Stellingen behorende bij het proefschrift
"High- T_c superconductors and magnetic electron lenses"
van J.P. Adriaanse

- 1) Het verbeteren van een magnetische objectieflens van een elektronenmicroscop, met als doel de interpretatie van het beeld te vergemakkelijken, kan het best worden bewerkstelligd door verdere ontwikkeling van spoelen met een hoge stroomdichtheid.
- 2) Het toepassen van hoge- T_c supergeleidende dunne films voor magnetische lenzen, zoals gebruikt in de elektronenmicroscop of de elektronenlithograaf, wordt aantrekkelijk indien het produkt van de maximale epitaxiale filmdikte en de kritische stroomdichtheid een factor vijf is toegenomen.
- 3) Dunne film lenzen, zoals beschreven in dit werk, lenen zich bij uitstek voor numerieke optimalisatie van hun optische eigenschappen.
- 4) Bij het optimaliseren van een elektrostatische lens met betrekking tot een specifieke toepassing is het bepalen van de optimale grootte in het algemeen veel belangrijker dan het bepalen van de optimale vorm.
- 5) Een elektronoptisch ontwerp is pas volledig indien er naast informatie over de optische kwaliteit ook gegevens beschikbaar zijn over de mate waarin deze kwaliteit door mechanische onnauwkeurigheden zal worden beïnvloed.
- 6) Naast het uitvoeren van in-situ experimenten in de elektronenmicroscop is het, middels een schaalverkleining van dit instrument, zinvol te trachten in-situ microscopie te bedrijven tijdens een experiment.
- 7) Bij het verhogen van de bundelenergie in een schaduwmaskerbeeldbuis, teneinde via een kleinere elektronenspot tot een verhoogde beeldscherpte te komen, gaat men vaak voorbij aan het feit dat het hierdoor veroorzaakte contrastbederf het beoogde effect teniet kan doen.
- 8) Daar onze samenleving gebaseerd is op onderlinge competitie op alle niveaus is het een illusie te denken dat er, door invoering van technieken die taken van de mens overnemen, meer vrije tijd zal ontstaan.
- 9) Indien de kritische temperatuur van supergeleiders de kamertemperatuur gaat benaderen, verdient het aanbeveling de definitie van het begrip "normal state" te herzien.
- 10) Een succesvolle wetenschapper wordt gekenmerkt door de eigenschap een eigenwijze bewering te kunnen uitwerken tot een eigen wijze.
- 11) Alle stellingen behorende bij dit proefschrift zijn verdedigbaar, zelfs deze.

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HIGH- T_c SUPERCONDUCTORS
AND
MAGNETIC ELECTRON LENSES

HIGH- T_c SUPERCONDUCTORS AND MAGNETIC ELECTRON LENSES

Proefschrift ter verkrijging van de graad van doctor aan de
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Prof.drs. P.A. Schenck, in het openbaar te verdedigen ten overstaan van een
commissie aangewezen door het College van Dekanen
op vrijdag 2 oktober 1992 te 19.00 uur

door

Jan-Peter Adriaanse,

geboren te Zierikzee,
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en

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Aan mijn ouders

Aan Esther

Contents

Introduction	1
1 Introduction into superconductivity	3
1.1 General properties	3
1.2 Various theories	4
1.2.1 London theory	4
1.2.2 Ginzburg Landau theory	7
1.2.3 microscopic theory	9
1.3 Magnetic flux structures	12
References	15
2 The utilization of classical superconductors in electron microscopy	17
2.1 Introduction	17
2.2 Superconducting lenses	19
2.2.1 solenoid lenses	19
2.2.2 trapped flux lenses	20
2.2.3 diamagnetic shielding lenses	21
2.2.4 shrouded coils and polepiece lenses	22
2.3 Microscopes equipped with superconducting lenses	25
2.4 Conclusions	26
References	28
3 High temperature superconductivity	31
3.1 High temperature superconducting materials	31
3.1.1 General structure	32
3.1.2 $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$	33
3.2 High temperature superconductor properties	35
3.2.1 Layered structure and anisotropy	35
3.2.2 Substitution of elements	35
3.2.3 Critical field and critical current density	36

3.2.4 Comparison of the critical current density in bulk-, thin film- and multilayered superconductors	43
3.2.5 Magnetic shielding	45
3.3 Possible mechanisms for electron pairing in high- T_c superconductors	45
3.4 Device applications	46
References	48
4 Thin film fabrication	55
4.1 Introduction	55
4.2 Sputtering	58
4.3 Laser Ablation	60
4.4 Evaporation	62
4.4.1 introduction	62
4.4.2 fabrication equipment	63
4.4.3 fabrication procedure	65
4.4.4 Thin film analysis	67
4.5 Other Techniques	70
References	71
5 Experiments with a thin film coil	75
5.1 Patterning	75
5.2 Measurements	76
5.2.1 superconducting properties	76
5.2.2 field distribution	78
5.3 Conclusions	79
References	80
6 Thin helical lenses and thin film lens elements	81
6.1 Introduction to thin helical lenses	81
6.1.1 focal properties	81
6.1.2 spherical aberration	86

6.2 Introduction to thin film lens elements	90
6.2.1 the lens element concept	90
6.2.2 various types of lens elements	93
6.3 The magnetic field of a lens element	98
6.3.1 the magnetic field using Biot Savart	98
6.3.2 unified representation for the magnetic field of a one turn spiral using a multipole expansion	100
6.3.3 the field of a lens element using a multipole expansion	105
References	106
 7 Design of a thin film stack pattern	 109
7.1 Introduction	109
7.2 Capacitive alignment	110
7.2.1 basic principles	110
7.2.2 a differential capacitive position sensor	112
7.3 Geometry of the stack element	113
7.3.1 introduction	113
7.3.2 helical pattern	113
7.3.3 capacitive electrodes	114
7.3.4 connection between adjacent layers	118
7.4 Optical properties	121
7.4.1 introduction	121
7.4.2 field distribution	123
7.4.3 first order properties	125
7.4.4 spherical- and chromatic aberration	126
7.4.5 parasitic aberrations	127
References	131
 8 Experimental work on a prototype lens element	 133
8.1 Fabrication of the lens element	133
8.2 The capacitive alignment procedure in practice	138
8.3 The prototype lens element using $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ thin films	142
References	143

9 Conclusions and future perspectives of high- T_c superconductors in particle optics . 145

9.1 The present state of high- T_c superconductors with respect to applications in particle optics 145

9.2 Potential applications 148

References 152

Summary 155

Samenvatting 159

Acknowledgement 163

Curriculum Vitae 165

Appendix A: Recipe for fabrication of a stack element. 166

Introduction

To investigate the applicability of superconductors seems a logical step if one wants to make iron-free magnetic lenses or wants to reduce the dimensions of a conventional lens, since, when decreasing the coil size, the current density is increased.

In the nineteen sixties and seventies, research, mainly concentrating on the application of superconductors for magnetic electron lenses took place on a relatively large scale. This is reviewed in chapter 2, prefaced, in chapter 1, by a general introduction into superconductivity. Superconducting lenses and microscopes did not grow into a success due to their inconvenient operation and the lacking popularity of high voltage electron microscopy. High voltage microscopy has been one of the main reasons to work on strong magnetic lenses and, consequently, on the utilization of superconductors.

In 1986, with the discovery of high temperature superconductivity, the discussion on the applicability of superconductors for magnetic electron lenses was re-opened. In the past, one of the most serious disadvantages concerning the operation of superconducting lenses had been related with the use of liquid helium refrigeration, so high- T_c superconductors might be employed to overcome this problem, because their cooling demands are much more relaxed. However, especially during the first years of high- T_c superconductivity, despite their high operating temperature, the materials themselves seemed extremely unfriendly, as they were brittle, sensitive to water, instable and difficult to produce. Fortunately, most of these disadvantages have disappeared now and the discussion concerning their utilization got a more fundamental character in the sense that most properties of the high- T_c materials are known, though a sound theoretical basis has not been defined yet. The properties of high- T_c superconductors are subject of chapter 3.

To investigate if high- T_c superconductors are more appropriate for applications in particle optics than their classical counterparts was considered to be an interesting research subject. Therefore, this work was started as a feasibility study to the use of high temperature superconductors in particle optics. Most short term applications in this field were expected to employ the high current density of these materials at temperatures

INTRODUCTION

above liquid helium, so this work concentrated on magnetic lenses as one of the most straightforward high current density applications.

Since conventional iron circuit lenses are already used to their limits, set by the saturation of the ferromagnetic circuit, significant improvements are only to be expected from iron free lenses or highly saturated polepiece lenses. Their performance is restricted by the current density allowed in the windings and further, for the iron free lens, by the attainable mechanical tolerances, since, in the absence of iron, a lack of axial symmetry in the windings directly results in parasitic aberrations. For making small iron free lenses, high- T_c thin films are a potentially interesting candidate, as they possess a high current density and can be patterned very accurately using lithographic techniques. Advantages to be expected from thin film lenses are smaller dimension and better optical properties. An overview of fabrication techniques for high- T_c thin films is in chapter 4.

The first attempt to make a coil in a superconducting thin film, using co-evaporated $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films, is subject of chapter 5.

A thin film has to be patterned with some kind of spiral in order to obtain a coil. Therefore, thin film lenses do basically not possess full axial symmetry. The relation between the geometry of a flat coil and its optical properties is given in chapter 6.

The geometry of a feasible thin film lens element, along with its corresponding optical properties, is in chapter 7 and the technology used to fabricate this lens element is subject of chapter 8.

Finally, based upon the work presented in this thesis, chapter 9 treats the potential applicability of high- T_c superconductors in instruments that employ particle beams.

1 Introduction into superconductivity

In this chapter, the most important properties of what now are called classical superconductors, together with a short overview dealing with the most important theories will be outlined briefly. The sections dealing with properties are section 1.1, which explains some general properties and section 1.3 that explains the flux structures appearing in a type II superconductor exposed to an external field. Section 1.2 indicates the major aspects of the theories of London, Ginzburg and Landau and Bardeen, Cooper and Schrieffer which are necessary to understand the origin of several parameters used throughout the rest of this thesis.

1.1 General properties

Superconductivity was discovered in 1911 by Kamerlingh Onnes in Leiden [Kamerlingh Onnes 1911]. A few years earlier, in 1908, he succeeded for the first time in liquefying helium and this provided Onnes with a method for studying phenomena in the 1 to 14 K interval. One of the first experiments was on the electrical resistivity of metals and to his surprise, Onnes found that the resistivity of a Mercury sample at about 4 K abruptly fell to zero. This phenomenon was called superconductivity.

That a superconductor is quite more than just a perfect conductor was discovered by Meissner and Ochsenfeld in 1933 [Meissner 1933]. They found that, in the presence of a magnetic field, there is a difference in behavior between a perfect conductor and a superconductor.

When an external field is raised, surface currents are induced on both a perfect conductor and a superconductor. These currents screen the field in order to maintain zero field in the sample. So far, there is no difference in behaviour between a perfect conductor and a superconductor. However, when the external field exceeds a critical value H_c , the superconductor turns into the normal state and the field can penetrate.

If the samples are in an applied field above H_c , and this field is lowered, the perfect conductor will internally keep up the initial field, but the superconductor will completely

expel the field from its interior as soon as the external field is below H_c . So, in a superconductor the internal field is zero for $0 < H_{\text{applied}} < H_c$. The situation for both a perfect conductor and a superconductor in a decreasing field, initially above H_c , is shown in Figure 1.1. Besides this sudden disappearance of superconductivity when the strength of an external field is raised above a certain critical value, superconductivity also disappears when a current through the superconductor exceeds a critical value.

1.2 Various theories

1.2.1 London theory

For a perfect conductor, the following classical equation describes the motion of an electron:

$$m^* \frac{dv}{dt} = -eE, \quad (1.1)$$

where m^* is the effective mass, v the electron velocity and E the electrical field strength in the conductor. If we multiply both sides by $-en_s$, where n_s is the density of superconducting electrons we obtain

$$\frac{dj}{dt} = \frac{n_s e^2}{m^*} E, \quad (1.2)$$

where $\mathbf{j} = -en_s\mathbf{v}$ is the induced current.

When eq.(1.2) is substituted into Faraday's law of induction

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} , \quad (1.3)$$

we obtain the following relation between current density and magnetic field:

$$\frac{\partial}{\partial t} \left(\nabla \times \mathbf{j} + \frac{n_s e^2}{m^*} \mathbf{B} \right) = 0 . \quad (1.4)$$

Substituting Maxwells equation

$$\nabla \times \mathbf{B} = \mu_0 \mathbf{j} \quad (1.5)$$

in eq.(1.4) gives:

$$\frac{\partial}{\partial t} \left(\nabla^2 \mathbf{B} - \frac{\mu_0 n_s e^2}{m^*} \mathbf{B} \right) = 0 . \quad (1.6)$$

From eq.(1.6) it can be seen that any time independent solution for $\mathbf{B}$ will satisfy this equation. So, when $\mathbf{B}$ in the perfect conductor is initially zero, the field inside the material will remain zero when an external field is applied. Also, when a non zero external field is removed, currents induced on the surface of a perfect conductor will keep up the internal field at its initial value. This was in contradiction with the observed behaviour of a superconductor which permits no interior fields below its critical temperature. The London brothers [London 1935] suggested that the full set of solutions eq.(1.6) should be restricted to only those that obey

$$\nabla^2 \mathbf{B} - \frac{\mu_0 n_s e^2}{m^*} \mathbf{B} = 0 , \quad (1.7)$$

which is a differential equation in B . Solutions of eq.(1.7) are typically of the form $B=C \cdot \exp(-r/\lambda_L)$, an exponential decay of the field inside the superconductor. Here λ_L is known as the London penetration depth given by

$$\lambda_L = \sqrt{\frac{m^*}{\mu_0 n_s e^2}} . \quad (1.8)$$

The London equation eq.(1.7) correctly predicts the observed Meissner effect. It also has the consequence, because of the Maxwell equation eq.(1.5), that the currents induced in a superconductor show the same exponential decay with depth as the field does. Hence, the induced currents are restricted to the surface of the sample.

In the superconducting state, strong correlations between the superconducting electrons are expected. Let this range be denoted by ξ_0 . From the foregoing it was concluded that the superconducting order is setup over a range λ_L , so for the validity of the London equations it is necessary that $\lambda_L \gg \xi_0$.

Another early theory has been developed by Gorter and Casimir [Gorter 1934], called the *two fluid model*. It is based on thermodynamics and assumes the coexistence of a normal- and superconducting fluid in a superconductor. Assuming perfect diamagnetism, Gorter and Casimir obtained for the difference in free energy between the superconducting and normal state:

$$F_n(T) - F_s(T) = \frac{1}{2} \mu_0 H_c^2 , \quad (1.9)$$

where F_n and F_s are the free energies in the normal and superconducting state, respectively, and H_c is known as the thermodynamic critical field. This two fluid model along with the phenomenological electrodynamic theory of the London brothers provided an early insight into superconductivity.

1.2.2 Ginzburg Landau theory

The Ginzburg-Landau theory is a phenomenological theory based on the following three postulates:

- The superconducting order can be described by a parameter ψ which goes to zero at the transition temperature.
- The free energy depends on the superconducting order and may be expanded in powers of ψ .
- The coefficients of the expansion are regular functions of T .

With the help of these three postulates, the free energy may be written as [Landau 1937]

$$F = F_n + \alpha(T) |\psi|^2 + \frac{\beta(T)}{2} |\psi|^4 + \dots, \quad (1.10)$$

where F_n is the free energy in the normal state. When the order parameter has a spatial variation, then spatial derivatives must be added to eq.(1.10) which gives

$$F = F_n + \alpha(T) |\psi|^2 + \frac{\beta(T)}{2} |\psi|^4 + \gamma |\nabla \psi|^2 + \dots. \quad (1.11)$$

The total free energy now has to be obtained by performing the integration

$$f = \int_{\Omega} F d\mathbf{r}, \quad (1.12)$$

which runs over the volume Ω .

Ginzburg and Landau [Ginzburg 1950] proposed an extension of eq.(1.11) to describe the superconductor in the presence of a magnetic field. They considered the order parameter as some kind of wave function for a particle of charge e^* and mass m^* and postulated Gauge invariance in the sense of quantum mechanics. This gives

$$F = F_n + \alpha(T) |\psi|^2 + \frac{\beta(T)}{2} |\psi|^4 + \frac{1}{2m^*} \left| (-i\hbar \nabla - e^* \mathbf{A}) \psi \right|^2 + \frac{1}{2} \mu_0 h^2, \quad (1.13)$$

where A is the vector potential and $\mu_0 h^2/2$ the magnetic energy density. It must be emphasized that h is the internal field.

Minimizing eq.(1.12) with respect to the order parameter Ψ and F given by eq.(1.13), yields the two Ginzburg-Landau equations:

$$\frac{1}{2m^*} (-i\hbar\nabla - e^*A)^2 \Psi + \alpha\Psi + \beta|\Psi|^2\Psi = 0, \quad (1.14)$$

$$j = \frac{e^*i\hbar}{2m^*} (\Psi^*\nabla\Psi - \Psi\nabla\Psi^*) - \frac{e^{*2}}{m^*} \Psi^*\Psi A. \quad (1.15)$$

Later e^* and m^* could be derived from microscopic theory (see section 1.2.3) and their correct values proved to be $e^* = 2e$ and $m^* = 2m$, two times the electron charge and -mass respectively.

In principle, the Ginzburg-Landau equations allow the order parameter, the field and hence the currents to be calculated. However, because the relations are obtained from a series expansion in Ψ , the following restrictions to the valid temperature interval apply:

If $\lambda_L(0) \ll \xi_0$ then:

$$\frac{T_c - T}{T_c} \ll \left(\frac{\lambda_L(0)}{\xi_0} \right)^2 \quad (1.16)$$

and the superconductor belongs to type I. In eq.(1.16), $\lambda_L(0)$ denotes the London penetration depth at zero temperature and ξ_0 denotes the intrinsic coherence length.

If $\lambda_L(0) \gg \xi_0$ then:

$$\frac{T_c - T}{T_c} \ll 1 \quad (1.17)$$

and the superconductor belongs to type II. The temperature interval where the Ginzburg-Landau theory is valid is, in both cases, close to T_c , but for type II superconductors the validity holds for a larger temperature range.

Whether a superconductor is type I or type II thus depends on the ratio $\lambda_L(0)/\xi_0$. When $\lambda_L(0)/\xi_0 < 1$ the superconductor is type I and for $\lambda_L(0)/\xi_0 > 1$ we have type II material. A striking difference in behaviour between the two types of superconductors occurs when a magnetic field is applied. A type I superconductor completely expels the external field as long as the magnitude is below the critical field H_c . An external field above H_c turns the type I superconductor in the normal state. This critical field H_c of a type I superconductor equals the thermodynamic critical field discussed in

section 1.2.1, eq.(1.9). A type II material shows the same behaviour for small fields, but when the magnitude exceeds a certain limit, known as the lower critical field H_{c1} , the Meissner effect becomes incomplete and flux penetrates the superconductor in small filaments of core radius $\xi(T)$. Schematic graphs of the magnetization of the sample and the internal field as a function of the applied field are given in Figure 1.2, both for type I and type II material.

1.2.3 microscopic theory

An important discovery for the development of a microscopic theory was that in the superconducting state, there seemed to exist a gap in the electron excitation spectrum. This gap was a strong evidence that the electrons are in some bound state. Another

important discovery [Maxwell 1950] was the so called isotope effect which in its simplest form gives

$$T_c \propto \frac{1}{\sqrt{M}}, \quad (1.18)$$

where T_c is the transition temperature and M is the ionic mass. This was an indication that the lattice plays an essential role in the formation of the superconducting state.

The concept of an effective electron-electron interaction caused by the dynamics of the system was worked out. It was considered that a lattice deformation caused by the presence of an electron takes a finite time to relax and can therefore influence a second electron passing by at a later time. This leads to an effective electron-electron interaction potential of the form

$$v(q, \omega) = \frac{4\pi e^2}{q^2 + k_0^2} \left(1 + \frac{\omega_q^2}{\omega^2 - \omega_q^2} \right), \quad (1.19)$$

where e is the electronic charge, q is the difference in wave vector between the two electrons and $\hbar\omega$ is their difference in energy. Furthermore, ω_q is the frequency of a phonon of wave vector q and $1/k_0$ is the Thomas-Fermi screening length which brings the screening of the bare electron-electron interaction by the rest of the electrons into account. The first term of eq.(1.19) represents the usual screened electron potential and the second term the phonon-mediated interaction which may dominate at low frequencies. For $\omega < \omega_q$, $v(q, \omega)$ is negative, hence the phonon-mediated electron-electron interaction is attractive for $\omega < \omega_q$. In other words, electrons which differ in energy by less than $\hbar\omega_q$ effectively attract each other. A typical value for $\hbar\omega_q$ is $\hbar\omega_D$, the energy as given by the Debye frequency.

An attractive electron-electron interaction, however, does not automatically imply a bound state, because the attractive interaction might simply be too weak. The crucial point Cooper [Cooper 1956] emphasized was, that in the presence of a Fermi sphere of normal electrons, a net attractive interaction always leads to a bound state, regardless

the strength of the attractive potential. The effect of the presence of the Fermi sphere is that it limits the number of possible electron levels by excluding those with a wave vector less than k_F .

When the concept of a phonon-mediated interaction in the presence of a Fermi sphere of normal electrons is worked out, it leads to a bound state separated by a gap

$$\Delta \approx 2\hbar\omega_D e^{-\frac{1}{V_0 N_F}} \quad (1.20)$$

from the excited states, where ω_D is the Debye frequency, V_0 is a simplified form of eq.(1.19) and N_F is the density of states at the Fermi level.

If the formation of a bound pair of electrons lowers the energy, many pairs of electrons can be expected to form, and it was the theory developed by Bardeen, Cooper and Schrieffer (BCS) [Bardeen 1957] which provided an explicit N-particle wave function for the superconducting ground state. This ground state can be written as

$$|\Psi_0(N)\rangle = \left(\sum_k \phi(k) a_{k\uparrow}^+ a_{-k\downarrow}^+ \right)^{N/2} |\Phi_0\rangle, \quad (1.21)$$

where $a_{k\uparrow}^+$ creates an electron with momentum $\hbar k$ and spin up, $a_{-k\downarrow}^+$ creates an electron with momentum $-\hbar k$ and spin down and

$$\sum_k \phi(k) a_{k\uparrow}^+ a_{-k\downarrow}^+ \quad (1.22)$$

is the Fourier transform of a Cooper pair. This ground state does not violate the Pauli exclusion principle since it is made up from two electron states.

The description of all superconducting electrons by just one N-particle wave function has far-reaching consequences. Since the electrons are highly correlated, the scattering of a Cooper pair involves disturbing many more electrons than just the two which form the pair, so that effectively scattering a Cooper pair implies a transition of the N-

electron state which is extremely unlikely. Hence, the Cooper pair can move through the system without hindrance.

With knowledge of the BCS theory the concept of the coherence length ξ , introduced in section 1.2.2 can be understood in a less abstract way, namely as the effective size of a Cooper pair. In classical pure superconductors, ξ_0 is typically a few hundred nanometers. The coherence length also depends on the mean free path l of the electrons in the normal state. Therefore, the effective coherence length ξ is given by

$$\frac{1}{\xi} = \frac{1}{\xi_0} + \frac{1}{l}, \quad (1.23)$$

where ξ_0 is the intrinsic coherence length. In pure systems, where l is large, $\xi \approx \xi_0$, while in so called dirty superconductors where the mean free path is short, $\xi < \xi_0$. Dirty superconductors are often alloys.

1.3 Magnetic flux structures

As already explained in section 1.2.2, at an external field of sufficient strength, the difference between type I and type II superconductors appears. A type I material transfers to the normal state, or, when the sample is suitably shaped, transfers to the intermediate state where the volume of the sample is subdivided into normal and superconducting regions. Those normal regions in a type I material are often referred to as flux-tubes. Type II materials, on the other hand, show a completely different behaviour for fields above H_{c1} , the so called lower critical field. Those materials transfer to the vortex state as was briefly outlined in section 1.2.2. Whether a material is type I or type II depends on the ration λ/ξ , known as the Ginzburg-Landau parameter κ .

The difference in behaviour between type I and type II materials can be understood when an energy is attached to the superconductor/normal interface. If this concept of surface energy is worked out within the theoretical frame of the Ginzburg-Landau equations, the following results appear. For $\kappa < 1/\sqrt{2}$ the surface energy is positive, so in

general the superconductor/normal interface will tend to be as small as possible, thus allowing no flux penetration. For $\kappa > 1/\sqrt{2}$ a negative surface energy comes out and this leads to the concept of normal filaments immersed in a superconducting environment. The structure of such a normal filament, mostly called vortex, can best be explained with the help of Figure 1.3. In the core of the vortex of radius ξ , the superconducting order parameter is depressed. Outside the core region, the field in the superconductor decreases over the characteristic length λ , and in order to shield the rest of the superconductor from the field, supercurrents surround the core. This supercurrent distribution is schematically given in Figure 1.3c.

The total energy per unit vortex length, made up from the magnetic field energy and the kinetic energy of the superconducting vortex currents, can be approximated by

$$E_l \approx \left(\frac{\Phi_0}{4\pi\lambda} \right)^2 \ln \left(\frac{\lambda}{\xi} \right), \quad (1.24)$$

where Φ_0 is the elementary flux quantum. From eq.(1.24) it can immediately be seen that because of the quadratic dependence of E_l on Φ_0 , it is most favorable to have only one flux quantum per flux line. Another important conclusion that can be drawn from eq.(1.24) is that the interaction between individual vortices is repulsive, again because a clustering of vortices would increase the energy quadratically. For high enough fields, the vortices become numerous and

if not disturbed by other forces, they form a regular triangular vortex lattice. This vortex

lattice shows a certain similarity to a crystal lattice in that departures from the ideal triangular lattice structure may occur in the form of deformations of the triangular structure and various lattice defects. This mostly results from the interaction between the vortex lattice and the crystal lattice.

If an electrical current is imposed on a superconductor in the vortex state, the vortices will experience a Lorentz force which tends to move them in a direction perpendicular to the current. This flux line motion induces a voltage V which according to Faraday's law is

$$E = -\nabla V = -\mathbf{v}_\phi \times \mathbf{B} . \quad (1.25)$$

In eq.(1.25) $\mathbf{v}_\phi$ is the flux flow velocity. This mechanism of a current induced voltage is mostly referred to as flux flow resistance. Besides flux flow induced by a current, the vortices may also be moved by a temperature gradient across the superconductor. Figure 1.4 shows a schematic diagram of the flux flow resistivity in a type II superconductor versus the magnetic field for different temperatures.

Flux flow can be suppressed if the vortex lattice is sufficiently anchored. This anchoring of the vortex lattice, known as flux pinning, is very important in technical applications where high fields and/or large transport currents are involved. The origin of the flux pinning are regions in the crystal lattice which in some way show different behaviour. A rough division between δT_c and $\delta \kappa$ pinning centers can be made [Kes 1990]; points where respectively the critical temperature or the Ginzburg-Landau parameter are different. To the first group belong precipitates and dislocations, $\delta \kappa$ pinning is mostly caused by grain boundaries, voids, vacancies and interstitials.

In this paragraph, we assumed both an external field and a transport current, but the theory in case of a transport current only is similar, as a current is always accompanied

by a magnetic field. A very important point with respect to applications is that in type II superconductors, the critical current is determined by the force exerted by defects on the vortices [Thuneberg 1989] and the larger these forces can be made, for example by artificially adding pinning centers, the more interesting the superconductor will be for high current applications.

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2 The utilization of classical superconductors in electron microscopy

In this chapter, we will outline the utilization of classical superconductors for electron lenses and electron microscopes which will serve as a starting point to investigate the applicability of high- T_c superconductors in later chapters. We first give some general information in section 2.1, followed by a systematic overview of the most important superconducting lens types in section 2.2. Section 2.3 deals with microscopes that employ superconducting lenses and finally we summarize, in section 2.4, the advantages and disadvantages that go with the use of classical superconductors in electron microscopes. For a more complete overview of the various superconducting lenses and microscopes, see e.g. Hawkes [1977] or Hardy [1973].

2.1 Introduction

Superconductors drew the attention of the workers in the field of particle optics when superconducting alloys such as Nb_3Sn and $NbTi$ became available. These alloys exhibit a high critical current density in a large external field, which is of course most important when aiming for small coils driven at a high current density. Approximately in 1965, when the first image produced by a superconducting coil had been realized [Fernández-Morán 1965], the major attention for superconductors in particle optics started. The interest in this field of research peaked in the mid-seventies and slowly faded away in the early eighties when it was generally realized that classical superconductors could not significantly improve the performance of an electron microscope [Lefranc 1982]. The marginal improvements that had been obtained were accompanied by complicated operating conditions caused by the presence of the helium cryostat. Today, the only established advantages of superconductors are not related to better optical properties, but deal with the cryo-protection of radiation sensitive samples and improved vacuum conditions [Iwatsuki 1988]. Those advantages are all due to the

liquid helium temperature in the sample region and not directly caused by the use of superconductivity.

The trends in electron microscopy in the early seventies were towards high resolution, observation of thick specimens, in-situ experiments and microanalysis [Hawkes 1977]. The observation of thick specimens requires a high beam voltage, because some extra matter has to be penetrated. This requires a high magnetic field and in order to prevent excessive coil dimensions, the use of superconductors seems obvious because they allow much higher current densities. The current density in conventional water cooled lenses does usually not exceed 100-300 A/cm². It can be raised to 10⁴ A/cm² in special designs [Podbrdský 1986]. Nonetheless, more than one extra order of magnitude in current density can be gained by application of a classical superconductor.

The advantages related to a higher beam voltage are [Dietrich 1975b]:

- greater penetration power for thick specimens
- improvements in the resolving power, due to the smaller wavelength
- a reduction in chromatic aberration, due to the smaller relative energy spread

Disadvantages are:

- radiation damage in the sample, which is therefore not stable during observation
- mechanical vibrations that are due to the much larger dimensions of the microscope

Especially the reduction of chromatic aberration may result in a significantly better resolution if thick specimens are to be observed, because a relatively large number of electrons are inelastically scattered in this case and the resolution is then determined by the chromatic aberration of the objective lens. This resolution as limited by chromatic aberration reads [Hardy 1973]

$$\delta = k C_c^{1/2} \left(\frac{\Delta V}{V} \right)^{1/2} \lambda^{1/2}, \quad (2.1)$$

where k is a constant close to unity, C_c is the coefficient of chromatic aberration, $\Delta V/V$ is the relative energy spread and λ is the electron wavelength. Eq.(2.1) shows that the effect of a better C_c on the resolution limited by chromatic aberration is larger than the

effect that a better C_s would have, in case the resolution is limited by spherical aberration, since the latter is proportional to $C_s^{1/4}$.

For high voltage applications, superconductors offer some advantages, such as a compact construction and a high magnetic field gradient in the lens gap. Especially at beam energies over 1 MeV, the optical properties of superconducting lenses are more favorable than those of conventional lenses [Dietrich 1975b], and the much higher current densities result in a size reduction. At smaller energies, the dimensions of the cryostat tend to cancel the reduction of the lens dimensions [Hardy 1973]. Other advantages related to the use of superconducting lenses are a reduction in power consumption and a high stability of the lens current if operated in the persistent mode [Hawkes 1977]. Advantages offered at medium voltage are again the high stability of the current in the persistent mode and the possibilities for ambient field shielding [Dietrich 1975a].

Concomitant advantages of superconducting lenses, not directly related with optical properties, are cooling of the sample and an improved vacuum due to the cryogenic pumping of the helium cryostat. A well known method in conventional electron microscopy for taking high resolution images of radiation sensitive samples is to use a helium refrigerated sample stage. However, contamination, specimen drift and bubbling of the coolant are complicating factors which are nearly absent when a superconducting objective lens is used [Iwatsuki 1988].

2.2 Superconducting lenses

2.2.1 solenoid lenses

By using superconducting materials that exhibit a high current density, it is possible to produce a high peak field with a solenoid only, because it is possible to scale down the coil dimensions [Dietrich 1975a]. Open coils are relatively easily made, but it proved to be very difficult to achieve the necessary rotational symmetry in those small coils. As a

result, those lenses suffered from excessive astigmatism [Hawkes 1977].

An interesting remark is that pancake coils as proposed by Mulvey (see chapter 6) show a much better performance if constructed with superconductors [Génotel 1967]. Instead of using only one pancake coil it is also possible to use a system of coils which has the advantage that the peak of the field can be shifted with respect to the main coil, thereby creating more room around the specimen [Merli 1970]. This principle is shown in Figure 2.1.

2.2.2 trapped flux lenses

Another relatively simple type of superconducting lens is the ring lens, also known as trapped flux lens. Trapped flux lenses consist of a superconducting ring or a stack of superconducting disks inside a magnetizing device such as a polepiece lens, or a superconducting coil. Flux is trapped in the ring by heating it above its critical temperature in the presence of the external field. After switching off the heater, the external field is slowly decreased towards zero which results in a field trapped in the ring, because the superconductor will try to keep up the initial field [Hardy 1973]. The principle

of a trapped flux lens is shown in Figure 2.2. Different types are rings, flat annuli or tubes [Hawkes 1977].

Focusing with a trapped flux lens is a problem because, once trapped, it is difficult to change the amount of flux in the ring. One method for focusing is to change the beam energy, but this requires a very careful alignment of the microscope that is not destroyed by small changes in beam energy. Another option is to use an additional focusing coil [Hardy 1973] which only needs a moderate excitation and thus has relaxed requirements on its rotational symmetry and current stability.

The advantage of the trapped flux lens over the solenoid is a better rotational symmetry. However, a non-homogeneous distribution of pinning centers in the superconductor may cause astigmatism, even if the ring is fully rotational symmetric in a mechanical sense. In order to diminish those non-rotational symmetry effects, a stack of disks may be used. The lens can be improved further by using disks which carry superconducting concentric annuli [Hawkes 1977]. A practical example of such a lens was published by Hibino et. al. [1973]. They used a stack of twenty disks, each disk carrying a 13 μm Nb₃Sn layer on both sides. The inner and outer diameters of the disks are 3 mm and 12.7 mm respectively. An excitation coil of Nb-Ti wire was used to produce the external field required for flux trapping. The field was trapped using the method outlined in the first paragraph of this section. The average current density in the Nb₃Sn was approximately $4 \cdot 10^5$ A/cm², resulting in a maximum axial flux density of 2.2 T. The distribution of the trapped field was close to Gaussian.

2.2.3 diamagnetic shielding lenses

Diamagnetic shielding lenses are lenses that explore the perfect diamagnetism of superconductors below the lower critical field (H_{c1}). Those lenses employ shielding cylinders for shaping the field and an outer casing to prevent flux leakage. An advantage of those lenses is the high maximum axial flux density which is allowed, because in this case, not the saturation of the iron, but the critical field of the superconductor is the limiting factor [Lefranc 1982]. Axial flux densities as high as 7 T can be obtained

[Hawkes 1977]. The principle of the diamagnetic shielding lens is shown in Figure 2.3.

A remarkable difference between iron circuit lenses and shielding lenses is that for the latter, the flux density in the gap decreases if the shielding cylinders are brought closer together, whereas the flux density in the gap of an iron circuit lens increases if the polepieces are moved towards each other. It is nevertheless possible to obtain very high gradients in

Figure 2.3 Principle of the diamagnetic shielding lens [Hardy 1973].

a field distribution shaped with a diamagnetic circuit. To achieve such a narrow field distribution with a high peak value, the cylinders must be close together and the field inside the diamagnetic casing has to be very intense [Hardy 1973].

Rotational symmetry defects in shielding lenses can be corrected by the application of stigmators and deflectors which can be placed in the lens gap (see Figure 2.3). Hence, the symmetry faults are corrected in the origin and thus do not give rise to magnification errors.

Dietrich et. al. [1975a] experimented extensively with shielding lenses and obtained the best results with Nb-Sn sinter material with some Cu additive. The active shielding material was Nb₃Sn which was formed during annealing at 700° C.

2.2.4 shrouded coils and polepiece lenses

Iron-shrouded solenoids can be considered as hybrid lenses in a sense that the axial flux density arises both from the solenoid itself as well as from the magnetization of the iron circuit. The principle of an iron shrouded solenoid is shown in Figure 2.4. The aim of this type of lens is to increase the peak of the field distribution and cut off the tails.

Mechanical astigmatism is still severe in an iron shrouded solenoid, due to the saturation of the iron and because the smoothing effect of the iron is least effective for the windings close to the axis which contribute most to the astigmatism [Hardy 1973].

The superconducting lens which best resembles the conventional polepiece lens is the shrouded coil with polepieces schematically shown in Figure 2.5. For superconducting lenses which employ polepieces there are two versions. One with cold polepieces made from rare earth materials that have a high saturation at liquid nitrogen or liquid helium temperature and one with room temperature polepieces [Lefranc 1982].

The latter do not provide the cryo-protection and cryogenic pumping which is considered to be an important additional advantage of a superconducting objective lens.

Table 2.1 Curie temperature and saturation magnetization of rare earth- and conventional polepiece materials.

Material	T_c [K]	B_s [T]	μ_r @ 1 T
Holmium	20	3.35	7
Dysprosium	85	2.45	3
Cobalt-iron	1220	2.35	$3 \cdot 10^3$
Iron	1040	1.6	$2 \cdot 10^4$

Examples of materials having a high saturation magnetization at low temperatures are Holmium and Dysprosium which are compared to conventional materials in Table 2.1 and Figure 2.6. A disadvantage of those rare earth polepiece materials is their low

Figure 2.4 Principle of an iron shrouded solenoid [Hardy 1973].

Figure 2.5 Principle of a superconducting polepiece lens [Hawkes 1977].

relative permeability. The relative permeability of Dysprosium is only three at three Tesla, so a large field strength has to be applied to achieve the required flux density between the polepieces. Holmium is a little better, but has a Curie temperature of 20 K and therefore has to be cooled with helium. The optical properties of lenses employing rare earth polepieces are mostly not much better than the properties of superconducting lenses equipped with iron polepieces [Hawkes 1977]. Furthermore, the rare earth polepiece materials are anisotropic and thus need to be oriented carefully prior to machining, in order to prevent a large basic astigmatism in the polepieces.

Iron circuit lenses with polepieces are primarily used in the saturated mode which results in a field distribution with long tails. The effect of astigmatism is however much smaller than in shrouded solenoids, because the iron, though saturated, also smooths the symmetry errors of the windings close to the axis.

Already mentioned advantageous side-effects related to a lens operated at liquid helium temperature are cryogenic pumping and radiation protection of the sample.

Another related advantage is the large thermal stability which results from the fact that the complete lens is inside a cryostat at liquid helium temperature.

2.3 Microscopes equipped with superconducting lenses

In most microscopes employing superconducting lenses, the only superconducting lens is the objective. One of the exceptions is the Siemens microscope as proposed by Dietrich et. al. [1975b], which fully utilizes superconductivity. The microscope is based on an objective lens of the diamagnetic shielding type. There are three different cryostats: a condenser cryostat, an objective cryostat and a projector cryostat which house a total of eight superconducting lenses, the objective included. The three condenser and three projector lenses, as well as the intermediate lens behind the objective, are of the superconducting polepiece type.

The Siemens microscope operates with a pulsed beam that has to be formed with a field emission source and a buncher. Acceleration to 3 MeV should be done with a superconducting microwave linear accelerator. The vacuum at the sample position is better than 10^{-9} Torr and the theoretical resolution is $0.7 \text{ \AA}$ with a C_s of 3.5 mm at a beam energy of 3 MeV.

Some principles of this microscope were experimentally verified in a test setup at 200 kV, using a normal electrostatic accelerator [Dietrich 1977]. The diamagnetic shielding lens functioned properly with a basic astigmatism comparable to that of a conventional lens. Moreover, all superconducting lenses as well as the deflector and stigmator coils were successfully operated in the persistent mode. The resolution obtained with this microscope was $1.7 \text{ \AA}$, close to the theoretical limit for this instrument which is $1.5 \text{ \AA}$. The effective current density in the coil of the shielding lens was $1.5 \cdot 10^4 \text{ A/cm}^2$ at a beam energy of 150 keV. The fact that it took two hours to cool down the cryostat and align the system, indicates the amount of extra effort needed to operate a microscope equipped with superconducting lenses.

Cryostats applied for the cooling of superconducting electron lenses have to be designed very carefully in order to eliminate vibrations. According to Lefranc [1982], one of the principle requirements to achieve this is to consider the cryostat and the lenses as one unit. For the same reason, only bath cryostats were used instead of evaporator cryostats, despite the more favorable dimensions of the latter. The most important requirements for a cryostat housing superconducting lenses are [Dietrich 1975a]:

- 1) good mechanical stability
- 2) low helium evaporation rate to avoid strong bubbling
- 3) rotational symmetric construction to avoid displacements during cooling
- 4) materials with comparable expansion coefficients, especially near the beam
- 5) no materials which become ferromagnetic at low temperatures
- 6) small apertures between the cold and warm microscope parts to avoid contamination in the sample region due to cryopumping
- 7) feed throughs constructed in such a way that no vacuum grease or other contamination can be transferred to the specimen region

2.4 Conclusions

The major effort concerning the application of superconductors in electron optics was mainly directed towards improvements of the objective lens, though there have been some other projects. One of these is dealing with the correction of spherical aberration in the objective lens of an electron microscope by the application of superconducting zone plates [Dekkers 1969]. The advantage of an active zone plate, which carries a current, is that the wavefront can be modified in such a way that positive phase contrast can be obtained for a large range of spatial frequencies.

Another application which deviated from the general interest was the use of superconducting cavities for the acceleration of an electron beam. Here the advantages related to the use of superconductors must come from lower power dissipation and smaller dimensions. General disadvantages of high frequency acceleration are that the

system can only deliver electrons with a specific energy [Hawkes 1977], due to the fixed geometry and dimensions of the cavity.

If the objective lens of an electron microscope has to be replaced by its superconducting counterpart, the following considerations apply [Hardy 1973]:

- 1) Does the lens, in theory, have better optical characteristics than a conventional lens would in the same circumstances?
- 2) Is the promised improvement in the theoretical resolving power useful?
- 3) Can this theoretical resolving power be achieved regularly in practice?

Those three items are all aiming at a better resolution, but more general there may be additional considerations such as:

- a) Does the superconducting lens offer features which are infeasible in a conventional way?
- b) Is a significant reduction in size possible without deterioration of the optical properties?
- c) Can the overall power consumption of the system be reduced?

The original aim of superconducting lenses was to obtain high magnetic peak fields which were necessary in high voltage electron microscopes. Today, the superconducting lens is applied for research of organic materials, sensitive to radiation damage, which only requires the low temperature ambient of the sample [Iwatsuki 1988].

The problems that arose with superconducting lenses in microscopes mostly originated from the cryostat which is a source of mechanical instabilities, and it hard to estimate those in a quantitative way beforehand. Moreover, the presence of a cryostat makes the electron microscope an instrument for specialists only which has to be operated in a laboratory where liquid helium facilities are present.

Given the overall scale of the research in electron optical instrumentation, there has been a large effort towards the utilization of superconductors in electron microscopes. Nevertheless, this exploration has not been so extensive and well established as in other fields of research. One of the best known examples is high energy physics, where the introduction of superconducting magnets meant a significant step forward. Those magnets allowed field strengths that were completely infeasible with conventional magnets, or were only possible at an excessive level of power consumption. Compared

to those applications, the requirements for electron microscopy are much more relaxed and can very well be attained using conventional lenses, especially at beam energies below 1 MeV, which are far the most popular. In practice the performance of superconducting lenses has been only as good as with conventional lenses and the majority of the workers in the field of electron optics took the attitude that they would like to be sure of the advantages before they discarded the conventional iron circuit lens [Hardy 1973].

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3 High temperature superconductivity

In this chapter we introduce the high- T_c materials in section 3.1. This section deals with the crystal structure in general, as well as, more specific, with the $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ structure. The properties of those materials are summarized in section 3.2. Section 3.3 briefly indicates the applicability of the BCS theory. Finally, in section 3.4, we give an overview of possible device applications which may utilize high- T_c materials.

3.1 High temperature superconducting materials

Until 1986 superconductivity was only known at relatively low temperatures, with the so called A-15 materials having the highest critical temperatures. These A-15 materials are all metallic superconductors with Nb_3Ge having the highest T_c of 23 K. Also some superconducting oxides were known but did not have a high- T_c . These materials are listed in Table 3.1 [Yvon 1989]. In 1986 Bednorz and Müller announced superconductivity in $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ with a T_c of 35 K, the highest ever reported [Bednorz 1986]. Already a year later, Wu et. al. [1987] reported the first observation of superconductivity above liquid Nitrogen temperature (77 K) in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ ($T_c=92$ K). The 100 K limit was passed by the Tl containing compounds and went up to the present highest T_c of 125 K in $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$.

Table 3.1 Superconducting oxides with critical temperatures above 5 K known before 1986 [Yvon 1989].

material	T_c [K]
LiTiO_4	13
$\text{Ba}(\text{Pb}_{1-x}\text{Bi}_x)\text{O}_3$	13
$\text{Rb}_{0.3}\text{WO}_3$	6

3.1.1 General structure

So far, thirteen oxide structures favorable for high- T_c superconductivity are characterized [Yvon 1989], as listed in Table 3.2. All high- T_c oxides can formally be derived from the perovskite structure, shown in Figure 3.1, with general formula ABO_3 . However, the oxygen-to-metal ratio is always lower than, or equal to this ratio in the perovskite, which is $3/2$. For this reason, the crystal structure of the high- T_c oxides is often referred to as "defect perovskite". Figure 3.2 shows an example of how the structure of $YBa_2Cu_3O_{7-x}$ can be derived from that of perovskite.

All high- T_c oxides have orthorhombic or tetragonal symmetry, except for $Ba_{1-x}K_xBiO_3$ which is cubic. The most obvious common structural feature among the high temperature superconductors, again except for $Ba_{1-x}K_xBiO_3$, are the CuO_2 layers and the CuO chains. A good example is $YBa_2Cu_3O_{7-x}$ which has two CuO_2 planes and one CuO plane per unit cell, as indicated in Figure 3.2. In the CuO planes, the Cu - and O -atoms form chains along the b -axis.

Table 3.2 The thirteen prototypes of high- T_c oxides with their critical temperatures [Yvon 1989].

material	T_c [K]
$(Ba_{1-x},K_x)BiO_3$	30
$(La_{2-x},Sr_x)CuO_4$	40
$(Nd,Ce,Sr)_2CuO_4$	28
$(Nd_{2-x},Ce_x)CuO_4$	24
$YBa_2Cu_3O_7$	90
$YBa_2Cu_4O_8$	80
$Y_2Ba_4Cu_7O_{14}$	40
$Pb_2Sr_2NdCu_3O_8$	70
$TlBa_2CaCu_2O_7$	103
$TlBa_2Ca_2Cu_3O_9$	120
$Tl_2Ba_2CuO_6$	90
$Tl_2Ba_2CaCu_2O_8$	112
$Tl_2Ba_2Ca_2Cu_3O_{10}$	125

3.1.2 $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$

The family of high- T_c materials composed with the elements Y, Ba, Cu and O (YBCO) is described in more detail, because one of them ($\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$) has been used in this work. Up to now three members of the YBCO family are known, as listed in Table 3.3. $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$, known as "123", is the oldest member. The structure of "123" is given in Figure 3.2. This orthorhombic "123" structure is instable with respect to oxygen removal, temperature, pressure and irradiation [Yvon 1989]. When the oxygen content in "123" is not 7, the oxygen vacancies occur in the CuO chains which leads to a tetragonal phase if the oxygen content is reduced to 6.5. In the tetragonal phase the oxygen sites are only partially occupied. In general, the following rule for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ applies:

- orthorhombic and superconducting for $0 < x < 0.5$
- tetragonal and non-metallic for $0.5 < x < 1$

As nearly all high- T_c superconductors, "123" is a very anisotropic material having lattice parameters $a=3.8177 \text{ \AA}$, $b=3.8836 \text{ \AA}$ and $c=11.6827 \text{ \AA}$ [Yvon 1989]. The a-b orientation in "123"

Table 3.3 Chemical composition and critical temperature of the YBCO family.

composition	T_c [K]	stoichiometric
$\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$	92	no
$\text{YBa}_2\text{Cu}_4\text{O}_8$	80	yes
$\text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15-x}$	40	no

is only fixed in finite domains and changes when crossing a domain border. This phenomenon of alternating a-b orientation is known as "twinning".

Another YBCO member, $\text{YBa}_2\text{Cu}_4\text{O}_{8+x}$, mostly referred to as "248", is closely related to "123" from which it derives by intercalation of a second CuO layer. This results in a stacking sequence of the metal-oxygen layers as follows: $-\text{Y}-\text{CuO}_2-\text{BaO}-\text{CuO}-\text{CuO}-\text{BaO}-\text{CuO}_2-\text{Y}-$ etc. (see Figure 3.2 for comparison with "123"). The last YBCO member is $\text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15-x}$ ("247"). This structure consists of alternating "123" blocks with single chains and "248" blocks having double chains. An important point with respect to applications is that "248" is stoichiometric and hence has a high stability. Figure 3.3 shows the oxygen release of all YBCO structures as a function of temperature [Karpinski 1990] and it can be concluded that "123" already decomposes at $T > 400^\circ\text{C}$ where "248" needs $T > 850^\circ\text{C}$ in order to release oxygen. An advantage with respect to measurements of physical properties is in "248" and "247" which do not show the a-b twinning and thus allow orientation dependent measurements.

3.2 High temperature superconductor properties

3.2.1 Layered structure and anisotropy

Nearly all high- T_c superconducting oxides have a layered structure, CuO_2 planes and a tetragonal or orthorhombic symmetry. Most high- T_c crystal shows a large anisotropy which has its effect on the superconductor parameters. For example the coherence length of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ is 0.5 nm along the c-direction and 3 nm in the a-b plane [Lynn 1990]. In general the following relation holds [Friedel 1989]:

$$\frac{\lambda_c}{\lambda_{ab}} \approx \frac{\xi_{ab}}{\xi_c} \approx 5-10 . \quad (3.1)$$

Another property resulting from the anisotropy is that the charge carriers are much more mobile in the CuO_2 planes. This is probably due to the fact that ξ_c is comparable to the distance between the CuO_2 planes, which suggests that the high- T_c materials are not only highly anisotropic, but that they truly behave as a stack of two dimensional superconductors with weak interlayer coupling. More and more evidence for the essential role of the CuO_2 planes in the high- T_c superconductors is accumulated nowadays.

Typical values for λ and ξ are 100 nm and 1 nm, respectively, indicating a high κ , or, in other words, all high- T_c materials are strong type II superconductors. The fact that ξ is so small, less than the mean free path, makes the high- T_c oxides clean superconductors and it makes them also extremely sensitive to disorder [Kes 1990].

3.2.2 Substitution of elements

In order to gain knowledge about the new metal oxide materials, an interesting experiment is to replace one or more elements in a particular superconductor by similar

elements. The effect of this replacement might provide insight into the mechanism responsible for the superconductivity.

Complete replacement of Y in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ by another magnetic or non-magnetic rare earth element, like for example Ce, Pm or Tb does not have any influence on the critical temperature.

Replacement of Cu is more critical, as can be expected because of the essential role the CuO_2 planes seem to play. Cu can only partially be replaced by other metals like e.g. the magnetic Co, Fe, Ni or the non-magnetic Zn, Al or Ga without loss of superconductivity. However, the critical temperature is strongly affected upon replacement [Yvon 1989].

3.2.3 Critical field and critical current density

The critical temperatures for many high- T_c superconductors are listed in Table 3.2. For $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$, T_c strongly depends on the oxygen content. According to Wang [1990] three different regimes for T_c apply as listed in Table 3.4.

Because of the strong type II character of the high T_c materials, they are particularly susceptible to flux penetration. The lower critical field is of the order of 10 mT. For fields above the lower critical field (H_{c1}), flux can penetrate the superconductor in small filaments which form a so called flux line lattice. This flux line lattice (FLL) shows a special behaviour in the high- T_c materials. Because of the weak coupling between the planes, the flux lines perpendicular to the planes are very flexible, so the order of the FLL is destroyed even when the temperature is significantly below T_c , a phenomenon often referred to as "melting" of the FLL [Tinkham 1991]. In the "molten" state, the flux lines are free to move, thus causing resistance. The melting line of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ at fields of order 1 T is about 10 K below T_c , thus allowing its use at 77 K.

Table 3.4 The three regimes for T_c in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ for different oxygen content [Wang 1990].

oxygen defect [x]	T_c [K]
$0 < x < 0.2$	90
$0.2 < x < 0.5$	60
$x > 0.5$	not sc

Though the lower critical field is small, the upper critical field (H_{c2}) is much larger than for the classical materials. For high quality thin films, $H_{c2}(0)$ was estimated to be about 340 T for the field parallel to the ab-plane. Even at liquid Nitrogen temperature $H_{c2}(77) > 20$ T. Another estimate of $H_{c2}(0)$ for films made by chemical vapor deposition is 180 T [Watanabe 1989]. The variation of H_{c2} with temperature at $T=T_c$ is about 3 T/K for the field parallel to the ab-plane and 0.6 T/K for the field parallel to the c-axis. Figure 3.4 shows the influence of an external magnetic field on the R-T curve of an epitaxial $YBa_2Cu_3O_{7-x}$ film, and Figure 3.5 shows the temperature dependence of the upper critical field $B_{c2}(T)$. Both curves were measured by Adrian [1991].

Figure 3.4 Resistive transitions of an $YBa_2Cu_3O_{7-x}$ film in external magnetic fields up to 12 T [Adrian 1991].

The critical current density (J_c) is very high for single crystal materials. Values over 10^6 A/cm² have reproducibly been obtained in thin single crystal films. Several mechanisms can be responsible for the maximum current density [Tsuei 1989]. The most obvious one is when the energy associated with the current locally exceeds the gap

Figure 3.5 Temperature dependence of the upper critical field $B_{c2}(T)$ [Adrian 1991].

energy and the Cooper pairs dissociate. This J_c limiting mechanism is known as the depairing limit. The London theory provides a relation between the thermodynamic critical field and the maximum current density:

$$J_c(T) = \frac{B_c}{\mu_0 \lambda(T)} . \quad (3.2)$$

The depairing J_c can also be deduced from the Ginzburg-Landau theory or, at low temperatures, from the BCS theory. For the high- T_c materials, the depairing $J_c(0) \approx 10^8 - 10^9 \text{ A/cm}^2$ [Tsuei 1989, Gross 1990], and $J_c(77) \approx 10^7 - 10^8 \text{ A/cm}^2$. The depairing limit is the theoretical upper limit for the critical current density. Figure 3.6 shows the depairing limit as resulting from the various theories, as well as the critical current density of a thin $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film as a function of temperature.

Another mechanism limiting J_c is flux creep, thermally activated motion of flux lines. Flux creep occurs if the pinning energy is insufficient. The hopping rate of the flux lines is proportional to $\exp[-U_0(B,T)/k_B T]$, where U_0 is the pinning energy. Flux creep in high- T_c superconductors is large because of a small U_0 and a possibly high operating temperature.

The third mechanism which may limit J_c are weak links between adjacent single crystalline grains of superconducting material. These weak links are normal regions, thin enough for the superconducting electrons to tunnel through. However, above a certain current density normal dissipation occurs in those weak links. In bulk materials weak links are the main cause for a low current density, especially in the presence of external magnetic fields.

Because high- T_c superconductors may be operated at relatively high temperatures, the Boltzmann factor $\exp[-U/k_B T]$ increases and thus thermal fluctuations are much larger. This is the basic key to the prominence of flux creep and the appearance of measurable

resistance in high- T_c superconductors. According to Gray [1989], the maximum attainable J_c in the classical superconductors due to flux pinning is less than the depairing limit and thus the relevant ultimate limit on J_c . Thuneberg [1989] indicates, that the pinning theory, developed for the classical materials, might also be applicable to the high- T_c oxides. For the depinning limited case in which the field and current are parallel to the Cu-O planes, a $J_{cmax} \geq 2 \cdot 10^8$ A/cm² at $B \leq 20$ T is predicted for the high- T_c materials [Gray 1989].

Figure 3.7 Critical current density $J_c(B)$ at fixed temperatures [Adrian 1991].

In order to achieve high critical currents, the superconductor must be of high quality. For thin films, c-axis orientation is not sufficient; epitaxial growth in c-axis orientation is needed [Adrian 1991]. In bulk high- T_c material the critical current density is low. Larbalestier [1988] measured critical current densities in 2-20 T fields at 4.2 K and reported values ranging from 100 A/cm² to 4 A/cm², respectively. Single crystalline $YBa_2Cu_3O_{7-x}$ films exhibit a much higher J_c . Among many others, the following results were reported for $YBa_2Cu_3O_{7-x}$. $J_c(77\text{ K}) > 10^6$ A/cm² in 0.3 μ m films made by laser

ablation [McGuire 1989]. $J_c(77\text{ K}) = 10^6\text{ A/cm}^2$, and $J_c(4.2\text{ K}) = 1.5 \cdot 10^7\text{ A/cm}^2$, for a laser ablated thin film [Schilling 1990]. $J_c(77\text{ K}) > 10^6\text{ A/cm}^2$ for films made by DC-sputtering [Tome-Rosa 1990].

Figure 3.8 Critical current density $J_c(T)$ for fixed magnetic fields [Adrian 1991].

The critical current density is lowered in the presence of an external magnetic field. Figure 3.7 and Figure 3.8 show the critical current density in a thin $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film as a function of the field at fixed temperatures and as a function of temperature for fixed fields, respectively. The direction of the field in those measurements was perpendicular to the ab-plane.

Figure 3.9, measured by Schultz [1991], shows the influence of the orientation of the magnetic field on J_c . From this figure it can be concluded that at 77 K and low field, the $B \parallel c$ orientation has a slightly higher J_c . This property is however not always observed by others. The fact that the critical current density is larger for the field oriented parallel to the Cu-O planes is, according to many authors, due to *intrinsic flux pinning*.

This intrinsic pinning results from the coherence length along the c-axis which is comparable to the distance between the Cu-O planes, where the super-current is localized. It is thus energetically favorable for the vortices to have their core located between the Cu-O planes. The situation for vortices perpendicular and parallel to the ab-plane is shown in Figure 3.10.

The situation for Bi-Sr-Ca-Cu-O, which has a slightly higher T_c than $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$, is worse with respect to practical applications concerning currents in magnetic fields, because its flux line lattice is already molten at 30 to 40 K below T_c . Therefore it must be cooled to a much lower temperature than $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ for practical use [Tinkham 1991]. The only candidate which probably shows a better high field behaviour than YBCO thin films do, are the artificially prepared superlattices. In those materials, very thin superconducting layers are sandwiched between layers of other material. An increase of the critical field for an $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ / $\text{PrBa}_2\text{Cu}_3\text{O}_{7-x}$ multilayered film has been reported by Fischer [1990].

3.2.4 Comparison of the critical current density in bulk-, thin film- and multilayered superconductors

Various techniques can be used for obtaining high T_c bulk samples. The most common one is the powder sintering process. In this process, a pellet, pressed from the constituent elements in their stoichiometric ratio, is sintered in ambient oxygen. This results in a superconductor consisting of individual single crystalline grains in a random orientation, separated by non-superconducting links. These links are thin enough for the superconducting pairs to tunnel through and thus provide a weak coupling between the single crystalline grains. For this reason, the regions between the grains are called weak links and it is their presence that is the reason for a large difference between intragrain and intergrain critical current densities in bulk materials. J_c in the superconducting grains, the intragrain critical current density which can be obtained by magnetization measurements, can be as high as 10^5 A/cm^2 . The transport current density, the intergrain J_c , on the other hand varies between $10\text{-}1000 \text{ A/cm}^2$ [Cave 1989] and is even

more reduced in the presence of an external magnetic field. Apart from weak links, also the mismatch between two grains with different crystallographic orientations, resulting from the random grain orientation in bulk materials, is responsible for their low critical current densities. This is due to the fact that the best superconducting properties occur in the *ab*-crystal plane. The critical current density in bulk high T_c superconductors can be improved if the problem with the intergranular weak links is solved and the grains are aligned. Grain alignment can for example be done in a mixture of single crystal grains and liquid epoxy which hardens in a large magnetic field [Fang 1989].

Bulk materials which are most suitable for power applications can probably only successfully be applied when wires can be manufactured. However, making high- T_c wires is very difficult from a technological point of view. This because high- T_c materials are brittle, need to have a sufficiently high oxygen content and suffer from grain boundaries and crystallographic misorientation. Nevertheless, rather promising results can now be obtained with silver-sheated superconducting tapes [Shibuta 1991], because the silver tends to solve the intergrain problems.

It has been indicated that the critical current density in epitaxial thin films is much higher than in bulk materials. At 77 K, $J_c > 10^6$ A/cm² is routinely obtained, and at 4.2 K critical current densities over 10^7 A/cm² are reported. The maximum film thickness for these high current densities is about 0.5 μ m, because the crystal orientation changes from *c*-perpendicular to *c*-parallel with the substrate if the film thickness exceeds this critical value. An upper limit for the film thickness was found to be present in films made by various fabrication methods [Mogro-Campero 1990, Carim 1991]. In the work of Carim [1991], the transition from *c*-perpendicular to *c*-parallel with the substrate, in a laser ablated $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film, occurs at a thickness of 0.4 μ m and is explained by strain accumulation which relaxes above a certain critical thickness. This strain is due to a mismatch between the substrate and the superconductor crystal lattice.

Concerning $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ multilayers, two types can be distinguished. The first type are the $\text{ReBa}_2\text{Cu}_3\text{O}_{7-x}$ (Re = rare earth) multilayers with an alternating rare-earth element, e.g. $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}/\text{DyBa}_2\text{Cu}_3\text{O}_{7-x}$ or $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}/\text{PrBa}_2\text{Cu}_3\text{O}_{7-x}$. The other type of multilayers are those in which the adjacent layers have different crystal structure and lattice parameters. The lattice misfit that occurs can be accommodated if the layer

thickness is below a critical value. This misfit accommodation introduces strain in the multilayers which might be advantageous in terms of critical current density. The highest J_c obtained in Nd/Y multilayers was 10^7 A/cm² at 77 K, which is above the values reported for single films. This high value, still being limited by flux creep, was attributed to the increased number of defects present in the multilayer structure [Gross 1990].

3.2.5 Magnetic shielding

The effectiveness of magnetic shielding can be defined as:

$$S = -20 \log \left(\frac{H}{H_0} \right) \quad (3.3)$$

where S is the shielding factor in dB, H is the field measured in the presence of the sample and H_0 is the field without sample. A typical shielding factor for a planar disk of sintered $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ is 108 dB [Chen 1990]. This shielding factor was almost independent of the applied field unto the critical field of 1.7 mT. Above this critical field, the shielding factor showed a sharp decrease. For a cup shaped sample, a higher value of 160 dB has been reported [Zavaritsky 1989].

3.3 Possible mechanisms for electron pairing in high- T_c superconductors

Before discussing the theoretical approach with respect to high- T_c superconductors, we first give an overview of the experimental facts which may backup theory:

- the charge quantum is $2e$
- the flux quantum is $h/2e$
- charges are positive, as found by Hall measurements
- there is an energy gap which is about $3-7 k_B T_c$
- quasi-two-dimensionality because of large anisotropy

- weak isotope effect

These experimental results strongly indicate that the standard BCS assumptions do not directly apply to the high- T_c superconductors. In the BCS scheme, the charge carriers are weakly coupled through phonons and T_c is related to the Debye temperature as [Allen 1990]

$$T_c = 1.13 \Theta e^{-\frac{1}{V_0 N_F}}, \quad (3.4)$$

where Θ is the Debye temperature, V_0 the electron-electron interaction potential and N_F the density of states at the Fermi level. Substitution of any realistic number in eq.(3.4) cannot explain the high critical temperatures observed. Because of the weak isotope effect and the relatively low characteristic energy of phonons, theories focus on other mechanisms than phonon mediated coupling in order to obtain electron pairing. Possible mechanisms for electron-electron interactions may be charge fluctuations or spin fluctuations [Schrieffer 1988]. The charge fluctuation mechanisms responsible for the electron pairing may for example be excitons or plasmons.

3.4 Device applications

In low T_c research, many superconducting devices have already successfully been developed. Examples are microwave components such as oscillators, amplifiers and mixers [Likharev 1990]. In the field of superconducting electronics, most devices make use of tunnel junctions. To date, the most successful devices using tunnel junctions are the superconducting quantum interference devices (SQUIDS) which are used as high sensitive detectors of magnetic flux. In logic and computer applications some promising research results were obtained, with the 4-bit Josephson data processor chip as one of the most advanced examples [Kotani 1990]. According to Likharev [1990], classical superconducting devices as A/D converters and specialized microprocessors will become available in the not too distant future. Concerning large scale devices, applications in

magnet technology are probably the best known utilization of the low T_c materials, but the high cost of helium refrigeration has prevented large scale use. An overview of potential power applications for both low- T_c and high- T_c is given by Schneider [1991].

The main advantage related with the use of high- T_c superconductors is their ability to operate in the liquid nitrogen temperature range. This drastically reduces the cost of refrigeration, because liquid nitrogen both is about one hundred times cheaper and has a 160-fold larger heat of evaporation than liquid helium. Disadvantages that go with the use of high- T_c materials are [Likharev 1990]:

- the necessity to use highly oriented crystalline material
- the small coherence length
- vulnerability of the high- T_c properties to small distortions in composition and structure
- high chemical activity at the surfaces and at interfaces

Therefore, initial applications will be small scale, in specialized areas, where the additional cost and effort of using novel devices is acceptable by the inability to achieve the same result by other means [Gallop 1990]. The first applications will be in the field of superconducting electronics rather than in power devices.

When high- T_c materials are used for shielding, the volume to be shielded should be covered with a film of a few λ_L thick. For a single crystalline film, this corresponds to a thickness of about 1 μm . For polycrystalline films this thickness must be larger. A limitation for the use of present high- T_c materials as diamagnetic shielding is that the magnitude of the field to be shielded must be in the milli-Tesla range. However, this is enough for ambient field shielding.

Material parameters that are of importance with respect to high frequency applications are the surface resistance and the dispersion. For high- T_c superconductors, the surface resistance is below that of copper (at 77 K) unto frequencies of about 100 GHz [Withers 1990]. Moreover, the penetration depth is virtually frequency independent unto several hundred GHz for Niobium and perhaps several THz for the high- T_c materials, so negligible dispersion unto these frequencies can be foreseen. The first high frequency devices in which high- T_c superconductors are expected to be applied are passive microwave components like filters, resonators and delay lines [Withers 1990,

Adam 1990]. Experimental delay lines and strip lines which exhibit very significant improvements over copper at 77 K have in fact already been made. However, in order to let the high- T_c materials be competitive at relatively high temperatures and in the presence of high microwave powers, improvements in the film microstructure are still necessary [Rowell 1991].

As superconducting temperatures have moved up, the operating temperature of some semiconductor devices has moved down, so high temperature superconductors are candidate for application as interconnects in semiconductor systems [Malozemoff 1988]. Especially for intermediate range interconnects, superconductors are advantageous because of their low dispersion. For short range interconnects, superconductors are not performing better than metals do at the same temperature and for long range interconnects, the use of fibre optics is probably a better option.

Applications which require Josephson junctions as active elements are developed at a slower pace, because high- T_c Josephson junctions are difficult to fabricate due to the small coherence length and their sensitive behaviour to small deviations in stoichiometry and orientation [Likharev 1990]. Nonetheless, high- T_c SQUIDS made from natural weak links, which are at the boundaries between the grains of polycrystalline superconductors, have been made [Gallop 1990].

In the field of high- T_c superconducting wires, results are improving, but yet, they are not on a level where applications are feasible. For silver sheathed Bismuth-compound wires with aligned grains, a critical current density of $3.5 \cdot 10^5$ A/cm² could be achieved at 77 K and in zero magnetic field. At 77 K, the current density was only high unto 0.1 T. At 4.2 K, J_c was $6.0 \cdot 10^5$ A/cm² and decreased to $2.5 \cdot 10^5$ A/cm² in 1 T. The critical current densities were strongly affected by the degree of crystal orientation [Matsumoto 1991].

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4 Thin film fabrication

The key problems related with the deposition of high- T_c thin films are to obtain the correct composition, to produce a film with a uniform thickness, to minimize the film-substrate interactions and to obtain the correct crystal structure. Moreover, it is also most important to have the right amount of oxygen in the film in order to obtain good superconducting properties. In order to tackle these problems, a number of deposition techniques were proposed and tested [Leskelä 1989]. In this chapter, our main interest will be directed towards the so called physical vapor deposition methods which include sputtering, laser ablation, electron beam evaporation and Molecular Beam Epitaxy (MBE). We treat sputtering in section 4.2 and laser ablation in section 4.3. Evaporation, which has been part of this work, is treated in more detail, in section 4.4. That section gives an overview of the instrumentation we used, along with the fabrication procedure and some of the results. Finally, in section 4.5, the principles of Molecular Beam Epitaxy (MBE), Chemical Vapor Deposition (CVD) and Spray Pyrolysis will be outlined briefly.

4.1 Introduction

In the field of high- T_c superconductivity, deposition of thin films has become a large effort, because the properties of thin films are superior to those of bulk material and because thin films are needed for electronic applications which are thought to become one of the major areas of interest for high- T_c devices in the future. Nearly all applications anticipated for high- T_c superconducting thin films involve high frequencies, so, apart from requirements on the critical temperature and the critical current density, good high-frequency properties are important. Furthermore, the films should allow thermal cycling and their properties should not degenerate when exposed to ambient. For device and multi-level applications, the quality of the film surface along with film smoothness is quite important. Thick films ($>0.5 \mu\text{m}$) are undesirable for those applications, because these films mostly possess rough surfaces and mixed crystal

orientations which affect the critical current density [DiIorio 1990]. Finally, the deposition technique selected for production should have a high throughput.

Concerning the physical vapor deposition methods, two types of thin film preparation can be distinguished; the ex-situ method and the in-situ method. In the ex-situ or two step method, the metallic elements are deposited as an amorphous layer, usually in compound form with oxygen and possibly fluorine. Only after annealing in oxygen at about 850° C, which is the second step, an epitaxial polycrystalline layer will be formed [Humphreys 1990]. For two step deposition of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$, the use of BaF_2 instead of pure Ba is advantageous, because BaF_2 is more stable when exposed to air. This simplifies preparation of the deposition setup and improves the stability of the as deposited films, which in most cases have to go through air before annealing. When BaF_2 has been used, a mixture of water vapor and oxygen has to flow over the film during a certain time segment of the post deposition annealing procedure in order to reduce BaF_2 to Ba and HF, where the latter diffuses out of the film. Besides the annealing temperature itself, the heating- and especially cooling rates are important factors for obtaining good superconducting properties. Heating rates are mostly normal and set by the equipment, but slow cooling rates are important for obtaining the superconducting phase [Leskelä 1989]. The superconducting properties, in terms of critical temperature and critical current density of the ex-situ prepared $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films, based on BaF_2 , are good.

In in-situ deposition techniques, the film is deposited in crystalline form. It may need some low-temperature annealing to achieve the correct oxygen stoichiometry, but this step can often be omitted. No substantial rearrangement of the lattice after growth is required which results in superior films in terms of a smooth surface morphology, a sharp film-substrate interface and a well controlled epitaxial orientation. Important factors for successful in-situ preparation of high- T_c films are that the composition is close to stoichiometric, the growing film is sufficiently oxidized, and that a suitable substrate at the right temperature is used.

As already indicated, in order to obtain crystalline films, the substrate needs to be heated, but this stimulates unwanted film-substrate interactions. The desired interaction with the substrate can be summarized in the following three items [Leskelä 1989]:

- 1) negligible reactions between film and substrate.
- 2) good lattice matching.
- 3) matching of the thermal expansion coefficient between the film and substrate.

The ideal substrate meets those three interaction requirements and moreover, is cheap, is available as a large size crystal and is easily cut and polished. For high frequency applications it should additionally have a low dielectric constant and -loss and it should be compatible with IC technology [Humphreys 1990]. This compatibility is easy to obtain with substrates such as Si or GaAs. However, those materials very easily diffuse into the film at the substrate temperatures required by the deposition process (about 800° C) [DiIorio 1990]. For those reactive substrates, a buffer layer is necessary, mostly not only to prevent film-substrate interaction, but also to accommodate the difference in thermal expansion coefficients [Xi 1991]. Among the ceramic buffer layers, ZrO₂ seems to be best.

Table 4.1 Some parameters of the most common substrates used in superconducting thin film growth. YBa₂Cu₃O_{7-x} is added for comparison.

substrate	interactions [-- = none ++ = much]	lattice parameters [Å]	expansion coefficient [@ 300 K, x10 ⁻⁶ /K]	lattice type	price [US\$/cm ²]
YBa ₂ Cu ₃ O _{7-x}		a=3.8177 b=3.8836	a-axis: 9.6	orthorhombic	
SrTiO ₃	-	3.905	9.4	cubic	130
MgO	--	4.212	8.0	cubic	50
Al ₂ O ₃	+	5.13	5.0	cubic	
ZrO ₂	--	a=5.21		monoclinic	
LaAlO ₃	--	a=5.357 α=60°6'		trigonal	20
Si	++	a=5.4305	2.5	cubic	1

The first substrate materials that have been used for thin film growth were Al₂O₃, MgO, ZrO₂ and SrTiO₃. The best films have been grown on single-crystalline ZrO₂, MgO and particularly SrTiO₃. SrTiO₃ is the most widely used, but it has the disadvantage of not being perfectly inert and being only available in small sizes at a high price.

SrTiO_3 is not well suited for high frequency applications, because it has a large dielectric constant and nearly ferro-electric properties ($\epsilon_r > 1000$ and $\tan(\delta) > 0.01$ at 77 K). MgO , which is also widely used, has a poorer lattice match, but the dielectric constant makes the substrate suitable for high frequency applications. LaAlO_3 , which was later introduced as a substrate for high- T_c thin film growth, combines a good lattice matching with suitable dielectric properties. The best films in terms of T_c , J_c and orientation have been grown on SrTiO_3 and LaAlO_3 . An overview of the most common substrates is given in Table 4.1.

X-ray diffraction in the Bragg-Brentano geometry is mostly used to determine the crystal structure. In this geometry, the diffracting planes are parallel to the substrate and this technique thus gives information about the preferred crystallographic orientation normal to the substrate, but is incapable to determine epitaxy. For checking this in-plane alignment, it is necessary to investigate the alignment of the in-plane crystallographic axis of the film with respect to those of the substrate. Suitable techniques are Transmission Electron Microscopy (TEM), Reflective High Energy Electron Diffraction (RHEED) or grazing incidence x-ray diffraction. The advantage of RHEED is that it can be used to obtain structural information during growth. The most widely used methods for determination of the film composition are Energy Dispersive X-ray (EDX) and Rutherford Backscattering Spectrometry (RBS). The latter technique is also capable to give a depth profile of the composition.

4.2 Sputtering

Sputtering was one of the first deposition techniques applied in high- T_c thin film fabrication. The most frequently used sputter techniques are DC magnetron sputtering, RF magnetron sputtering, DC diode sputtering and RF diode sputtering. A less common sputter technique is ion-beam sputtering. Here material is sputtered off a target and onto a substrate using a Kaufmann ion gun operated with Ar. The technique was used by Gao et. al. [1988], who used an ion energy of 800-1000 eV and a beam current of 100-150 mA.

Target materials can be divided in sintered oxide targets and metal alloy targets. Sputter gasses are Ar or Ar+O₂. DC techniques offer less freedom in target choice, because the target needs to be conducting or semiconducting, which means for YBa₂Cu₃O_{7-x} that it must fully react to the orthorhombic structure. This can be a disadvantage when one wants to adjust the film composition by changing the target composition.

For deposition by means of sputtering, one expects the film composition to equal the target composition. However, for the "123" films, deviations in stoichiometry between target and film occurred. These deviations were found to be caused by resputtering of the growing film by negative oxygen ions [Xi 1991]. Solutions found to overcome this negative ion bombardment are:

- 1) Placing the target off-axis, in order to get the film out of the negative ion impact area.
- 2) The use of a high oxygen pressure (0.1-2.5 mbar) in order to decelerate the negative ions. This high oxygen pressure is also required when aiming for in-situ film processing.
- 3) The use of a sputter deposition geometry known as inverted cylindrical magnetron sputtering (ICMS).

Figure 4.1 Setup for inverted cylindrical magnetron sputtering (ICMS) [Xi 1991].

The ICMS sputter geometry is indicated in Figure 4.1. This system uses a cylindrical stoichiometric target, a low cathode voltage (100-150 V) and it can work with Ar/O₂ mixtures to a pressure of 800 mbar. The advantage of this geometry is that it minimizes oxygen resputtering, because oxygen ions are emitted in the direction of the opposite wall. The deposition rate of this system is high (500 nm/hour at a power of 50 W) compared to other off-axis geometries, because the cylindrical target has a large area and a gas flow from the top facilitates material transfer [Xi 1991].

A more advanced technique is multi target sputtering, which can be done with three sources, all pointing at the same substrate location. This multi target sputtering offers additional freedom for adjusting the composition, but, for the same reason, it is a more difficult technique, because the right composition is not easily obtained in the presence of a high oxygen background pressure [DiIorio 1990]. Multi-target sputtering can also be applied for growing multilayers. When insulating layers between the YBCO are required, PrBa₂Cu₃O_{7-x} is an ideal candidate, because it has an orthorhombic crystal structure with lattice parameters close to YBCO and a resistivity of 10⁷ Ωcm at 4.2 K. YBCO/PrBCO multilayers have successfully been grown using planar DC magnetron sputtering in a 400 mbar Ar+O₂ mixture with two magnetron guns, 180° apart, in combination with a rotatable substrate holder [Fischer 1990].

The electrical properties of sputtered films are among the best reported. T_c > 88 K and J_c is over 10⁶ A/cm² at 77 K [Stritzker 1990].

4.3 Laser Ablation

Laser ablation is a relatively simple technique, giving high quality films. The technique was first reported by Smith and Turner [1965] and turned into a well known thin-film deposition technique today. Laser ablation also became the simplest and most reproducible method for the in-situ fabrication of high-T_c 123 films. The advantages that go with laser ablation include [Blank 1991]:

- Power source outside the vacuum, which relaxes the demands on the vacuum system

- Deposition can take place in an oxygen background pressure high enough to enable in-situ thin film preparation
- The number of targets can easily be extended, which makes the in-situ fabrication of multilayers possible
- The laser may also be applied for patterning of the film

The reason for using laser ablation in high- T_c film deposition is that, as a result of the enormous energy density of the laser beam at the target, it allows to deposit materials containing elements with different vapor pressures [DiIorio 1990]. Therefore, the target composition is copied to the substrate. A drawback of the laser ablation technique is the small area which is uniformly deposited, because of the limited dimensions of the plume (see Figure 4.2). This problem can be overcome by using an off-axis rotating substrate.

Laser deposition occurs in a medium- or ultra high vacuum chamber. The laser beam is focused via a lens on a sintered pellet which is evaporated by means of short laser pulses. In order to provide the laser beam with fresh material, the target is rotated

during deposition. The vaporized material forms a plume which will partially sublime on the substrate, placed at the edge of the plume. A possible setup for laser ablation is given in Figure 4.2.

A short pulse and a short wavelength need to be used in order to minimize the spread of heat into the target and to minimize the reflectivity. Lasers applied for high- T_c thin film deposition include the pulsed excimer laser used in the ArF or the XeCl mode, the pulsed CO_2 laser and the Nd:YAG laser [Leskelä 1989]. Ablation with an excimer laser is very suitable for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$, because the reflectivity is only about 0.1-0.2 in the region of operation and the pulse time is short. In comparison, the reflectivity of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ is 0.8-1.0 for wavelengths above $1\text{ }\mu\text{m}$ [Blank 1991].

Introduction of oxygen into the vacuum chamber is required for oxidation. The oxygen pressure used by Blank et. al. [1989] is 0.3 mbar. The laser energy is 100 mJ/pulse and the energy density ranges from 0.5-3 J/cm². The pulse frequency range is 3-300 Hz, the pulse widths are in the millisecond to nanosecond range and the peak intensity at the target is about 10^7 - 10^8 W/cm² [Leskelä 1989].

Laser deposited films on SrTiO_3 or MgO orient with their c-axis perpendicular to the substrate. Good results in terms of T_c and J_c can be obtained. T_c above 90 K and J_c values as high as $5 \cdot 10^6$ A/cm² at 77 K are reported [DiIorio 1990].

The electrical and structural properties of laser ablated and sputtered films are similar. However, their surfaces are quite different, due to the presence of small droplets on top of laser ablated films. These droplets are absent on the surface of sputtered films. The droplet problem can be minimized by a proper adjustment of the ablation parameters [Blank 1991].

4.4 Evaporation

4.4.1 introduction

Evaporation is usually done in a high vacuum chamber using metals or metal alloys as source materials. The evaporation sources may be heated with a resistance wire, or, when metals with a high melting point are used, by means of an electron beam.

Evaporation of high- T_c materials cannot be done from a single source, because the source pellet would thermally decompose into the original oxides (CuO , BaO and Y_2O_3 for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$), which have different vapor pressures and thus does not result in stoichiometric films.

Good films can be made ex-situ as well as in-situ, but for the in-situ technique the high oxygen pressure is a problem. Fabrication of in-situ films in a relatively high oxygen background pressure, as in sputtering or laser ablation, cannot be applied for evaporation, because the high pressure would drastically reduce the mean free path of the evaporated material. Moreover, rate monitoring of the flux of e-beam sources becomes a problem as quartz crystal monitors are too slow and mass spectrometers work only reliable at low pressure. To overcome this problem, a rate monitoring technique based on atomic absorption was worked out by Missert et. al. [1989]. Furthermore, activated oxygen like O or O_3 may be used, which already gives full oxidation at a much lower pressure, enabling mass spectrometer rate monitoring. By using activated oxygen, in-situ films with $T_c=90\text{ K}$ and $J_c(77\text{ K})=4\cdot 10^6\text{ A/cm}^2$ have been obtained [Beasley 1989]. Another technique to circumvent background pressure problems is differential pumping.

Part of this work was devoted to the fabrication of ex-situ and in-situ $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films by means of evaporation. This will be described in the rest of 4.4.

4.4.2 fabrication equipment

The equipment used for fabricating $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ thin films is based on a Balzers UTS 500 vacuum system. The system is pumped by means of a turbo molecular pump and a Ti sublimation pump. The base pressure of the system, as measured by means of an ionization gauge near the turbo pump, is 10^{-8} mbar.

The vacuum system is equipped with three evaporation sources, two Balzers ESQ 300U electron guns and one Luxel Radak II effusion cell. In order to obtain a stable flux profile from the electron guns, the partial pressure above the melt is lowered by sweeping the e-beam over the melt with 500 Hz in x-direction and 4 kHz in y-direction.

Figure 4.3 Setup of the evaporation system used for growth of superconducting thin films.

As fabrication of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films requires control over the Y:Ba:Cu ratio within 1 %, a feedback loop is employed to maintain this ratio to 1:2:3. The monitoring part of the feedback loop is based on a mass spectrometer which is operated in a multiplexed mode in order to control all three elements with just one spectrometer. The flux signals coming from this spectrometer are calibrated by comparing them with a quartz crystal rate monitor. Flux adjustment is split into a high frequency and a low frequency part. Low frequency flux control is obtained by adapting the filament current of the electron gun, while the Wehnelt voltage is adapted in order to stabilize the high frequency flux variations. Under normal circumstances, the effusion cell does not need feedback, because once adjusted at a certain evaporation rate, the cell is stable enough for the duration of the run. However, the barium flux from the cell we used turned out to

sensitively depend on the pressure. Therefore, the flux from this cell is also controlled by means of a feedback loop.

In order to obtain the right $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ structure, the substrates need to be heated during deposition. In our system, the substrates are mounted in a stainless steel holder which is radiatively heated using a tantalum wire. The complete setup is given in Figure 4.3. More details about this system and the evaporation rate control are to be found in Schellingerhout [1988], Schellingerhout [1989] and Appelboom [1992].

4.4.3 fabrication procedure

For the fabrication of ex-situ films, the following scheme was used:

- melt the materials in the sources one at a time in order to remove the oxidized top layer as well as to remove contamination.
- calibrate the mass spectrometer signals for the various elements by comparing them with the quartz crystal rate monitor.
- set the fluxes of the various sources to the ratio 1:2:3 for Y, BaF_2 and Cu, respectively.
- open the sample shutter until a film of the desired thickness has been grown.
- close the sample shutter.
- shut down all sources.
- vent the vacuum system with dry Nitrogen.

After deposition, the samples were taken out and annealed under flowing oxygen according to the following scheme:

- turn the oxygen flow on and ramp the temperature up to 850 °C within one hour.
- pass the oxygen through boiling water prior to oven inlet, during ten minutes at 850 °C.
- keep the temperature at 850 °C during twenty minutes.
- ramp the temperature down to ambient at a rate of 100 °C/hour.

Figure 4.4 Setup of the ozone generator as used in the experiments.

For the fabrication of in-situ films, we applied ozone in order to obtain sufficient oxidation at a relatively low background pressure. The ozone was made in a so called "silent" discharge [Horváth 1985]. The ozone generator consists of a stainless steel rod in a glass tube through which oxygen flows (see Figure 4.4). The rod is driven by a high voltage source at 500 Hz. The ozone/oxygen mixture is leaked into a liquid nitrogen cooled glass chamber through a small orifice. When enough mixture is condensed, the chamber is pumped down in order to purify the ozone. This purification results from the fact that an ozone/oxygen mixture with a higher ozone content has a lower vapor pressure. When the ozone is needed in the growth chamber, the glass chamber is heated relative to the liquid nitrogen bath and the ozone flows into the growth chamber. Great care has been taken to avoid recombination of the ozone before it can reach the substrates. For this reason, the materials outside the growth chamber which come into direct contact with the ozone are either glass, PTFE or stainless steel. The ozone flow rate into the growth chamber can be controlled by adjusting the temperature of the glass chamber.

The scheme used to fabricate in-situ films was:

- melt the materials in the sources, one at a time in order to remove the oxidized top layer as well as to remove contamination.

-
- calibrate the mass spectrometer signals for the various elements by comparing them with the quartz crystal rate monitor.
 - heat the substrates to the desired temperature (700° C).
 - set the fluxes of the various sources to the ratio 1:2:3 for Y, Ba and Cu respectively.
 - turn the ozone flow on by heating the ozone container.
 - open the sample shutter until a film of the desired thickness has been grown.
 - close the sample shutter.
 - shut down all sources.
 - re-open the sample shutter and shut down the substrate heating in order to allow post oxidation by the ozone.
 - cool the substrates down to room temperature in about half an hour while the ozone flow remains on.
 - vent the vacuum system with dry Nitrogen.

The maximum ozone pressure that can be used during evaporation is limited to a value where the mass spectrometer sensitivity starts to decrease. This decreasing mass spectrometer sensitivity results in an increasing flux, because the feedback loop tends to keep the mass spectrometer signals constant. The increase in flux can be observed on the quartz crystal monitor and the ozone pressure has been raised until the mass flux had increased by 50 %. This happened at a pressure of 10^{-5} mbar as measured by the ionization gauge located near the pump. The ozone pressure at the substrate position was estimated to be 10^{-2} - 10^{-3} mbar.

4.4.4 Thin film analysis

The films were analyzed by means of R-T measurements, X-ray diffraction, Rutherford Backscattering (RBS) and Scanning Electron Microscopy (SEM). R-T curves of four films on various substrates, made either ex-situ or in-situ are shown in Figure 4.5. Table 4.2 shows their onset- and zero critical temperatures.

X-ray diffraction showed that the post annealed film on SrTiO_3 had better pronounced c-axis peaks than the in-situ films [Appelboom 1991]. RBS spectra were taken in order

to verify the composition, as well as the interdiffusion of the $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ with the substrate. According to these spectra there has been negligible interdiffusion for the films deposited on SrTiO_3 and Al_2O_3 . For the films deposited on Si, however, we found complete interdiffusion upto the film surface. The RBS spectra further indicated that the composition of the films deviates from the 1:2:3 ratio. This may be due to the difference in position of the quartz crystal rate monitor and the sample holder [Appelboom 1991].

Table 4.2 The R-T parameters $T_{c \text{ onset}}$ and T_{c0} for the various films.

type of deposition	$T_{c \text{ onset}}$ [K]	T_{c0} [K]
ex-situ on SrTiO_3	85	77
in-situ on SrTiO_3	90	85
in-situ on Al_2O_3	85	70
in-situ on Si	85	60

Figure 4.6 SEM photograph of the surface of a post annealed film. The size of the bar is 1 μm .

Figure 4.7 SEM photograph of the surface of an in-situ film. The size of the bar is 1 μm .

SEM photographs of the surface of the post annealed films show needle like out-growths, as can be seen in Figure 4.6. These needles, also observed by others [Mogro-Campero 1990, Watanabe 1989], are regions where the *c*-axis is in the plane of the film. SEM photographs of the surface of the in-situ films show submicron size droplets which

may be secondary phase particles (Figure 4.7). These secondary phases can be explained by the small deviations in stoichiometry as observed by RBS [Appelboom 1991].

4.5 Other Techniques

Searching for a well determined deposition technique, the choice for Molecular Beam Epitaxy (MBE) seems obvious, because in MBE the lattice is build layer by layer in an artificial way, thereby enabling the control over many parameters. However, for the deposition of high- T_c superconducting films, some problems arise, because a large number of individual elements has to be controlled in the presence of some form of oxygen. Even more than in e-beam evaporation, the presence of oxygen is incompatible with the principle of MBE, which requires a very low background pressure. For this reason, MBE is mostly similar to co-evaporation so far, although successful shuttered growth of $\text{Bi}_2\text{Sr}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_x$ on an atomic layer by layer basis has been reported [Schlom 1990].

Another technique which is not applied on a large scale yet, is Chemical Vapor Deposition (CVD). In CVD, film growth occurs via the decomposition of reactant gasses on a substrate in an oxygen rich atmosphere. Those gasses consists of metal in an organic solvent mixed with a non reactant carrier gas. Therefore, this deposition method is often referred to as Metal Organic-Chemical Vapor Deposition (MO-CVD). The properties of films made by CVD can be good. Watanabe [1989] reported a critical current density of 10^5 A/cm^2 in a 2 T field. CVD is often quoted as a production technique, as the throughput can be high when a large size CVD reactor is used where many wafers can be coated in parallel. Another advantage of CVD is the ability to coat non flat surfaces.

A simple non vacuum technique, which unfortunately does not give results comparable to those obtained with vacuum deposition techniques is spray pyrolysis of metalorganic compounds. During spray pyrolysis, a metalorganic solution is sprayed onto the substrate using a fine air brush with nitrogen used as a carrier gas. The substrate is usually at 400-500 °C. Some post deposition heat treatment is necessary in order to form the

required crystal structure and to fully oxidize the film. The T_c of those films is about 80 K and their J_c is in the order of 10^4 A/cm² at 65 K [Leskelä 1989].

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5 Experiments with a thin film coil

Before going to a more complete lens concept, we worked on the technology for making spiral shaped coils in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ thin films. This technological work, along with the results is presented in this chapter.

Section 5.1 introduces the process steps required to make a test pattern in an $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ thin film. The pattern consists of a 50 turns flat coil with a total length of about 70 cm. The width of the spiral track is $50\text{ }\mu\text{m}$. We employed Ar ion etching for patterning, since the films that were available to us for these tests were very sensitive to aqueous solutions. Later, when better films could be obtained, we applied wet chemical etching instead. This wet etching method is presented in chapter 8. The fabrication procedure for the thin films utilized in these experiments has been given in chapter 4.

Measurements on the patterned samples are in section 5.2, superconducting properties in section 5.2.1 and the axial field distribution of the 50 turns test coil in section 5.2.2.

5.1 Patterning

We patterned the $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films with the mask shown in Figure 5.1. This mask, made by means of e-beam lithography, is used to work out a patterning process and to measure the R-T and J_c -T behavior of long tracks in thin films. The pattern consists of a 50 turns spiral with an inner diameter of 2 mm and an outer diameter of 8 mm. The total length is about 70 cm, the width of the spiral track is $50\text{ }\mu\text{m}$ and the spacing between the turns is $10\text{ }\mu\text{m}$. Both the inner ring, which has a width of 0.5 mm, and the triangular pad are meant for contacting.

Figure 5.1 Pattern used to test the concept of high-Tc thin film coils [Adriaanse 1991b].

Prior to patterning, we deposited a $0.5\ \mu\text{m}$ layer of silver. This silver layer serves to protect the superconductor during patterning and also to provide an alternative path for the current at those spots in the superconductor that are of poor quality.

The patterning process itself consists of the steps listed below:

- spin Shipley primer on the silver layer at 4000 rpm during 40 seconds, in order to provide a better adhesion of the photoresist
- spin Shipley AZ 1375 on the substrate at 4000 rpm during 40 seconds
- bake the photoresist at 90°C during 15 minutes on a hotplate
- expose the photoresist with a mercury lamp during 4 minutes and 50 seconds
- develop the photoresist during 75 seconds in a 1:1 solution of Shipley microposit Developer and deionized water
- bake the photoresist at 120°C during 30 minutes on a hotplate
- etch the silver and the $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film with 300 eV argon ions at a current density of about $1\ \text{A}/\text{m}^2$ during 90 minutes
- remove the photoresist with acetone

Cooling of the $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film during ion etching is necessary in order to maintain the initial oxygen content in the superconductor. In our case, this has been taken care of by a substrate holder equipped with a water cooling.

5.2 Measurements

5.2.1 superconducting properties

Prior to the measurements, we attached wires to the $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ using silver paint, which results in contacts of sufficiently low resistivity. Figure 5.2 shows the resistance versus temperature curve of an evaporated $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film patterned with the mask shown in Figure 5.1. For comparison, Figure 5.2 also shows the R-T curve of the plain film, as measured prior to evaporation of the silver top layer and patterning. Figure 5.3 gives the J_c -T curve of the patterned film.

Figure 5.2 R-T curves of the plain and patterned film respectively. On the plain film no silver layer was present [Adriaanse 1991a,b].

Figure 5.3 J_c -T curve of the patterned film. The voltage criterion applied was $500 \mu\text{V}/\text{cm}$ [Adriaanse 1991a,b].

All curves, except the one obtained with the plain film, were measured over entire length of the spiral (70 cm), using a four probe DC technique. The voltage criterion applied in the J_c measurements was 500 $\mu\text{V}/\text{cm}$. We took this high value, instead of the more common 1 $\mu\text{V}/\text{cm}$, because the resistance of the patterned film was not entirely zero below T_c , as can be seen in Figure 5.2. The low temperature tail in the R-T curve of the patterned film shows a metallic behavior, which can be explained if one assumes bad spots in the superconductor where the current has to flow through the silver. The nearly horizontal part above T_c in the J_c -T curve (Figure 5.3) can also be attributed to the silver.

5.2.2 field distribution

The axial field distribution of the experimental thin film coil was measured by means of a pickup coil. We ran the experimental coil at an alternating current of 100 mA at 10 kHz in order to induce a voltage in the pickup circuit. The diameter of the pickup coil was 0.5 mm. We used a Si substrate with an Al layer, patterned with the experimental thin film coil in order to enable room temperature measurements of the axial field distribution.

This measured field distribution has been compared with a simple analytical model that only takes the rotational symmetric part of the field into account. The model, effectively being the sum of the axial field distributions of a series of concentric ring currents, reads:

$$B(z) = \frac{1}{2} \mu_0 I \cdot \sum_{j=1}^N \frac{R_j^2}{(R_j^2 + z^2)^{3/2}}, \quad (5.1)$$

where the number of current loops is N and their corresponding diameters range from R_1 to R_N . Figure 5.4 displays the axial field distribution as measured with the pickup coil, together with the distribution obtained with eq.(5.1). Figure 5.4 immediately shows that the fit between the model and the measured data is good in the region not too close to

the plane of the lens. The misfit near the lens plane is due to the finite size of the probe which is relatively more important close to the lens.

5.3 Conclusions

The previous sections indicate that patterning of a high- T_c thin film with a long track is possible and that its current density is reasonable, if we take into account that the $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film used was not entirely stoichiometric. A simple comparison with the current densities reported in literature, indicates that our result is more than two orders of magnitude lower, but one must keep in mind that those high values were mostly measured on short lines (e.g. $50\text{ }\mu\text{m} \times 200\text{ }\mu\text{m}$). In order to achieve such a high current density along a 70 cm long line, the film must be of high quality over a large area.

Another conclusion is that the silver top layer indeed seems to support conduction at bad spots in the superconductor. This can be concluded from the R-T curve of the patterned film which shows a metallic behavior below T_c (Figure 5.2).

Finally, we have the field distribution experimentally verified and showed that it can be measured relatively accurate by means of a pickup coil.

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6 Thin helical lenses and thin film lens elements

In this chapter we will introduce, in section 6.1, thin helical lenses as proposed by Bassett and Mulvey [1969]. In section 6.2, it will be concluded that lenses to be made in a superconducting thin film will, due to their flat geometry, always resemble this shape and that, moreover, two films are needed to make a round lens. In that section, we also treat several possible geometries for making thin film lenses. In section 6.3, we give the formulae for calculating the three dimensional field distribution of a thin film lens. First we apply the Biot-Savart law to obtain a set of integral equations for the three components of the magnetic field and finally, in section 6.3, we derive the axial multipole coefficients that are present in the field distribution of a thin film magnetic lens.

6.1 Introduction to thin helical lenses

6.1.1 focal properties

In conventional magnetic electron lenses, a large amount of space is occupied by the coil and the magnetic circuit. For some applications, those large dimensions are among the performance limiting factors. Especially in probe forming instruments, large lens dimensions limit the extraction of secondary electrons and x-rays from the specimen. For secondary electrons, the acceptance may be enlarged to a 2π solid angle by application of a through-the-lens-detection system [Kruit 1991], but for the detection of photons emitted in secondary processes, the size of the final lens is directly related to the performance.

A simple estimate for the focal properties of polepiece lenses can be obtained if the axial field distribution is assumed to be equivalent to that of a solenoid, shown in Figure 6.1, of axial length S and diagonal length $L = (S^2 + 0.45D^2)^{1/2}$ [Durandeau 1957]. The axial flux density distribution of a long thin solenoid is

$$B_z = \mu_0 \frac{NI}{2S} (\cos \alpha_1 - \cos \alpha_2), \quad (6.1)$$

where α_1 and α_2 are indicated in Figure 6.1. If the field distribution of this so called equivalent solenoid [Mulvey 1973] is assumed to be a block field, the first order properties of all symmetrical magnetic polepiece lenses within the geometrical limits $0.2 < S/D < \infty$ and the excitation limits $0 < NI < 1.5NI_0$ are readily calculated [Dugas 1961]. NI_0 is the excitation needed to obtain minimum projector focal length.

When the lens properties are normalized against the previously mentioned length L and the excitation is normalized against $NI_0/\sqrt{V}$, where V is the beam energy in electron volts, the properties can be represented in the form of one universal curve. This curve gives a general representation of the lens properties for a wide variety of polepiece geometries and excitations, provided the iron circuit is not saturated. Because the universal curve is based on the equivalent solenoid principle, it is also possible to use this curve to determine the focal properties of real solenoids with $S/D > 0.3$ [Bassett 1969]. A conclusion which can be drawn immediately from the equivalent solenoid principle as indicated in Figure 6.1, is that an iron free solenoid is able to produce the same focal properties as a conventional polepiece lens at much smaller dimensions. This is only true, however, if the coil can carry the necessary current, because where polepiece lenses are limited by the saturation of the ferromagnetic circuit, solenoid or iron-free lenses are limited by the maximum current density allowed in the windings.

A physical realization of the equivalent solenoid principle is the *mini lens* [Le Poole 1964] designed to overcome the space problem in an electron probe forming system. This mini lens is a long solenoid with a current density of 8000 A/cm^2 , which is fairly large for a non-superconducting coil. However, because of the lack of precision in

Figure 6.1 Equivalent solenoid. The length of the equivalent solenoid is equal to the gap length and its diameter is 2/3 of the polepiece bore [Mulvey 1973].

Figure 6.2 Flat helical lens and corresponding parameters [Bassett 1969].

obtaining axial symmetry, a large astigmatism resulted.

In contrast to the long solenoid, a flat helical lens or pancake lens [Bassett 1969], as shown in Figure 6.2, with D_2/D_1 large appears to be the optimum shape for an iron free lens of least spherical aberration. The axial flux density of a flat helix may be written as [Mulvey 1982]

$$B_z = \mu_0 \frac{NI}{2l} \left[\cos \alpha_1 - \cos \alpha_2 + \ln \left\{ \frac{1 + \cos \alpha_2}{1 + \cos \alpha_1} \frac{D_2 \cos \alpha_1}{D_1 \cos \alpha_2} \right\} \right], \quad (6.2)$$

where l , α_1 , α_2 , D_1 and D_2 are indicated in Figure 6.2. The angles α_1 and α_2 are defined between the lines joining the ends of the coil and a line *perpendicular* to the axis, instead of parallel as in Figure 6.1. Therefore, eq.(6.2) has the same form as eq.(6.1) and the analogy between the radial length l of a thin helical lens and the axial length S of a thin solenoid becomes clear [Mulvey 1973]. The peak flux density of a flat helix may be written as

$$B_{\max} = \mu_0 \frac{NI}{l} \cdot \ln \left(\frac{D_2}{D_1} \right)^{1/2}, \quad (6.3)$$

which is, apart from the logarithmic factor, identical to the peak flux of a thin solenoid. So, if $D_2/D_1 > 7.4$, the logarithmic factor is larger than one and the maximum flux density of a flat helical lens of radial length l will exceed that of a long solenoid of axial length S . As a consequence, the Full Width at Half Maximum (FWHM) value of the field distribution of the helical lens must be smaller than that of the solenoid, as the flux density integral over the axis must be equal to the number of ampere turns for both coil types. Therefore, the spherical aberration which is proportional to the previously mentioned half width is smaller in a helical lens than in a long solenoid. The favorable optical properties associated with a flat coil are valid for $S/D_m < 0.1$ [Mulvey 1982], where D_m denotes the mean diameter.

Figure 6.3 Unified focal properties of a flat helical lens. $(f_p)_{\min}$ denotes the minimum projector focal length and NI_0 is the corresponding excitation [Mulvey 1973].

Because the optical properties of flat lenses are not limited by the properties of the iron, they can very well be described in terms of universal curves as shown in Figure 6.3. Those generalized focal properties of thin helical lenses, given in Figure 6.3 for two different values of D_2/D_1 , approach closely those of long solenoids for relative excitations $NI/NI_0 < 1.5$.

Table 6.1 Polepiece lens excitation for minimum projector focal length and relative minimum projector focal length for several gap to bore ratios. S and D denote the gap length and bore respectively and L denotes the diagonal of the equivalent solenoid as shown in Figure 6.1 [Lenz 1982].

$\frac{S}{D}$	0	0.25	0.5	1.0	2.0	∞
$\frac{NI_0}{\sqrt{V}}$	14.4	14.3	14.1	13.5	12.7	10.9
$(\frac{f_p}{L})_{\min}$	0.565	0.549	0.520	0.492	0.491	0.550

Table 6.2 Helical lens excitation for minimum projector focal length and relative minimum projector focal length for several values of D_2/D_1 . L is the diagonal of the coil which reduces to $(D_1 + D_2)/2$ for a flat lens [Mulvey 1973].

D_2/D_1	1.22	1.50	1.86	2.33	3.00	4.00	5.67	9.00	19.00
$NI_0/\sqrt{V}$	15.2	15.4	15.6	15.8	16.0	16.2	16.4	16.6	16.8
$(f_p/L)_{\min}$	0.481	0.476	0.472	0.467	0.455	0.440	0.425	0.385	0.350

From Figure 6.3 it can be seen that for $D_2/D_1=19$, the objective focal length has a minimum at $NI/NI_0 \approx 1.4$. This minimum objective focal length at a physically attainable excitation is one of the characteristic features of flat helical lenses. In general, the objective focal length has a minimum if the tail of the field distribution falls off according to a power law, $B(z) \propto |z|^{-\nu}$ and $\nu > 3/2$ for large z [Lenz 1982]. If the field falls off exponentially, $B(z) \propto \exp(-C|z|)$, f_{obj} decreases monotonically with increasing lens strength, but has a finite limit for the excitation going to infinity. When the field falls off more than exponentially, so $B(z) \propto \exp(-C|z|^\gamma)$ with $\gamma > 1$, f_{obj} decreases monotonically towards zero for the excitation going to infinity. For a flat helical lens, the rotational symmetric part of the field distribution may be approximated by eq.(5.1) which is the superposition of N circular current loops, all located at $z=0$, with diameters ranging from D_1 to D_2 . For large z , this field distribution falls off as $B(z) \propto z^{-3}$, which is well within the class of fields which exhibit a minimum objective focal length. This minimum

objective focal length of flat helical lenses is appreciably larger than the focal length of polepiece lenses. On the other hand, the minimum projector focal length for helical lenses can be appreciably shorter than for long solenoids and iron polepiece lenses. However, as can be seen by comparing Table 6.1 with Table 6.2, the excitation to reach the point of minimum projector focal length is larger in helical lenses [Mulvey 1973].

6.1.2 spherical aberration

In order to make the comparison between helical lenses and polepiece lenses more complete, we should not stick to the first order properties, but extend the comparison with aberrations. A general trend with respect to aberrations is that a long small diameter solenoid corresponds to the lens of optimum chromatic aberration. A thin helix with a large ratio of outer to inner diameter corresponds to the lens of least spherical aberration. However, C_s of a long solenoid is infinite, whereas C_c of a helix is only of the same order as its focal length [Mulvey 1982].

A more or less fundamental lower limit of spherical aberration, for every image made with a rotational symmetric lens, free of space charge, is $C_s B_{\max}/\sqrt{V} \geq 2.23 \cdot 10^{-6} \text{ mTV}^{-1/2}$ [Tretner 1954, 1955]. For comparing the spherical aberration of a physical lens with this minimum, its coefficient of spherical aberration needs to be multiplied by $B_{\max}/\sqrt{V}$ which results in a normalized value for the spherical aberration that is independent of both lens size and beam energy. For polepiece lenses, this results in $C_s B_{\max}/\sqrt{V} \approx 5 \cdot 10^{-6} \text{ mTV}^{-1/2}$ as the best attainable value. Flat helical lenses on the other hand, show $C_s B_{\max}/\sqrt{V} = 3.05 \cdot 10^{-6} \text{ mTV}^{-1/2}$ for $D_2/D_1 = 19$, and even go down to $2.87 \cdot 10^{-6} \text{ mTV}^{-1/2}$ at $D_2/D_1 = 1000$ [Mulvey 1982]. A graphical comparison between the relative spherical aberration of polepiece lenses and flat helical lenses is given in Figure 6.4. In this figure it can be seen that, for $D_2/D_1 = 19$, the relative spherical aberration has a strongly pronounced minimum.

Theoretically, the phenomenon of minimum relative spherical aberration is not restricted to helical lenses, although in practice, the excitation at the point of minimum aberration cannot be reached in polepiece lenses due to saturation effects.

Figure 6.4 Relative spherical aberration in polepiece lenses and helical lenses [Mulvey 1973].

Figure 6.5 Relative spherical aberration against relative objective focal length for a polepiece lens and a flat helical lens [Mulvey 1973]. The limits according to Tretner [1959] are indicated.

A graphical representation of the relative spherical aberration against the relative objective focal length is in Figure 6.5. Besides a minimum spherical aberration, this graph also denotes the minimum relative focal length which is obtained in a constant field and has a value of $6.75 \cdot 10^{-6} \text{ mTV}^{-1/2}$ [Tretner 1959].

From Figure 6.4 it is clear that we have to make D_2/D_1 as large as possible in order to minimize the relative spherical aberration. However, in order to determine a minimum value for given physical constraints such as the dimensions and the current density, we better use the curves of Figure 6.6 [Bassett 1969]. Using Figure 6.6, we may write for the spherical aberration:

$$C_s = D_1 \cdot g(\Gamma, k) , \quad (6.4)$$

where $\Gamma = D_2/D_1$, $k = NI/\sqrt{V}$ and $g(\Gamma, k)$ are the curves displayed in the figure. Figure 6.6 also indicates that the minima of $g(\Gamma, k)$ are close to $k = 25$ for the larger values of Γ and that $g(\Gamma, k)$, for smaller values of Γ , does not differ significantly from the minimum value over a wide range of k . Therefore, $k = 25$ results in minimum spherical aberration for most Γ . Those minima of the normalized spherical aberration as a function of Γ are in

Figure 6.7 Minimum values of the normalized spherical aberration coefficient versus Γ .

Figure 6.7. If we look at Figure 6.7 and fit a straight line through the points, we get

$$g(\Gamma, 25) \approx C_0 + C_1 \Gamma, \quad (6.5)$$

where C_0 and C_1 are constant and shown in Figure 6.7.

Substituting eq.(6.5) in eq.(6.4) gives

$$C_s \approx D_1 \cdot [C_0 + C_1 \Gamma] = C_0 \cdot D_1 + C_1 \cdot D_2. \quad (6.6)$$

For the number of ampere turns NI , we may write

$$NI = \frac{1}{2} \eta JS(D_2 - D_1), \quad (6.7)$$

where S is the axial coil length, J the current density and η the coil fill factor ($\eta \leq 1$). If we substitute $NI = k\sqrt{V}$ in eq.(6.7) and rearrange the equation we obtain the following expression for D_2 :

$$D_2 = D_1 + \frac{2k\sqrt{V}}{\eta JS} . \quad (6.8)$$

Finally, if we substitute eq.(6.8) in eq.(6.6), we obtain the following expression for the spherical aberration of a helical lens:

$$C_s \approx C_1 \frac{2k\sqrt{V}}{\eta JS} + (C_0 + C_1)D_1 . \quad (6.9)$$

So, for a given beam voltage, critical current density, axial coil length and coil fill factor, the choice for D_1 determines C_s , because k is always close to 25. However, in practice, due to a limited current density and film thickness, the spherical aberration may be dominated by the first term of eq.(6.9). Substituting $k\sqrt{V}=NI$ and $J=NI/(S l)$ in eq.(6.9), shows, that this first term is in fact the radial coil length $l=(D_2-D_1)/2$. Therefore, when aiming for minimum spherical aberration at a given radial coil length l , which follows from practical constraints, the upper limit for D_1 can be found using eq.(6.9).

It must be emphasized that eq.(6.9) is only an approximation based on eq.(6.5). For a more exact treatment one needs to take the full function $g(\Gamma, k)$ into account.

6.2 Introduction to thin film lens elements

6.2.1 the lens element concept

One of the objectives of this work is to investigate the high T_c materials on their applicability in particle optics. When wires made of high T_c material with reasonable critical current density become available, the superconducting lens technology already developed in the nineteen sixties and seventies could be applied at liquid nitrogen temperature, which would take away some of the previous disadvantages. But high T_c superconducting coils and wires are not available yet. This is one reason why we have focused on thin films. The other reason is that thin films can be patterned very

accurately, a critical demand when aiming for small iron-free lenses, for which the mechanical accuracy requirements apply directly to the coil.

Using present high- T_c thin film technology, which results in films on flat substrates, we are forced by the geometry to construct flat helical lenses. In order to create a magnetic lens in a thin film, the film must be patterned with some kind of spiral as shown in Figure 6.8. An estimate for the number of ampere turns can be obtained using eq.(6.7). We have substituted the following numbers in eq.(6.7):

- $J = 10^{10} \text{ A/m}^2$ (10^6 A/cm^2) which is an acceptable value for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ at 77 K.
- $S = 0.5 \text{ } \mu\text{m}$, which is roughly the maximum film thickness on SrTiO_3 [Carim 1991].
- $D_1 = 1 \text{ mm}$, in order to allow the electron beam to pass the lens with a reasonable opening angle and in order not to make the demands on the accuracy too strict.
- $D_2 = 1 \text{ cm}$, which is the maximum area homogeneously deposited in laser ablation.

Substituting those values in eq.(6.7) results in about 20 ampere turns. At liquid helium temperature, where the current density is at least an order of magnitude higher, the number of ampere turns is about 200. So, although the critical current density in high- T_c thin films is large, the maximum number of ampere turns generated by a single thin film coil is small compared to the excitation needed in a typical application.

An estimate for the number of ampere turns needed in a helical lens, operated at its minimum projector focal length, can be obtained from Table 6.2. The average value listed there for $NI/\sqrt{V}$ is 16, so

$$NI \approx 16\sqrt{V} . \quad (6.10)$$

Thus, for a 10 kV beam, 1600 ampere turns are needed, which is eight times the maximum generated with a thin film coil at liquid helium temperature and eighty times

Figure 6.8 Pattern of a thin helical lens to be made in a thin film.

the maximum at liquid nitrogen temperature. From the example just given it is clear that, using present high- T_c technology, more than one thin film element is needed for any practical application with a beam energy over a few hundred electron volt.

If we somehow increase the number of ampere turns, for example by stacking thin film elements, we raise the field strength. However, due to the effects described in chapter 1 and chapter 3, the superconductor exposed to this strong field may exhibit a maximum current density that is significantly reduced. Whether the maximum current density will be influenced much by the lens field, being mainly perpendicular to the substrate, can be estimated if we use eq.(6.3) for the maximum axial field. If we substitute 10^4 ampere turns, the maximum used in conventional transmission microscopes, and again $D_1 = 1$ mm and $D_2 = 1$ cm, the maximum axial field according to eq.(6.3) is about 3.2 T. Especially at liquid helium temperature this field does not have too much influence on the current density. However, for stronger fields or for operation at liquid nitrogen temperature, we have to take the deterioration of the current density into account.

Besides increasing the magnetic field, there is another argument in favor of stacking. This is the demand that the lens field must be highly rotationally symmetric, a requirement which cannot be met when using only one spiral-shaped coil. This non-rotational symmetry can be understood if one recognizes that the innermost turn of a spiral coil ends in a bonding pad where a wire transporting the current to or from the coil center has to be connected. This

Figure 6.9 Thin film lens element consisting of two spirals in a face to face geometry. The direction of the current flow is indicated.

asymmetry can be eliminated by making contact to a second outward spiralling coil, which moreover, because of its opposite spiralling sense, corrects for the non-rotational symmetry inherently present in a spiral (see sections 6.2.2 and 6.3.2).

In-situ stacking of thin films, with alternating structures of superconducting and substrate layers is definitely the best way to obtain a strong rotationally symmetric lens

field. Unfortunately, this is not a common technology yet, especially not when aiming for an individual patterning of the superconducting layers. This individual patterning is necessary, as inward and outward spiralling layers need to be stacked in an alternating sequence. Therefore, we took an intermediate step and developed a technique for stacking two patterned superconducting films, together with their substrates, in a face to face geometry. In this geometry, the innermost turns of the spirals are to be connected by a non-superconducting contact. We will call such a stack of two films a *thin film lens element* throughout the rest of this thesis, because it is the smallest entity for building thin film magnetic lenses. Such a lens element is shown in Figure 6.9.

An alternative for this face to face stacking is a substrate having patterned films on both sides, with the innermost turns of the spirals connecting each other through the substrate. However, we prefer face to face stacking over a substrate patterned on both sides, because the length of the non-superconducting path between the spirals can be short and because, in this way, the two spirals are closer together which results in a more rotationally symmetric field. Besides, deposition of a superconducting layer on both sides of a substrate is a much more difficult technique, because the substrate cannot be heated by gluing its backside to a heater, as this would damage the backside film. Moreover, some special tools are needed in order to facilitate patterning of a two sided substrate without damaging the backside layer.

6.2.2 various types of lens elements

As indicated in section 6.2.1, a lens elements consists of two thin film spirals in a stacked geometry. In that section we did not mention the shape of the spirals. The most simple spiral, to which we will sometimes refer to as first order spiral, is described by:

$$R(\varphi) = R_0 + \frac{\delta}{2\pi} \cdot \varphi . \quad (6.11)$$

The parameters used in eq.(6.11) are shown in Figure 6.10, where the center of the spiral track is drawn with a dotted line and the corresponding central radius with a dash-

dotted line. For a spiral having N turns, ϕ runs from 0 to $2\pi \cdot N$. The x-y plane projection of a lens element built with two first order spirals is given in Figure 6.11. In this lens element, the mean radius along every line $A-A$ fulfills

$$\frac{\rho_1 + \rho_2}{2} = \frac{1}{2}D . \quad (6.12)$$

This stacking of two first order spirals in a face to face geometry greatly improves the rotational symmetry of the field. However, the contribution to the field of a certain section of the spiral is, for z -values near the lens plane, proportional to $1/R$ and we may thus try to further improve upon the rotational symmetry if we replace eq.(6.12) by

$$\frac{1}{R(\phi)} + \frac{1}{R(2\pi - \phi)} = \frac{2}{\rho_c} , \quad (6.13)$$

where the two lefthand terms denote the contributions of the two spirals in the lens element and ρ_c denotes the central

radius. Because of the rotation about the x -axis, the radius of the second spiral at an angle ϕ equals the radius of the first one at $2\pi - \phi$ and we can, therefore, express eq.(6.13) in the coordinates of just one spiral $R(\phi)$.

If we want the radius of the spiral to be increased by δ at each revolution, we obtain the following boundary condition

Figure 6.10 One turn of a first order spiral. The dash-dotted line indicates the central diameter.

Figure 6.11 XY-plane projection of a lens element, built with two first order spirals. The mean radius along every line oriented according to A-A is $(\rho_1 + \rho_2)/2 = D/2$.

$$\begin{aligned} R(0) &= R_0 , \\ R(2\pi) &= R_0 + \delta . \end{aligned} \tag{6.14}$$

If we now substitute those values, valid for $\varphi=0$, in eq.(6.13), we obtain for ρ_c :

$$\rho_c = \frac{R_0 + \delta}{1 + \frac{\delta}{2R_0}} , \tag{6.15}$$

which for small δ/R_0 is approximately $R_0 + \frac{1}{2}\delta$. In a first order spiral, eq.(6.13) can only be satisfied in one point, which is at $\varphi=0$ in case of the current definition for ρ_c . To satisfy eq.(6.13) in two points, we need a second order spiral given by

$$R(\varphi) = R_0 + c_1\varphi + c_2\varphi^2 , \tag{6.16}$$

where c_1 and c_2 are set by the boundary conditions. To determine c_1 and c_2 , we use the following boundary conditions

$$\begin{aligned} R(\pi) &= \rho_c , \\ R(2\pi) &= R_0 + \delta . \end{aligned} \tag{6.17}$$

This gives for c_1 and c_2

$$\begin{aligned} c_1 &= \frac{4(\rho_c - R_0) - \delta}{2\pi} , \\ c_2 &= -\frac{2(\rho_c - R_0) - \delta}{2\pi^2} , \end{aligned} \tag{6.18}$$

with ρ_c given by eq.(6.15). It must be noted that c_1 and c_2 need to be calculated for each individual turn and that for small δ/R , where $\rho_c \approx R_0 + \frac{1}{2}\delta$, $c_1 \rightarrow \delta/2\pi$ and $c_2 \rightarrow 0$. So, the difference between a first and second order spiral is maximum for the innermost turns.

The previous paragraph indicates that a solution for $R(\varphi)$ which satisfies eq.(6.13) for all φ from 0 to 2π must have the form of an infinite power series, given by

$$R(\varphi) = \frac{R_0}{1 - a\varphi} . \quad (6.19)$$

The constant a can be found through the boundary condition eq.(6.14) which gives

$$a = \frac{\delta}{2\pi(R_0 + \delta)} . \quad (6.20)$$

Note that eq.(6.19) is only valid for φ from 0 to 2π . Consequently, R_0 must be increased by δ each revolution.

In the previous sections we did not take the *finite width* of the track into account. If we assume that a certain section of the spiral contributes to the total strength proportional to $1/R$, we can map the path followed by a real, non-zero width, spiral into that of a zero-width spiral having the same field distribution. For a spiral where the track has a width $2w$ (see Figure 6.10), we obtain the path of the corresponding zero-width spiral as follows

$$\frac{1}{\rho(\varphi)} = \frac{1}{2w} \int_{-w}^w \frac{d\xi}{S(\varphi) + \xi} , \quad (6.21)$$

where $\rho(\varphi)$ is the path of the zero-width track and $S(\varphi)$ describes the center of the path followed by the non-zero width track. If we solve $S(\varphi)$ from eq.(6.21), we get

$$S(\varphi) = w \cdot \coth \left(\frac{w}{\rho(\varphi)} \right) , \quad (6.22)$$

which simplifies to

$$S(\varphi) = \rho(\varphi) + \frac{1}{3} \frac{w^2}{\rho(\varphi)} - \dots \quad (6.23)$$

for small w/ρ . As a result, we may use eq.(6.22) to prescribe the path of a non-zero width spiral for a given zero-width distribution $\rho(\varphi)$, which can for example be eq.(6.19). However, eq.(6.23) shows that the corrections will be negligible small for most practical dimensions.

If we want to obtain a very high rotational symmetry, we can make components of a lens element using only half circles connected by radial parts. This geometry is indicated in Figure 6.12(a). In Figure 6.12(b), which gives the corresponding complete lens element, it shows that the circular parts add up to complete circles and the current in the radial parts cancels. However, this is only true if the axial spacing between the parts is zero. A disadvantage of the type of lens element indicated in Figure 6.12 is that only half of the available area is used.

The previous paragraphs have indicated that a non-rotational symmetry is inherently present in the field distribution of any thin film lens element. The effect this asymmetry imposes upon the performance is of course application dependent and can be manipulated by a proper selection of both the winding pitch and the type of spiral. In the next section, we will find the magnetic field distribution of a first order spiral. The non-rotational symmetric part of this field may serve as an upper limit for the parasitic fields of spiral lenses, as a first order spiral has the lowest rotational symmetry of all spiral types presented in this section.

6.3 The magnetic field of a lens element

6.3.1 the magnetic field using Biot Savart

For a general spiral described by $\rho(\varphi)$, we can calculate the field distribution at an arbitrary point (x,y,z) using the Biot Savart law

$$\mathbf{B}(x,y,z) = \frac{\mu_0 I}{4\pi} \int_p \frac{\mathbf{dl} \times \mathbf{r}_1}{r^2}, \quad (6.24)$$

where $\mathbf{dl}$ points along the spiral $\rho(\varphi)$, $\mathbf{r}_1$ points from a point at the spiral to (x,y,z) and r is the distance between a point at the spiral and (x,y,z) . Those parameters are indicated in Figure 6.13. For points at the spiral we may write,

$$\begin{aligned} x &= \rho(\varphi) \cos(\varphi), \\ y &= \rho(\varphi) \sin(\varphi), \\ z &= 0. \end{aligned} \quad (6.25)$$

This gives for $d\mathbf{l}$

$$d\mathbf{l} = d \begin{pmatrix} \rho(\varphi) \cos(\varphi) \\ \rho(\varphi) \sin(\varphi) \\ 0 \end{pmatrix} = \begin{pmatrix} \rho'(\varphi) \cos(\varphi) - \rho(\varphi) \sin(\varphi) \\ \rho'(\varphi) \sin(\varphi) + \rho(\varphi) \cos(\varphi) \\ 0 \end{pmatrix} d\varphi, \quad (6.26)$$

for $\mathbf{r}_1$

$$\mathbf{r}_1 = \frac{1}{r} \begin{pmatrix} x - \rho(\varphi) \cos(\varphi) \\ y - \rho(\varphi) \sin(\varphi) \\ z \end{pmatrix}, \quad (6.27)$$

and for r

$$r = \sqrt{(x - \rho(\varphi) \cos(\varphi))^2 + (y - \rho(\varphi) \sin(\varphi))^2 + z^2}. \quad (6.28)$$

Substituting eq.(6.26), eq.(6.27) and eq.(6.28) in eq.(6.24) gives for the field components of an arbitrary spiral $\rho(\varphi)$:

$$B_x(x,y,z) = \frac{\mu_0 I}{4\pi} z \int_0^{\varphi_{\max}} \frac{\rho'(\varphi) \sin(\varphi) + \rho(\varphi) \cos(\varphi)}{((x - \rho(\varphi) \cos(\varphi))^2 + (y - \rho(\varphi) \sin(\varphi))^2 + z^2)^{3/2}} d\varphi, \quad (6.29)$$

$$B_y(x,y,z) = -\frac{\mu_0 I}{4\pi} z \int_0^{\varphi_{\max}} \frac{\rho'(\varphi) \cos(\varphi) - \rho(\varphi) \sin(\varphi)}{((x - \rho(\varphi) \cos(\varphi))^2 + (y - \rho(\varphi) \sin(\varphi))^2 + z^2)^{3/2}} d\varphi, \quad (6.30)$$

$$B_z(x,y,z) = \frac{\mu_0 I}{4\pi} \int_0^{\varphi_{\max}} \frac{(\rho'(\varphi) \cos(\varphi) - \rho(\varphi) \sin(\varphi))(y - \rho(\varphi) \sin(\varphi)) - (\rho'(\varphi) \sin(\varphi) + \rho(\varphi) \cos(\varphi))(x - \rho(\varphi) \cos(\varphi))}{((x - \rho(\varphi) \cos(\varphi))^2 + (y - \rho(\varphi) \sin(\varphi))^2 + z^2)^{3/2}} d\varphi. \quad (6.31)$$

It must be noted that for the simple spiral as described by eq.(6.11), $\rho'(\varphi)$ reduces to $\delta/2\pi$. In principle, eq.(6.29) to eq.(6.31) allow to calculate the field of any arbitrary spiral at any point in space. However, to the authors knowledge, no analytical solutions exist for those equations, not even for the case of a simple spiral as given by eq.(6.11). It is of course possible to perform numerical integration and obtain the field in this way, but this requires a lot of computer time, especially if the number of turns is large. Consequently, raytracing in this field, which is needed to investigate the effect of the non-rotational symmetric aberrations, will be very slow. This is one reason why we worked out a method to determine the axial multipole coefficients of a spiral and describe the three dimensional field using a radial series expansion. Another reason is that a multipole expansion provides a tool to gain a more general insight in the effect the geometry imposes upon the various parasitic aberrations. Furthermore, it enables to make the description more general.

6.3.2 unified representation for the magnetic field of a one turn spiral using a multipole expansion

A well known method in electron optics for describing a three dimensional field is by means of a multipole expansion, as e.g. given by Hawkes and Kasper [1989]. Throughout this work we will follow their notation with the exception of B_1 and B_2 , the x- and y-components of the dipole field, respectively, for which we substituted P_1 and Q_1 in order to make the notation more uniform. Their multipole expansion unto terms of third order gives:

$$\begin{aligned}
 B_x(x,y,z) = & -\frac{x}{2}B'(z) + \frac{x}{16}(x^2+y^2)B'''(z) \\
 & +P_1(z) - \frac{1}{8}(3x^2+y^2)P_1''(z) - \frac{1}{4}xyQ_1''(z) \\
 & -xP_2(z) - yQ_2(z) + \frac{1}{6}x^3P_2''(z) + \frac{1}{12}(3x^2y+y^3)Q_2''(z) \\
 & + \frac{1}{2}(x^2-y^2)P_3(z) + xyQ_3(z) \\
 & - \frac{1}{6}(x^3-3xy^2)P_4(z) + \frac{1}{6}(y^3-3x^2y)Q_4(z) ,
 \end{aligned} \tag{6.32}$$

$$\begin{aligned}
 B_y(x,y,z) = & -\frac{y}{2}B'(z) + \frac{y}{16}(x^2+y^2)B'''(z) \\
 & +Q_1(z) - \frac{1}{8}(3y^2+x^2)Q_1''(z) - \frac{1}{4}xyP_1''(z) \\
 & -xQ_2(z) + yP_2(z) - \frac{1}{6}y^3P_2''(z) + \frac{1}{12}(x^3+3xy^2)Q_2''(z) \\
 & + \frac{1}{2}(x^2-y^2)Q_3(z) - xyP_3(z) \\
 & - \frac{1}{6}(x^3-3xy^2)Q_4(z) - \frac{1}{6}(y^3-3x^2y)P_4(z) ,
 \end{aligned} \tag{6.33}$$

$$\begin{aligned}
 B_z(x,y,z) = & B(z) - \frac{1}{4}(x^2+y^2)B''(z) \\
 & +xP_1'(z) + yQ_1'(z) - \frac{1}{8}(x^2+y^2)(xP_1'''(z) + yQ_1'''(z)) \\
 & - \frac{1}{2}(x^2-y^2)P_2'(z) - xyQ_2'(z) \\
 & + \frac{1}{6}(x^3-3xy^2)P_3'(z) - \frac{1}{6}(y^3-3x^2y)Q_3'(z) ,
 \end{aligned} \tag{6.34}$$

where the first line in each of the previous three equations denotes the rotational symmetric part of the field, the second line the dipole part, the third line the quadrupole part, etc. P and Q denote the orthogonal components of the various multipoles. By taking the partial derivatives of eq.(6.29) and eq.(6.30) with respect to x and y unto third

order and combining those equations in the appropriate way, we obtain for the multipole coefficients:

$$\begin{aligned}
 P_1 &= B_x, \\
 Q_1 &= B_y, \\
 P_2 &= \frac{1}{2} \frac{\partial B_y}{\partial y} - \frac{1}{2} \frac{\partial B_x}{\partial x}, \\
 Q_2 &= -\frac{\partial B_x}{\partial y}, \\
 P_3 &= \frac{1}{4} \frac{\partial^2 B_x}{\partial x^2} - \frac{3}{4} \frac{\partial^2 B_x}{\partial y^2}, \\
 Q_3 &= \frac{3}{4} \frac{\partial^2 B_y}{\partial x^2} - \frac{1}{4} \frac{\partial^2 B_y}{\partial y^2}, \\
 P_4 &= -\frac{1}{8} \frac{\partial^3 B_y}{\partial y^3} + \frac{3}{4} \frac{\partial^3 B_x}{\partial x \partial y^2} - \frac{1}{8} \frac{\partial^3 B_x}{\partial x^3}, \\
 Q_4 &= -\frac{1}{2} \frac{\partial^3 B_y}{\partial x^3} + \frac{1}{2} \frac{\partial^3 B_x}{\partial y^3}.
 \end{aligned} \tag{6.35}$$

It is implicitly assumed in eq.(6.35) that the partial derivatives are taken in $x=y=0$ and that both the axial multipole coefficients as well as the derivatives of B_x and B_y are a function of z .

In order to obtain the axial multipole coefficients of a one turn first order spiral (see section 6.2.2), we found the required partial derivatives of eq.(6.29) and eq.(6.30) using the computer algebra program MAPLE [Char 1988]. Using MAPLE, we were not able to find the solutions for the multipole coefficients of a first order spiral entirely analytical, but got the result by using numerical integration.

If we want to make a general description for the multipole coefficients of a first order spiral, we have to find the scaling rules. For a first order spiral with an orientation equivalent to the one displayed in Figure 6.10, we found, in lowest order, the e_1 -multipole coefficients to be proportional to $(\delta/D)^2$ and the e_2 -multipole coefficients to be proportional to (δ/D) . The e_1 - and e_2 -coefficients denote strength along orthogonal directions for the various multipoles. In the case of a dipole they simply are the x - and y -dipole components.

Table 6.3 Relative and absolute scaling factors for obtaining the real multipole coefficients from the unified coefficients of a one turn spiral. Quadrupole e_1 and e_2 denote orthogonal components.

	coefficient (M_i)	absolute scaling	m	relative scaling	k	relative scaling error
axial field	B	$1/D$	1	1	0	0
dipole X	P_1	$1/D$	1	$(\delta/D)^2$	2	$(\delta/D)^3$
dipole Y	Q_1	$1/D$	1	δ/D	1	$(\delta/D)^2$
quadrupole e_1	P_2	$1/D^2$	2	$(\delta/D)^2$	2	$(\delta/D)^3$
quadrupole e_2	Q_2	$1/D^2$	2	δ/D	1	$(\delta/D)^2$

Besides this proportional scaling, relative to the deviation from the central radius, there is of course a scaling factor which is proportional to the absolute size. For this we took the central diameter D as a scaling factor. D for the first order coefficients and D^2 for the second order coefficients. An overview of the scaling factors for the various multipole coefficients is in Table 6.3. If we scale the multipole coefficients as obtained by MAPLE with the reciprocal factors of Table 6.3, we get unified representations for the axial multipole coefficients of a one turn first order spiral. Those unified multipole coefficients are, unto quadrupole components, displayed in Figure 6.14. An estimate of the error that occurs because we do the relative scaling only in lowest order of (δ/D) , is given in Figure 6.15. It shows the next term in the series expansion. A similar procedure, omitted here, can be used to make a unified representation for the derivatives of the multipole coefficients. When obtaining the curves of Figure 6.14 and Figure 6.15, care has been taken that (δ/D) is sufficiently small, because the series expansion resulting in the proportionalities as given in Table 6.3, has been taken about $(\delta/D)=0$.

Because we had to obtain the curves numerically, we only have the solution available at discrete points. Therefore, we made a cubic spline interpolation between these points in order to enable evaluating the curves for all z/D .

Figure 6.14 Unified representation of the multipole components of a one turn first order spiral, oriented according to Figure 6.10.

Figure 6.15 Unified representation of the errors made in relative scaling (scaling of δ/D).

6.3.3 the field of a lens element using a multipole expansion

In a lens element we deal with two multi-turn spirals in a face to face geometry, separated in axial direction by a distance δz . So, the first step in obtaining the field of a lens element is to calculate the field of one multi-turn spiral. The axial coefficients of this multi-turn spiral can be derived from the unified one-turn coefficients as displayed in Figure 6.14, according to the following scheme:

$$M_i(z) = \frac{1}{N} \sum_{j=1}^N \frac{1}{D_j^m} \left(\frac{\delta}{D_j} \right)^k U_i(z/D_j) , \quad (6.36)$$

where M_i are the axial multipole coefficients of the multi-turn spiral and U_i are the corresponding unified one-turn coefficients displayed in Figure 6.14. Further, N denotes the number of turns and k and m are, for each coefficient, given in Table 6.3. Again, a similar procedure can be used to obtain the derivatives required in the series expansion. The algorithm just given, for obtaining multi-turn coefficients from tabulated one-turn coefficients, is only valid for a first order spiral (see section 6.2.2), because the relative scaling rules as well as the tabulated curves of Figure 6.14 are based on the assumption that a first order spiral is used. Eq.(6.36), together with eq.(6.32) to eq.(6.34) and the curves of Figure 6.14 enable to calculate the three dimensional field of a first order spiral unto quadrupole components. This choice for quadrupole components as the multipole of highest order is arbitrary and may be extended with higher order components if necessary.

Once we know the field of one of the spirals, the field of the other one can be added by means of a simple coordinate transformation as given in Table 6.4. This transformation is obtained by combining a rotation about the x-axis with a reversal of the current. Moreover, if we introduce the displacements δx , δy and δz for the two parts of the lens element with respect to each other, the three dimensional field of the lens element is given by

$$\begin{aligned} Bx_{le}(x,y,z) &= Bx_{sp}(x+\frac{1}{2}\delta x,y+\frac{1}{2}\delta y,z+\frac{1}{2}\delta z) - Bx_{sp}(x-\frac{1}{2}\delta x, -(y-\frac{1}{2}\delta y), -(z-\frac{1}{2}\delta z)) \;, \\ By_{le}(x,y,z) &= By_{sp}(x+\frac{1}{2}\delta x,y+\frac{1}{2}\delta y,z+\frac{1}{2}\delta z) + By_{sp}(x-\frac{1}{2}\delta x, -(y-\frac{1}{2}\delta y), -(z-\frac{1}{2}\delta z)) \;, \\ Bz_{le}(x,y,z) &= Bz_{sp}(x+\frac{1}{2}\delta x,y+\frac{1}{2}\delta y,z+\frac{1}{2}\delta z) + Bz_{sp}(x-\frac{1}{2}\delta x, -(y-\frac{1}{2}\delta y), -(z-\frac{1}{2}\delta z)) \;, \end{aligned}$$

(6.37)

where lens element components are marked with the subscript *le* and the components of the multi-turn spiral with *sp*.

Table 6.4 Transformation used to calculate the field components of a spiral with current in the opposite direction and rotated about the x-axis with respect to the already known spiral. The subscripts "1" and "2" denote the coordinates and field components of the already known spiral and the unknown spiral, respectively.

rotation about x-axis	reversing the current	overall transformation
$x_2 = x_1$ $y_2 = -y_1$ $z_2 = -z_1$	$x_2 = x_1$ $y_2 = y_1$ $z_2 = z_1$	$x_2 = x_1$ $y_2 = -y_1$ $z_2 = -z_1$
$Bx_2 = Bx_1$ $By_2 = -By_1$ $Bz_2 = -Bz_1$	$Bx_2 = -Bx_1$ $By_2 = -By_1$ $Bz_2 = -Bz_1$	$Bx_2 = -Bx_1$ $By_2 = By_1$ $Bz_2 = Bz_1$

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7 Design of a thin film stack pattern

7.1 Introduction

In this chapter, we work out the exact geometry of the lens element for which the theoretical background has been given in the chapter 6. This lens element was built in order to verify the theoretical concepts. As already indicated, the best way to obtain a lens element would be by in-situ stacking of two superconducting layers separated by an insulating layer. However, currently we lack a reliable technology for this and alternatively, we have chosen to stack two single thin films on their substrates in a face to face orientation. In this face to face orientation, we shall not be able to use an optical technique for the mutual alignment of the two spirals. Therefore, we have chosen a capacitive alignment technique which requires, in addition to the lens pattern, a set of capacitive electrodes on both substrates. In the face to face geometry, these electrodes will form a set of position sensitive capacitors that can be used to align the two patterns with respect to each other. Another advantage related with a capacitive method is that the technique facilitates monitoring of the alignment afterwards, e.g. during cool down of the lens element.

When designing a stack element, using the high T_c technology presently available, we must take the following technological limitations into account:

- the maximum area homogeneously deposited in laser ablation is about 1 cm^2 .
- the maximum thickness of an $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film on SrTiO_3 is about $0.5 \text{ }\mu\text{m}$.
- the largest substrate that fits in the laser ablation setup is $2 \times 2 \text{ cm}$.

We have chosen SrTiO_3 as substrate material, because the quality in terms of current density is very high for films grown on this substrate. Since we use DC current only, the poor high frequency properties of this material are irrelevant. We have chosen a substrate of $10 \times 19 \text{ mm}$, because standard tools are available for this size.

In section 7.2, we will introduce the basic principles related with capacitive position sensors and show a technique to measure capacitance with sufficient accuracy. This knowledge will be used, in section 7.3, to work out the exact geometry both of the lens spiral and the capacitive electrodes. In sub-section 7.3.4, we solve the problem how to

connect the innermost windings of both spirals with each other, without disturbing the rotational symmetry of the lens field. Finally, in section 7.4, we calculate the optical properties of the lens design as developed in previous sections. Focal properties, as well as spherical and chromatic aberration will be given. Besides, we calculate the effect of the most important parasitic aberrations arising from the non-rotational symmetry of the windings. Since rotational symmetric as well as non-rotational symmetric aberrations are involved, we obtained all properties by means of ray tracing.

7.2 Capacitive alignment

7.2.1 basic principles

In an ideal parallel plate capacitor, the capacitance is given by

$$C = \epsilon \frac{O}{d} , \quad (7.1)$$

where ϵ is the permittivity of the material filling the space in the capacitor, O the surface area of the plates and d the distance separating the plates. However, in case of

just two capacitor plates as shown in Figure 7.1(a), eq.(7.1) is only an approximation, due to fringes and parasitic capacitances. These unwanted effects can be suppressed for the greater part by using a guard ring enclosing one of the electrodes, combined with an enlargement of the other electrode, as shown in Figure 7.1(b). Now eq.(7.1) approximates the true capacitance with great accuracy, provided that the gap between the guard ring and the electrode is small compared to the distance between the capacitor plates. The correction formula in case of a gap between the guarded electrode and the guard ring is [Heerens 1975]:

$$C = C_0 \left(1 - \frac{s^2}{2\pi R_i d} \coth \left(\frac{\pi R_i}{d} \right) \right), \quad (7.2)$$

where R_i is the radius of the detector electrode, d the distance between the capacitor plates and s the width of the gap between the guard ring and the detector electrode (see Figure 7.1(b)).

For precise measurements on a capacitor with guard ring, a bridge circuit of the principle shown in Figure 7.2(a) can be used. We will call the electrodes connected to

the generator, voltage electrodes and the ones connected to the detector, detector electrodes. Figure 7.2(b), which is the equivalent circuit of Figure 7.2(a), shows the parasitic capacitances. The capacitances C_{p1} and C_{p2} between the leads to the voltage electrodes and earth, only form a load for the generator and thus will pull the voltage on the measured capacitors down but do not influence the zero of the bridge. The parasitic capacitance parallel to the detector, C_{p3} , does not influence the null of the bridge either, because in case of balance, there is no voltage across this capacitor. In the bridge circuit, there is on each of the capacitors a voltage $V \sin(2\pi f_1 t)$, which results in a current $I_D = 2\pi f_1 V (C_1 - C_2) \cos(2\pi f_1 t)$ through the detector. This detector current can be measured very accurately using a lock-in amplifier with the voltage signal used as a reference [Heerens 1986]. Accordingly, the minimum capacitance which can be detected, provided that the guarding is correct, can be as low as 10^{-6} pF.

A bridge circuit as introduced in the previous paragraph is well suited to be used in differential measurements, because in this case, the two capacitors of the differential sensor can be connected with the two sides of the bridge. The geometry of a differential capacitive sensor is introduced in the next section.

7.2.2 a differential capacitive position sensor

Using the principles of the previous section, it is possible to construct capacitor geometries suitable for sensing displacements. If the electrodes are used in a differential geometry, indicated in Figure 7.3, then a displacement of the upper electrode "3" over a distance Δx with respect to the lower electrodes "1" and "2" results in a capacitance difference between the capacitors "1-3"

Figure 7.3 Differential capacitor geometry, used to measure displacements Δx [Heerens 1986].

and "2-3" respectively. This difference is given by

$$\Delta C = 2 \epsilon_0 \epsilon_r \frac{l}{d} \Delta x , \quad (7.3)$$

where l , d and Δx are indicated in Figure 7.3. Eq.(7.3) is valid with a relative inaccuracy better than 10^{-6} for $b/2 - |\Delta x| > 5d$.

7.3 Geometry of the stack element

7.3.1 introduction

In this section we present a geometry which consists of a helix forming the lens, combined with capacitive electrodes for the alignment. This geometry has to fit on a 10×19 mm substrate. In normal lens design, one must try to find the best balance between optical quality and physical constraints. However, when designing the thin film test pattern, we can only take the physical constraints into account. Since we do not have a particular application in mind, the optical requirements cannot be used to indicate the best geometry. We will develop a pattern that can be obtained in practice and calculate the optical properties of this pattern afterwards in order to verify whether thin film lenses are a realistic option with the current level of technology.

7.3.2 helical pattern

The helical pattern used for the prototype lens is a first order spiral for reasons of simplicity and because we have the three dimensional field of this type of spiral available in terms of axial multipole coefficients (see chapter 6).

When we want to stack two patterned substrates in a face to face geometry, it must still be possible to have electrical contacts on both substrates. We thus cannot let the

substrates fully overlap, since we have to keep the edges of both substrates available for contacting. This situation is indicated in Figure 7.4. If we have a substrate of size $A \times B$ and we chose $a \times b$ for the contact area, it means that we have an area $(A-a) \times B$ available for the pattern. However, as we do not want the extent of the pattern too close to the rim, we take a margin b on all sides.

Figure 7.4 Two substrates in a face to face geometry with one edge available for contacting at each substrate.

This results in an area $V \times W$ which is available for the pattern.

In our case we have a substrate of 10×19 mm and say A is 10 mm and B is 19 mm. For a and b we have chosen 1.5 mm and 0.5 mm, respectively. We thus limited the maximum outer diameter of the spiral to V , which is 7.5 mm in this case.

We took 1 mm for the diameter of the central hole, because this was the smallest size available to us in the ultrasonic technique we applied for drilling. Ultrasonic drilling results in rough edges which made us decide to take a 0.1 mm margin around the hole. These considerations set the inner diameter of the spiral to 1.2 mm.

Once we have fixed the inner and outer diameter to 1.2 mm and 7.5 mm, respectively, the only free parameters left are the width of the spiral track and the winding pitch. We took a width and pitch of $20 \mu\text{m}$ and $25 \mu\text{m}$, respectively, because we have a relatively large pattern which must be completely error free. Therefore, we can push the size of the details not too close to the resolution attainable with a light optical patterning technique. As a result, we have a spiral of 125 turns.

7.3.3 capacitive electrodes

If we position the helix in the center of $V \times W$ (see Figure 7.4), we have, on both sides of the helix, an area available for the capacitive electrodes. The electrode pattern must fulfill the following constraints:

- all electrodes must have their bonding pads in the area $a \times W$ (see Figure 7.4), which is the part that does not overlap with the opposite side substrate.
- the electrodes must form capacitive position sensors if the two substrates are in a face to face orientation.
- the geometry of the capacitive electrodes must be identical on both substrates, as we intend to use only one mask for patterning.

Since we require identical electrode geometries, there will always be an even number of position sensors, because on each substrate we need at least one voltage and one detector electrode which from two sensors in the face to face geometry. Therefore, if we intend to measure displacements on both sides of the helix, the face to face geometry contains at least four sensor regions.

Figure 7.5 (a) Detector- and voltage electrodes of the position sensor.
 (b) Complete sensor obtained by combining the geometry indicated in (a) with an identical geometry rotated about the line AA.

The layout of one of the electrode pairs, as to be placed on both sides of the helix, is indicated in Figure 7.5(a). This geometry consists of four voltage electrodes and one

detector electrode which will belong to different sensors in the face to face setup. This face to face setup is indicated in Figure 7.5(b) and is obtained by combining the electrode geometry of Figure 7.5(a) with an identical geometry rotated about the line A-A. In Figure 7.5(b), the two individual sensors appear. The geometry consists of two differential displacement sensors, using the principle shown in Figure 7.3, in a mutually perpendicular setup. The electrode arrangement allows, at each of the regions, detection of displacements along four different directions. Those are listed in Table 7.1 and indicated in Figure 7.5(b). Moreover, measuring the total capacitance of the electrodes "1-4" with respect to the detector provides information about the distance between the two substrates.

Table 7.1 Electrode configurations to be combined in order to detect displacements along four different directions. The electrode numbers and directions correspond to those indicated in Figure 7.5(b).

electrode numbers belonging to the first electrode	electrode numbers belonging to the second electrode	direction
1	3	II
2	4	IV
1 and 2	3 and 4	III
1 and 4	2 and 3	I

If we put the electrode pattern of Figure 7.5(a) on both sides of the lens spiral, we obtain a geometry as indicated in Figure 7.6(a). Figure 7.6(b) shows the stacked setup, obtained by combining two samples of the geometry indicated in Figure 7.6(a) in the

proper way. Figure 7.6(b) shows the four capacitive sensor areas which enable alignment of the two substrates with respect to each other in all six degrees of freedom. In Figure 7.6(b), the pattern on the upper substrate is drawn with a dashed line.

The dimensions of the voltage and detector electrodes have been chosen as large as possible in order to obtain easily detectable signals. We took $s=400\text{ }\mu\text{m}$ and $t=1000\text{ }\mu\text{m}$ (see Figure 7.5(b)), resulting in a capacitance between each voltage electrode and the detector of $5.7\cdot 10^{-18}/d$ Farad, where d is the distance between the voltage- and the detector electrodes. In practice d will be between 1 and $10\text{ }\mu\text{m}$ and the capacitance will thus be 5 to 0.5 pF, respectively. If we apply eq.(7.3), substitute the actual width of a voltage electrode, $l=5.7\cdot 10^{-5}\text{ m}$, and take $d=10\text{ }\mu\text{m}$ as the distance between the plates, we obtain $\Delta C=10^{-10}\cdot\Delta x$ (ΔC in F, Δx in m). So, for a position accuracy of $1\text{ }\mu\text{m}$, we have to measure a change in capacitance of 10^{-4} pF which is far above the detection limit of 10^{-6} pF . Reasons for choosing an alignment accuracy of $1\text{ }\mu\text{m}$ are given in the final section of this chapter.

As already indicated, the use of guarding is of capital importance for correct performance of the sensor. Accordingly, we surrounded the detector electrodes as well as the voltage electrodes with guard material which can be grounded by connecting the bonding pads 3 and 11 (see Figure 7.6(a)) with earth. We separated the two grounded areas from each other in order to avoid a closed loop in the earth circuit. In section 7.2.1, it has been concluded that only guarding of the detector is necessary. The reason for guarding the voltage electrodes as well is the fact that our geometry does not resemble the one shown in Figure 7.1(b), where the voltage electrode is far larger than the detector. In our setup the detector and voltage electrodes have approximately equal dimensions and we thus need to define a potential in the direct vicinity of the voltage electrodes in order to demarcate its extent which would, in case of floating surroundings, be inadequately specified.

7.3.4 connection between adjacent layers

The principle of a lens element is based on two spirals in a face to face geometry with their innermost turns connected. This connection cannot be superconducting, as this requires manufacturing of superconducting multilayers, a technique not mastered yet. Moreover, the contact must be symmetrical about the line of rotation, because we want an identical upper and lower geometry in the lens element which enables to use the same mask for both patterns. The layout of the innermost turn is indicated in Figure 7.7(a). Because of symmetry requirements, this innermost turn is circular. Note that the fields arising from the currents in the radial parts cancel as long as the two halves of the lens element are sufficiently close together.

To avoid a short circuit between the two spirals as well as a short circuit in the capacitive electrodes, we need to deposit an insulating layer covering the complete pattern except for the contacts and the innermost turn.

A contact between the innermost turns of the spirals on the two substrates can now be achieved by depositing a metal film on the inner turn. This film must have a thickness larger than the insulating layer. If we do this deposition for both parts of the lens element, we are able to make the desired connection by clamping the two substrates together in the face to face geometry. In order to enlarge the area of the contact, we have extended the ring with radial tracks (Figure 7.8(a)) which are on top of the insulating layer. The function of those tracks is to expand the contact area without

disturbing the rotational symmetry of the field. This is shown in Figure 7.8(b). The figure indicates the situation in case a contact between two radial tracks occurs at a certain position p . The current flow is directed from the center in the lower track and directed towards the center in the upper track, but, as the two substrates are clamped together, d is very small, so the effective disturbing field is negligible. It is not important exactly where the contacts between the upper and lower pattern occur. They can be either directly between the inner rings of both patterns or at one or several radial tracks. This geometry assures that currents in the contact do not give rise to a parasitic magnetic field.

Figure 7.9 shows a plane view and a cross section of the substrate near the center of the spiral. It can be seen that the thickness of the insulating layer must be larger than that of the superconducting layer in order to prevent a short circuit between the windings, caused by the metal top layer. This top layer must also have a thickness larger than the superconducting film, in order to ensure a reliable contact in radial direction at those positions where steps in the film occur (see Figure 7.9), such as from the inner ring to the radial track.

An overview of the complete pattern in exploded view is in Figure 7.10.

Figure 7.10 Complete pattern consisting of the superconducting film, the intermediate insulating layer and the metal top layer [Adriaanse 1991a,b].

7.4 Optical properties

7.4.1 introduction

To determine the optical properties of the lens element, we calculated the field distribution employing the axial multipole method outlined in the previous chapter. Next, we used ray tracing to determine the focal properties, the geometrical- and chromatic aberration and the parasitic aberrations. The latter are caused by the non-rotational symmetry of the windings.

The focal properties as well as the geometrical and parasitic aberrations were obtained by tracing 17 rays approaching the lens nearly parallel to the axis. Those ray positions are indicated in Figure 7.11 in a plane in front of the lens. The rays 2-9 enter the lens at a radius r , whereas the rays 10-17 enter at a radius $2r$. This facilitates computation of

the spherical aberration as indicated in Figure 7.13. Definitions of the parameters that determine the focal properties are given in Figure 7.12.

Because of the dipole component in the field, the axis of the lens element is not straight. Therefore, we used ray number 1 (Figure 7.11) to determine the displacement and calculated all properties with ray 1 acting as the axis of reference.

We found the spherical aberration by determining the mean distance between the axial crossings of the rays 2-9 and 10-17. This distance, shown in Figure 7.13, is $\Delta z_{Cs} = 3 C_s \alpha^2$. Accordingly, $C_s = \Delta z_{Cs} / 3 \alpha^2$. The axial astigmatism, Δz_f , was estimated just by determining the spread in the crossover positions of the rays 2-9 or 10-17.

Finally, we found the chromatic aberration by tracing a third set of rays, representing a slightly higher energy, and used the shift of the focal plane with respect to the previous traces as an estimate for $C_c \cdot \Delta V / V$. Here V denotes the beam energy in electron volts and ΔV the energy shift with respect to the previous traces. We also determined the chromatic effect on the displacement of the axis (ray number 1). This aberration is due to the dipole component in the field in combination with an energy spread.

7.4.2 field distribution

The axial multipole components of the multi-turn spiral can be obtained by application of eq.(6.36), thereby using the curves of Figure 6.14. The parameters for the multi-turn spiral are:

- inner diameter: 1.2 mm
- outer diameter: 7.5 mm
- number of turns: 125
- spacing between the turns: 25 μm

We did not take the width of the track into account, as, according to eq.(6.23), corrections to the spiral track are less than 30 nm. To find the coefficients for the complete spiral, we may use eq.(6.37), where the actual separation between the two components of the lens element is $dz=2.5 \mu\text{m}$ and $dx=dy=0$. However, we did not follow the procedure of eq.(6.37), because we effectively have to calculate the field at two points in that case. Since we do not use the displacements dx and dy here, the field of the complete lens element may equally well be obtained by a proper combination of the axial multipole coefficients only. These coefficients are a function of z and we can, therefore, easily incorporate the displacement dz by means of a coordinate transformation.

The multipole coefficients for both the single- and the double spiral (=lens element) are given in Figure 7.14. In order to obtain the complete field, we made the series expansions eqs.(6.32) to (6.34). This field serves as an input for the ray tracing which is employed for evaluating the optical properties.

Figure 7.14 Axial multipole coefficients of the 125 turns spiral with an inner diameter of 1.2 mm, an outer diameter of 7.5 mm and a winding pitch of 25 μm .

- (a) Axial multipole coefficients of one 125 turns spiral.
(b) Axial multipole coefficients of the complete lens element consisting of two spirals as indicated in (a) in a face to face orientation, separated in axial direction by $dz = 2.5 \mu\text{m}$.

7.4.3 first order properties

The focal properties of the lens element for both projector and objective mode are given in Figure 7.15 and Figure 7.16 as a function of the lens excitation $NI/\sqrt{V}$. The parameters f_{obj} , f_{proj} , zf_{obj} and zf_{proj} are defined in Figure 7.12.

7.4.4 spherical- and chromatic aberration

The spherical aberration was calculated according to the method outlined in section 7.4.1. The results are given in Figure 7.17, as a function of the lens excitation. The chromatic aberration is shown in Figure 7.18.

Figure 7.17 Spherical aberration for the projector- and objective mode, as a function of the lens excitation.

Figure 7.18 Chromatic aberration for the projector- and objective mode, as a function of the lens excitation.

7.4.5 parasitic aberrations

As already indicated in section 7.4.1, due to the non-rotational symmetry of the windings, parasitic aberrations arise. Figure 7.19 shows the angle between the axial ray (ray number 1 in Figure 7.11) and a straight axis in the objective focal plane and at the end of the lens field. Figure 7.20 shows the displacement of the axial ray with respect to a straight axis, at the position of the objective focal plane and at the projector focal plane. Figure 7.21 shows the axial astigmatism as a function of the lens excitation.

Due to the dipole component in the field, in combination with the energy spread in the beam, we have an extra chromatic effect analogous to the chromatic error that occurs in deflectors. The effect of energy spread on the angle between the axial ray and a straight axis is indicated in Figure 7.22 and axis displacement due to energy spread is shown in Figure 7.23.

Figure 7.20 Distance between the axial ray (ray number 1 in Figure 7.11) and a straight axis at the objective focal plane and at the projector focal plane.

Figure 7.21 Axial astigmatism at the objective- and projector focal plane as a function of the lens excitation.

For estimating the effect of a misalignment between the two parts of the lens element with respect to each other, we calculated the multipole coefficients that arise when two identical *circular* current loops are displaced over dx , dy , dz . This is a simplification in that we only take the rotational symmetric part of the field distribution into account and ignore the multipole terms, which, in case of a misalignment, also contribute to some extra parasitic aberrations. However, a calculation in which we took these terms into account showed that the simplified result is good enough to be used as an estimate for the required alignment accuracy. The multipole coefficients thus obtained, given in terms of unified quantities, are shown in Figure 7.24. The figure displays the curves as obtained in lowest order in dx , dy and dz and only exhibits multipole terms unto quadrupole components.

We want to obtain a rough estimate for the accuracy needed in the capacitive alignment procedure. We therefore simply compared the magnitude of the multipole components that arise from a misalignment with those that are due to the non-rotational symmetry of the windings. The advantage of this procedure is that it does not require any ray tracing.

The magnitude of the dipole field (P_1 and Q_1) that arises from a misalignment, is obtained from Figure 7.24 and reads

$$\begin{aligned} P_1 &= P_{1u} \cdot \frac{1}{D} \frac{dx dz}{D^2} , \\ Q_1 &= Q_{1u} \cdot \frac{1}{D} \frac{dy dz}{D^2} , \end{aligned} \quad (7.4)$$

where P_{1u} and Q_{1u} denote the unified coefficients shown in Figure 7.24, D is the diameter of the turns and dx , dy and dz are the misalignments along the x, y and z direction, respectively. If we only look at the peak values of the unified coefficients, we see $P_{1u} = Q_{1u} \approx 2 \cdot 10^{-6}$ Tm/A. We take $D = 10^{-3}$ m, which is roughly equal to the inner diameter of the spiral and we know that $dz = 2.5 \cdot 10^{-6}$ m. Substituting those values in eq.(7.4) gives, $P_1 = 5 \cdot 10^{-3} dx$ T/A and $Q_1 = 5 \cdot 10^{-3} dy$ T/A. In Figure 7.14, we see that Q_1 of the lens element has a peak value of $2.5 \cdot 10^{-9}$ T/A and if we equate this with the dipole components arising from a misalignment, we get $dy = 0.5 \mu\text{m}$. A similar procedure holds for the quadrupole components P_2 and Q_2 and it results in nearly the same answer for the accuracy required for the alignment.

We have taken the inner diameter of the spiral as a reference for D and thus obtained results that apply for the innermost turns. For the outer turns, we may accept more misalignment and the overall value will be some weighted average. Just to be on the safe side, we have chosen a maximum misalignment along x and y of $1 \mu\text{m}$.

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8 Experimental work on a prototype lens element

This chapter reports on the experimental methods applied for fabrication of a thin film stack device and on the results obtained with these methods. Results obtained with the capacitive alignment technique are also given.

The method utilized for fabrication is presented in section 8.1. We tested this fabrication process on Si substrates carrying a vapor deposited Aluminum film, in order not to waste the much more expensive SrTiO_3 substrates during the numerous experiments. Except for the substrate and the basic film, all layers and fabrication steps carried out on the test device were identical to those required for the real device. Therefore, we could also employ the test samples to examine the capacitive alignment procedure, since this method is virtually not depending on the resistivity of the film in which the capacitive electrodes are made. This alignment test is described in section 8.2. In order to be able to do the alignment with sufficient accuracy, we developed a special tool, the *alignment unit*, also presented in section 8.2. With the alignment unit, in combination with the capacitive sensors, we achieved a minimum displacement below $0.1\text{ }\mu\text{m}$ and an overall misalignment less than $0.5\text{ }\mu\text{m}$.

Finally, in section 8.3, we report on the results obtained with the real superconducting devices. A current-voltage characteristic of a complete 125 turns spiral made in an $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film is presented. Unfortunately, we did not perform measurements on a stack of two superconducting samples. This would have been an interesting experiment in order to study the influence of the non-superconducting intermediate contact between the innermost turns of the two spirals.

8.1 Fabrication of the lens element

As indicated several times, patterning of a high- T_c superconducting film with a fine pitch spiral covering a relatively large area is a difficult job due to inhomogeneities present in most films. Therefore, we first tested the stack element technique using well known methods available for patterning of aluminum films on silicon.

We intend to cover the superconducting track with a silver toplayer which serves as an alternative path for the current at bad spots in the film and also to protect the superconductor during patterning. We thus have to work with a lift-off process. This need for a lift-off process can be understood if we realize that patterning of an $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film with a silver top layer cannot be done in one step, using one etchant, because, as the etchant required for silver is much too strong for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$, we would remove most of the $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ immediately after we etch through the silver. If a lift-off process is employed, we can make the negative pattern in photoresist on top of the superconducting film, followed by a deposition of silver. Patterning of the silver is now simply done with lift-off and as the silver is hardly affected by the etchant for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$, we can use this silver top layer as a stencil mask for patterning of the $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$. In order to test this lift-off process, we patterned the aluminum film on the silicon substrate by means of this method.

As outlined in the previous chapter, a lens element is made from two parts. Each part is based on a substrate carrying a superconducting film that is patterned with a spiral and capacitive electrodes. On top of this superconducting film are an intermediate insulating layer and a metal top layer, where the latter is to be used as a connection to the opposite spiral in the face to face geometry. For patterning, we use a mask technique. Three masks were made on two separate quartz plates by means of e-beam lithography. On forehand, all masks were meant to be used in combination with positive photoresist. We planned to pattern the first layer by means of etching and to employ lift-off for patterning the second and third layers, but, as we later on decided to pattern the first layer with lift-off as well, we had to employ negative photoresist here.

The layers that make up the device are:

- (1) 500 nm $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ on a 10×19 mm (100) SrTiO_3 substrate
- (2) 2 nm Cr + 200 nm Ag
- (3) 800 nm Al_2O_3
- (4) 900 nm Ag + 100 nm Au

Instead of the layers (1) and (2), we used 700 nm Al on a silicon substrate for the test pattern. As already indicated in the previous chapter, the thicknesses of both the insulating layer (3) and the metal top layer (4) must be larger than those of the lens

spiral. A relatively thick Al_2O_3 layer is not only required to avoid short circuits between the windings of the spiral in the plane of the substrate, caused by the radial tracks of the upper pattern, but also to avoid pinholes that might result in short circuits between the spiral and the radial tracks. The pinhole based types of short circuits are oriented perpendicular to the plane of the substrate.

A complete schematic overview of the fabrication process is given in Figure 8.1. A detailed description of the recipe for fabrication of the devices is given in appendix A. Figure 8.1 shows a cross section through the device after each process step. Those cross section views are numbered from 1 to 16. Note that step number six is only required for patterning of the superconductor and that this step can be omitted when fabricating the test device on silicon. The pattern overviews, labelled with the Greek numbers I to III, show the status of the device after all process steps required for one specific layer have been completed.

Figure 8.1 Fabrication steps for one part of the lens element. The sub figures with numbers 1-16 are the device cross sections as they appear after the various processing steps. The three pattern overviews with numbers I to III give a topview of the complete sample after completing all steps required for a specific layer.

The results obtained with a 700 nm Al film on silicon are indicated in Figure 8.2 to Figure 8.4.

8.2 The capacitive alignment procedure in practice

The alignment procedure, using the integrated capacitive electrodes, was examined with the test devices made on silicon. To obtain a correct alignment, we not only need accurate position sensors, but we must be able to manipulate the two parts to within the required alignment accuracy as well. Therefore, we developed a special tool to do the alignment which we will call *alignment unit*. The alignment unit is based on two leaf-spring suspended tables in a frame, where each table rests on three adjustable supports. This enables translations in the z-direction and rotations about the x- and y-axis. The function of the leaf springs is to prevent translations and rotations of the table in the xy-plane. In order to have all six degrees of freedom under control, plates can be mounted on each of the tables. These plates are manipulated at three points and bring the other degrees of freedom under control, which are translations in the xy-plane and rotations about the z-axis. Figure 8.5 gives a schematic overview of the alignment unit and Figure 8.6 is a photograph of the lower part of the unit. Note the leaf-spring suspended table with the plate bearing the sample. The sample holders, described below, were not yet mounted when the photograph was taken. The alignment unit contains two tables, since we have both coarse and fine manipulators available for all degrees of freedom. We need these two sets of manipulators, since the range of the fine manipulators is insufficient for correcting the initial misalignment, resulting from pre-positioning. The fine alignment, on the other hand, is too subtle to be done with the coarse manipulators. These coarse manipulators are just simple micrometer screws giving a displacement of 0.5 mm per revolution and the fine manipulators, which employ these screws in combination with a reduction mechanism, give a 5 μm displacement for each revolution.

Figure 8.8 Photograph of the silicon samples mounted in the holders. Note the rim present on the biggest holder (lower holder).

As the final goal of the alignment is to fix the two parts of the lens element with respect to each other, using epoxy resin, we need special holders on which we can mount the samples and that allow filling with epoxy when the alignment has been completed. Moreover, the stacked substrates, including holders, should be removed from the alignment unit without any difficulty after curing the epoxy. Figure 8.7 shows the

principle of the holders. The upper part of the figure is a top view of the samples in a face to face position, mounted in the holders. The part beneath is a cross-section and clearly displays the rim present on the lower holder, needed to create some sort of box that allows filling with epoxy resin. Figure 8.8 is a photograph of the silicon samples mounted on the holders.

A disadvantage in using flatcable for all leads is that the leads to the detector are unshielded in the region between the sample and the flatcable connector. We use this flatcable connector for attaching the voltage- and ground leads only. In the same region we have two coaxial connectors available for the detector leads. The electronics we used are based on the principles outlined in the previous chapter and were available to us in a suitable form without any modifications needed. We only had to build a set of switches which serve to select a certain capacitor geometry.

The smallest displacement that has been observed in practice, for the case where both parts of the lens element are close together, is below $0.1\text{ }\mu\text{m}$. Because of the large number of capacitive electrodes, we can employ several electrode subsets to do the alignment along a certain direction. As a result, we can check this alignment using another subset of electrodes. The error we found, as checked with an alternative set of electrodes, after alignment along a certain direction was within $0.5\text{ }\mu\text{m}$. This result is well within the margin allowed for misalignment. In the previous chapter, we found that a maximum misalignment of $1\text{ }\mu\text{m}$ is acceptable.

After the alignment, we fixed the two sample holders by filling the lower holder with the epoxy resin Stycast 1266. This resin, when used in combination with Catalyst 24 LV, needs a 24 hours curing time at room temperature. We employed room temperature curing in order to minimize the stress induced by the low operating temperature of the device. We monitored the misalignment of the two parts while curing and found that the displacement during the 24 hours curing time was less than $0.1\text{ }\mu\text{m}$.

We estimated the power dissipation in the intermediate contact. For a worst case situation, where there is only one contact between two tracks at the outermost radial point, we estimate the resistance of the intermediate contact to be $250\text{ }\mu\Omega$. This results in a power dissipation of $250\text{ }\mu\text{W}$ at a current of 1 A , which is the maximum current that can be obtained in our device with present high- T_c films.

8.3 The prototype lens element using $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ thin films

For testing the lens element principles in a superconducting film, we had 10×19 mm SrTiO_3 substrates available. On those substrates, $0.5 \mu\text{m}$ $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ was deposited by means of laser ablation. We applied the complete procedure depicted in Figure 8.1 to make the device, which includes patterning of the silver toplayer using lift-off and employing this patterned silver layer as a stencil mask for etching the $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$. This etching is an additional step in comparison with the test element on silicon, described in section 8.1. We have had problems with the etching due to grains present in laser ablated films. These grains tend to pierce the silver and are thus exposed to the etchant which results in holes in the $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film at those particular grain positions. Another problem related with the grains is that they sometimes shortcircuit two windings. To overcome these problems, it is necessary to obtain a superconducting film that does not hold any grains with a size over $2 \mu\text{m}$ or any other large imperfections in the region of the spiral, which is a disk of 7.5 mm diameter. This area is relatively large in comparison with other patterns made in high- T_c films and although our pattern does not hold small details, the stringent condition that the spiral must be without interruptions over the entire length results in strict requirements on the superconducting film and the patterning process.

We measured the current-voltage characteristic of a 125 turns spiral made in a $0.5 \mu\text{m}$ thick laser ablated $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film by a two probe method at a temperature of 43 K. This result is shown in Figure 8.9 where the circles indicate the measured points. We made a least square fit to the first twenty points which is indicated with a solid line. The points marked with crosses indicate the difference between this fit and the measured points. From Figure 8.9, we estimated the critical current density to be $1.65 \cdot 10^6 \text{ A/cm}^2$ at 43 K. Although this current density is below the values reported in literature (e.g. $J_c(43 \text{ K}) > 10^7 \text{ A/cm}^2$ [Adrian 1991]) the result is not bad if one takes the total length of the track into account, which is about 2 meters.

Figure 8.9 Induced voltage versus applied current for a 125 turns spiral made in a $0.5 \mu\text{m}$ $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film, measured at 43 K.

The circles indicate the measured data points and the solid line is a least square fit to the first twenty points. The lower line, marked with crosses, has been obtained by subtracting the fit from the measured data and was added in order to clearly indicate the point of maximum current density.

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9 Conclusions and future perspectives of high- T_c superconductors in particle optics

9.1 The present state of high- T_c superconductors with respect to applications in particle optics

The main goal of this thesis has been to describe the present possibilities with respect to the applicability of high- T_c superconductors in particle optics. We have mainly been concentrating on high- T_c thin films utilized in magnetic lenses. Thin films, because of their superior properties in comparison with bulk materials and because they can be patterned accurately. The reason to apply those films for magnetic lenses is the fact that the pattern required for lenses is rather straightforward and that a lens is a basic optical element.

The present state of the art of high- T_c materials is as follows. Thin films which exhibit a current density over 10^6 A/cm² at 77 K in an applied field of several Tesla's are available. At liquid helium temperature, the attainable current density is even one order of magnitude higher and patterning of the films without deterioration of the properties can now routinely be done using a chemical etchant. The main problem of those films with respect to their applicability for magnetic lenses is that, in spite of the high current density, the number of ampere turns which can be generated with a single film is small. This is due to the maximum film thickness of about 0.5 μ m. However, it is to be expected that better substrates will allow larger thicknesses, which increases the number of ampere turns in the device accordingly. With state-of-the-art current densities we have, for a pancake coil of 1 cm outer diameter, 50 Ampere turns/ μ m at 77 K and about 500 Ampere turns/ μ m at 4.2 K. Consequently, if we use only two thin films for making a magnetic lens, we thus require a thickness of several micrometers per film in order to obtain the lens excitation necessary for most applications.

In addition to a larger film thickness, the lens strength can also be increased by stacking of thin film lens elements. Stacking of lens elements, including substrates, is not very desirable though, because, due to the space occupied by the substrates, the effective current density goes down dramatically. The best alternative would be to use multilayers,

but the technology for making those is in a preliminary state. No breakthrough with respect to applications in particle optics may be expected in the near future. Moreover, present multilayer technology mainly aims at three layer structures, the intermediate insulating layer included, which is another reason for not too high running expectations. As far as thin films are concerned, we think that the single lens element structure as presented in this work is the most promising one, especially if the maximum film thickness can be increased to a few micrometers. Another future perspective is offered by the current density, which is not at its fundamental limit yet. The critical current density may be further enhanced through a better understanding of the mechanism responsible for flux pinning and to utilize this knowledge by for example creating pinning centers in an artificial way.

Concerning the bulk materials, there also has been major progress during the past four years. High- T_c wires in silver tubes now exhibit current densities over 10^5 A/cm² at liquid helium temperature [Kase 1991] and their sensitive behaviour to external magnetic fields has been improved impressively, which offers perspectives for coil applications. In fact, the first high- T_c bulk coils with a reasonable current density have already been made. In a triple pancake coil, made from a Bismuth based high- T_c superconductor, Shibuta [1991] obtained a current density of $4 \cdot 10^3$ A/cm² at 77 K and about 10^4 A/cm² at 4.2 K, both in an external field of 23 Tesla. Another coil application has been reported by Shimoyama [1992] who obtained a maximum current density of $2.5 \cdot 10^4$ A/cm² at 4.2 K in a 12 Tesla external field. The coil, with a total length of 6 m, has an inner diameter of 13 mm, an outer diameter of 45 mm and generated a self field of 0.6 Tesla. The corresponding number of ampere turns was close to 15,000. These results strongly indicate that the bulk materials have come to a level close to that required for applications in electron optics.

A problem related with operating at a high current is how to keep the contact resistance between the normal conductor and the superconductor acceptably low. The dissipation in the contact may be decreased for thin film lenses, as well as for bulk coils, by increasing the number of *turns* at a constant number of *ampere-turns*, but this option is limited by technological considerations such as the inability to fabricate a completely error free coil having a large number of small pitch windings. Better prospects are

offered by an enlargement of the contact area and research for lower contact resistances.

Another, more basic application, of bulk material is shielding. For the ambient field, shielding factors as high as 10^8 have been obtained with sintered high- T_c materials [Zavaritsky 1989]. However, the use of diamagnetic shielding for shaping lens fields is not to be expected, since the lower critical field of the high- T_c materials is far below the field strengths needed in lenses. This lower critical field is the upper limit for applications related with shielding. An interesting application in electron optics, for both bulk and thin film materials, might arise if non flat surfaces can be coated, because this allows three dimensional structures to be made. In this respect, bulk superconductors probably have the highest potential.

As for the temperature where potential devices have to be operated, we think liquid nitrogen is the best choice for simple applications, although, for devices which are more demanding in terms of current density or critical field, we consider the temperatures offered by the closed cycle cryo coolers to be more appropriate. Those cooling machines operate in the 10 K to 40 K range where the current densities are nearly an order of magnitude above those at 77 K. For ultimate performance, liquid helium is of course still the best choice, but the applications of the past learned us that the use of a helium cryostat severely deteriorates the ease of operation. In fact, it has been one of the main reasons for the lack of success of classical superconductors in particle optics. Therefore, cooling must not be too complicated or occupy too much space, nor may it introduce vibrations that affect the resolution. Especially with respect to the first two requirements, we think that cryo cooler temperatures are the most appropriate. The vibrations introduced by those machines are a major problem though, because the plunger in most coolers runs at a two Hertz cycle which is difficult to eliminate. However, new developments in the field of cryo coolers take place and machines running at a fifty Hertz cycle rate are now commercially available.

During the first years of high- T_c superconductivity, superconductor properties were very sensitive to moisture and tended to degrade in time. Today, most of those problems have disappeared due to better quality of the high- T_c materials which are now stable enough to operate in applications for at least a few years.

Concluding this review about the present state in high- T_c superconductivity, it is clear that the present standard is not yet sufficient for profound applications in particle optics. The trend is however unmistakably towards possible applications in the not too distant future. A severe limitation for research on the applicability of high- T_c superconductors in particle optics now is that these materials are not commercially available. This makes experimental work only meaningful for research groups that have close connections with colleges who operate in the field of superconductivity, or more specifically, who own fabrication facilities for high- T_c superconductors.

9.2 Potential applications

One of the first applications for high- T_c materials may be shielding of the ambient field in regions of low beam energy. Most other potential applications should, in our opinion, take advantage of the high critical current density of the ceramic superconductors. For the ambient field, a shielding factor of 10^8 can be obtained with sintered high- T_c material [Zavaritsky 1989]. This value is very high in comparison to metals with a high permeability (μ -metals) which have an effective shielding factor of 10^5 when used under most favorable conditions [Boll 1990].

An elaborate application would be to use high- T_c thin films for magnetic lenses in electron microscopy. When aiming at an objective lens which has to be an improved version of its conventional counterpart at 300 kV, we should mainly concentrate on spherical aberration. The spherical aberration of any pancake lens as given by eq.(6.10) is, for present current densities, dominated by the first term. This basic C_s is independent of the lens size and reads:

$$C_{s_{\min}} = C_1 \frac{2k\sqrt{V}}{\eta JS} , \quad (9.1)$$

where C_1 is constant, k is the lens excitation, V the beam energy, η the coil fill factor, J the current density and S the axial length of the coil. Substituting 0.028 for C_1 (see

Figure 6.7), 25 for k and 1 for η , we have, at a beam voltage of 300 kV and a current density of 10^7 A/cm², a basic C_s of 10^{-8} /S meters. In other words, for a superconducting film of one micrometer thickness we have a basic C_s of 10 mm. An effective film thickness of at least 10 μ m is therefore needed in order to have a performance comparable with a conventional objective lens at 300 kV. An advantage related with iron-free pancake coils is their size reduction with respect to the iron circuit lens.

The Variable Axis Immersion Lens (VAIL), as used in e-beam lithography machines is another possible application. By joining two thin film lens elements, thereby shifting the optical axis of the one with respect to the other, a VAIL could be obtained in a simple way. The optical axis of the assembly can be shifted by changing the relative excitations of the two coils. The lens could be extended with two slightly elliptical lens elements in a mutual perpendicular orientation, which enable beam stigmatism. A constraint for this type of application is, that the four stacked lens elements have to be mounted sufficiently close together in order to form just one single optical element. This requires either a well developed multilayer technology or very thin substrates.

Another application that requires mastering of the technique for stacking several lens elements is shaping the (axial) field distribution by adding the fields of several pancake coils. An early implementation of this method, using classical superconductors, was proposed by Merli [1970] (see section 2.2.1). The device could be further refined if the local winding density in the thin film coils is optimized with respect to the shape of the field, or in other words, is optimized with respect to the optical requirements.

When aiming at some kind of hybrid device that combines conventional technology with high- T_c superconductivity, cooling of the superconducting device has to be taken into account. This may be a severe complication of the mechanical design. A hybrid device could for example be a VAIL with superconducting deflection coils, or an objective lens with a thin film lens element in the gap, which is able to improve the performance, or, when mounted eccentrically, is able to shift the optical axis in an alternative way. Another option is to put two elliptical lens elements, in a mutually perpendicular orientation in the gap of a magnetic lens to correct the astigmatism. This type of correction has the advantage over the conventional post-lens method, that the astigmatism is corrected in the origin and accordingly does not give rise to magnification

errors. However, it cannot be used in an objective lens, since the lens gap is already occupied by the sample.

An application related to hybrid devices are persistent current lenses which are excited by an external magnetic field that for example comes from a polepiece lens. Extremely stable lens fields can be obtained with those kind of superconducting lenses. One of the first tests of the persistent mode of high- T_c superconductors was performed by Fröhlingsdorf [1989], who obtained a current density of $0.9 \cdot 10^6$ A/cm² at 77 K in a 450 nm thick laser ablated ring. The ring had an outer and inner diameter of 9.5 mm and 6.5 mm, respectively. The critical current density did not depend on the presence of an external magnetic field of about 5 mT, which indicates that the magnetic flux lines are sufficiently pinned at this field strength in order not to decrease the current. If the superconductor is not able to carry the current necessary to take up the complete lens field, a stabilization can probably still be obtained by having a superconducting ring in the lens gap, because the ring tends to oppose changes in the field by means of the induced currents.

Quite another class of possible applications are the electron probe instruments. A small scanning column could for example be added to a machine working under vacuum, such as a Molecular Beam Epitaxy (MBE) apparatus, or an instrument for Auger depth profiling. This SEM column would enable in-situ monitoring of the process. For those SEM applications, the method of field shaping with several thin film coils could probably be applied.

An advantage of thin film lens elements is that they can rather simply be combined with capacitive electrodes, because those electrodes can be incorporated in the lens mask. This technique, worked out for capacitive alignment, has been demonstrated in this work. Besides using the position sensors for mutual alignment of the lens element parts, it may also be possible to apply those sensors for monitoring the position of the lens with respect to the rest of the column. Moreover, combining the sensor with an actuator such as a piezo may give rise to completely new features as automatic tilt and shift correction, or even enable dynamic corrections in scanning instruments.

The only lens application demonstrated with high- T_c superconductors to date, is the tube lens made from sintered material, as proposed by Matsuzawa [1989a-b, 1990,

1991a-c]. The principle, shown in Figure 9.1, is that a pulsed electron beam is focused upon injection into a superconducting tube, due to the field induced in the tube. However, the beam current of about 1000 Ampere, needed to induce a field of sufficient strength, is large. Apparently, those lenses are not

suitable for low current applications like microscopy or e-beam lithography. Proposed applications for the tube lenses are, among others, particle accelerators and free electron lasers.

If very strong magnetic fields can be generated with high- T_c materials, then applications in ion-optics might become feasible. However, a simple estimate learns that, for Hydrogen atoms, we need something like 10 to 50 Tesla to obtain focal properties comparable with those of magnetic electron lenses. The corresponding number of ampere turns is in the order of 10^5 . It is obvious here, that ferromagnetic materials cannot be applied for concentrating the field, but that high current density materials are required. To achieve those high lens fields, we need further development of the high- T_c materials, either to obtain a thick layer, or to increase the current density. At the moment, it is hard to foresee the time scale for these developments, but it will certainly exceed a few years.

How to improve upon the present properties of magnetic lenses? This question has been one of the background key issues throughout this work. It is known that polepiece lenses are already used to their limits set by the nature of the ferromagnetic materials. A slight improvement may be obtained if the saturation of the iron is used to optimize the field distribution of a certain design. This is now possible through the use of Finite Element software. However, no drastic improvement may be expected. Besides, the result only holds for one specific beam energy. The only alternative to obtain small field distributions with high peak values is to put more effort into the development of iron free high current density devices, and for those, high- T_c superconductors seem to be the best candidate.

Figure 9.1 Principle of the superconducting tube lens [Matsuzawa 1989a].

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Summary

This thesis presents the results of a study to the applicability of high- T_c superconductors in particle optics and introduces a possible technology for making magnetic lenses using $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ superconducting thin films.

In the nineteen sixties and seventies, several groups worked on the utilization of superconductors for magnetic electron lenses. This research had been triggered by the ambition to observe thick specimens which requires a high beam energy and thus strong lenses. With the availability of the high current density superconductors such as Nb_3Sn and NbTi , research directed towards their implementation for magnetic lenses was an obvious step. Superconducting lenses did not become very popular though, since they could only marginally improve upon the performance attained with ohmic conductors and, in addition, their operation required careful planning and specialized skills. Furthermore, high voltage microscopy did not grow into a successful technique.

Since one of the major reasons for the lack of popularity of superconductors in electron microscopy was related with the inconveniences of the helium cryostat, the discovery of high- T_c superconductors, which do not need liquid helium refrigeration, may revive the subject. High- T_c materials can be obtained in the form of sintered bulk material, as thin film or as single crystal. With respect to critical current density and critical magnetic field strength, single crystal material is far better than bulk material. For shielding, high- T_c superconductors are only good to fields of about 1 milli Tesla.

Various techniques can be applied to make high- T_c superconducting thin films. The most important ones are the so called physical vapor deposition techniques that include sputtering, laser ablation, electron beam evaporation and Molecular Beam Epitaxy (MBE). During this work, thin $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films have been made using e-beam evaporation. The films were made either in a two step process, or in-situ, with ozone applied for oxidation.

Because of the high critical current density of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ thin films, it is interesting to investigate the applicability of this material for small iron-free magnetic lenses. Advantages may be in better optical properties and smaller dimensions with respect to the conventional iron circuit lens. The first experimental step for creating a magnetic

lens in a thin superconducting film was to make a 50 turns flat coil with an inner diameter of 2 mm and outer diameter of 8 mm. The coil, with total length of 70 cm, had a critical temperature of 80 K and a critical current density of $3 \cdot 10^4$ A/cm² at 35 K.

When comparing the properties of iron-free helical lenses with those of conventional polepiece lenses, it is concluded that properly dimensioned helical lenses exhibit a lower coefficient of spherical aberration than their conventional counterparts. The limiting mechanism for ultimate performance of iron-free lenses is the maximum current density allowed in the windings and not, as in conventional lenses, saturation of the ferromagnetic circuit. Therefore, high- T_c superconducting thin films are a serious candidate to serve as basic material for making small iron free lenses.

In order to obtain a round lens, two spiral patterned thin films need to be stacked in a face to face orientation. This is a so called lens element. In order to verify the optical properties of this setup, the magnetic field distribution can be derived with the aid of the Biot-Savart law. However, to use these expressions in a ray tracing is not realistic, since the required run time is far too long. Therefore, the axial multipole coefficients of a one turn spiral have been derived, together with their scaling rules. This enables to find the multipole coefficients of any multi turn spiral, simply by adding properly scaled one turn coefficients. Once the axial coefficients of the complete lens element are known, the three dimensional field distribution is readily available through a radial series expansion. This procedure only takes a fraction of the time required by the Biot-Savart method.

In order to test the theoretical concept of a thin film lens element, a prototype has been developed. It consists of two thin films, together with their substrates, in a face to face orientation. Because the patterned substrates have to be aligned with respect to each other in a face to face setup, an optical alignment technique cannot be used. Therefore, a capacitive technique has been employed, which requires capacitive electrodes surrounding the lens spiral. Those electrodes form capacitive position sensors in the face to face setup and are constructed in the thin film together with the lens spiral. The prototype lens spiral has an inner diameter of 1.2 mm, an outer diameter of 7.5 mm and a track width of 20 μ m. The total number of turns is 125. The connection between the innermost turns of the spirals in the face to face orientation will be made

by means of a specially shaped metal film, contacting the innermost turn of the spiral. The actual connection can now be established by clamping the two substrates together.

The optical properties of this prototype lens element have been obtained by means of a ray tracing in the field as found with the axial multipole method described above. Apart from focal properties and round lens aberrations, due to the non-rotational symmetry of the lens field, parasitic aberrations arise, mainly causing deflection and axial astigmatism. Moreover, due to the deflection component of the field, chromatic deflection errors occur.

Apart from the superconducting device, based on a SrTiO_3 substrate covered with a laser ablated $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film, for reasons of costs and availability, most of the experimental work has been effected on a Si substrate covered with an Al film. The capacitive alignment procedure was tested using the Si based device and an overall alignment better than $0.5 \mu\text{m}$ was obtained, well within the criterion of $1 \mu\text{m}$. The critical current density of the 125 turns superconducting spiral, which has a total length of about 2 m, was $1.65 \cdot 10^6 \text{ A/cm}^2$ at 43 K.

It is concluded that with the present level of technology, about 50 Ampere turns can be obtained at liquid nitrogen temperature and about 500 Ampere turns at liquid helium temperature. This is insufficient for most practical applications. The number of ampere turns is likely to be raised by an increase in current density or by the application of films with a thickness over $0.5 \mu\text{m}$, which is the present upper limit. Another future option probably is the application of bulk materials which have impressively been improved, but when aiming for small iron-free lenses made from high- T_c wire, accurate positioning of the windings will be a severe problem.

If the high- T_c materials can be improved further in the sense that a sufficient number of ampere turns is obtained with a thin film lens element, some potentially useful applications arise. Apart from small iron free lenses, these include thin film deflectors or stigmators, all integrated with a thin film lens. Another more advanced application may be a small scanning electron microscope to be used for the in-situ monitoring of processes under vacuum, such as Molecular Beam Epitaxy.

SUMMARY

Since present polepiece lenses are already used to their limits as set by the nature of the ferromagnetic circuit, significant improvements are only to be expected from small iron free lenses for which high- T_c superconductors are a good basic material.

Samenvatting

Deze dissertatie beschrijft de resultaten van een onderzoek naar de toepasbaarheid van hoge temperatuur supergeleiders in de deeltjesoptica en introduceert een mogelijke techniek voor het vervaardigen van magnetische lenzen, gebaseerd op het gebruik van dunne supergeleidende $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films.

In de jaren zestig en zeventig is er door diverse groepen gewerkt aan het toepassen van supergeleiders voor magnetische lenzen. Aanleiding voor dit onderzoek was de wens om dikkere preparaten te kunnen analyseren, waarvoor een hoge bundelenergie noodzakelijk is en dientengevolge dus ook sterke lenzen nodig zijn. Vanwege het beschikbaar komen van supergeleiders met een hoge stroomdichtheid, zoals Nb_3Sn en NbTi , was het een logische stap om het onderzoek te richten op toepassingen van deze materialen in magnetische lenzen. Supergeleidende lenzen zijn echter nooit doorgebroken, omdat hun optische eigenschappen slechts marginaal beter waren dan die van hun conventionele tegenhangers en hun gebruik bovendien specialistische vaardigheden en een zorgvuldige planning van de experimenten vereiste. Daarnaast is de hoogspanningsmicroscopie niet uitgegroeid tot een techniek die op grote schaal wordt toegepast.

Eén van de belangrijkste oorzaken voor het matige succes van de supergeleiders in de elektronen microscopie was de helium cryostaat die voor veel ongemak zorgde. De ontdekking van hoge temperatuur supergeleiders kan het onderwerp supergeleidende lenzen echter opnieuw doen opleven, omdat deze supergeleiders niet gekoeld behoeven te worden tot de temperatuur van vloeibaar helium. Hoge temperatuur supergeleiders kunnen worden verkregen in de vorm van gesinterd bulk materiaal, als dunne film of als éénkristal. Met betrekking tot de kritische stroomdichtheid en de kritische magnetische veldsterkte heeft monokristallijn materiaal veel betere eigenschappen dan gesinterd bulk materiaal. Voor het gebruik in afscherming zijn hoge temperatuur supergeleiders slechts bruikbaar tot velden in de orde van 1 milli Tesla.

Voor het vervaardigen van supergeleidende dunne films kunnen diverse technieken worden toegepast. De meest belangrijke zijn de zogenaamde verdampings depositie technieken zoals sputteren, laser ablatie, elektronen straal verdampen en Moleculaire

Bundel Epitaxi (MBE). Als onderdeel van dit werk zijn dunne $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films vervaardigd met behulp van een elektronen straal verdampings techniek. De films zijn ofwel gemaakt via een tweestaps proces, ofwel via een in-situ procédé, waarbij ozon gebruikt werd voor de oxydatie.

De hoge kritische stroomdichtheid van dunne $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films maakt het interessant om te onderzoeken of deze materialen aangewend kunnen worden in magnetische lenzen. Mogelijke voordelen van zulke lenzen kunnen zijn gelegen in betere optische eigenschappen en/of kleinere afmetingen ten opzichte van de conventionele magnetische lens met ijzercircuit. De eerste experimentele stap op weg naar een magnetische lens gemaakt in een supergeleidende dunne film was het vervaardigen van een platte spoel met 50 windingen die een binnendiameter heeft van 2 mm en een buitendiameter van 8 mm. De spoel, met een totale lengte van 70 cm, had een kritische temperatuur van 80 K en een kritische stroomdichtheid van $3 \cdot 10^4 \text{ A/cm}^2$ bij 35 K.

Wanneer de optische eigenschappen van ijzerloze platte lenzen vergeleken worden met die van conventionele poolschoenlenzen, kan worden vastgesteld dat juist gedimensioneerde platte lenzen een lagere coëfficiënt van sferische aberratie bezitten dan hun conventionele tegenhangers. Het mechanisme dat de lenseigenschappen van ijzerloze lenzen begrensd is de maximaal mogelijke stroomdichtheid in de windingen en niet, zoals in conventionele lenzen, de verzadiging van het ijzercircuit. Daarom zijn dunne films van hoge temperatuur supergeleidend materiaal een serieuze kandidaat om te dienen als basis voor de vervaardiging van kleine ijzerloze lenzen.

Om een ronde lens te verkrijgen dienen twee films, beide voorzien van een spiraalpatroon, te worden gestapeld in een dusdanige oriëntatie dat de spiraalpatronen op elkaar komen te liggen. Hierdoor ontstaat er in de bijbehorende veldverdeling een betere rotatiesymmetrie. Dit is een zogenaamd lens element. Om de elektron optische eigenschappen van deze lens te bepalen is de magnetische veldverdeling afgeleid met behulp van de wet van Biot-Savart. Deze formules zijn echter ongeschikt om te worden gebruikt in een raytracing, daar de benodigde rekentijd veel te groot is. Daarom is gezocht naar een andere methode en zijn de axiale multipool coëfficiënten en schalingsregels afgeleid voor een spiraal met één winding. Dit maakt het mogelijk om de multipoolcoëfficiënten te bepalen voor een willekeurige spiraal, simpelweg door de

coëfficiënten van de spiraal met één winding op te tellen na voorvermenigvuldiging met de juiste schaalfactoren. Indien de axiale multipoolcoëfficiënten van het complete lens element bekend zijn, kan de drie dimensionale veldverdeling worden bepaald door middel van een radiële reeksontwikkeling. Deze procedure neemt slechts een fractie van de tijd die benodigd is voor een berekening via de Biot-Savart methode.

Om het theoretisch concept van een dunne film lens te testen is een prototype ontwikkeld. Het bestaat uit twee dunne films, die met hun substraten in een zodanige oriëntatie worden geplaatst dat de spiralen op elkaar liggen. Omdat de spiraal patronen op elkaar liggen en de substraten dus naar buiten gekeerd zijn, kan het uitlijnen niet met behulp van een optische techniek worden uitgevoerd. Daarom is een capacitieve techniek gebruikt waarvoor het noodzakelijk is om rond de lens spiraal capacitieve elektrodes aan te brengen. Deze elektrodes vormen capacitieve positie sensoren in de lens element opstelling en worden tezamen met de lens spiraal in de dunne film aangebracht. Het prototype van de lens spiraal heeft een inwendige diameter van 1.2 mm, een uitwendige diameter van 7.5 mm en een spoorbreedte van 20 μm . Het totaal aantal windingen is 125. De verbinding tussen de binnenste windingen van de spiralen in het lens element wordt gemaakt met behulp van een metaal film met een speciale geometrie die aansluit op de binnenste winding van de spiraal. De eigenlijke verbinding wordt dan gelegd door de twee substraten op elkaar te klemmen.

De optische eigenschappen van dit prototype lens element zijn verkregen met behulp van raytracing in het veld gegeven in de vorm van axiale multipoolcoëfficiënten, zoals in het bovenstaande beschreven. Als gevolg van de niet rotatie symmetrische veldverdeling vertonen deze dunne film lenzen, naast eerste orde eigenschappen en ronde lens aberraties, ook parasitaire aberraties, die zich voornamelijk uiten als afbuiging van de bundel en astigmatisme. Bovendien ontstaan er ten gevolge van de afbuigcomponent in het veld ook extra chromatische fouten.

Naast het supergeleidende lens element, gebaseerd op een SrTi_3 substraat bedekt met een $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film is, omwille van de kosten en de beschikbaarheid, het meeste experimentele werk uitgevoerd aan een Si substraat bedekt met een Al film. De capacitieve uitlijnprocedure is getest met een lens element gebaseerd op Si en een uitlijnnaauwkeurigheid beter dan 0.5 μm is hiermee aangetoond, hetgeen onder het

gestelde criterium van $1\text{ }\mu\text{m}$ valt. De gemeten kritische stroomdichtheid van de spiraal met 125 windingen, die een totale lengte heeft van ongeveer 2 meter, is $1.65 \cdot 10^6\text{ A/cm}^2$ bij 43 K.

Er kan worden geconcludeerd dat er bij de huidige stand van de technologie met een dunne film lens element ongeveer 50 Ampere-windingen kunnen worden verkregen bij vloeibaar stikstof temperatuur en ongeveer 500 Ampere-windingen bij vloeibaar helium temperatuur. Dit is ontoereikend voor de meeste toepassingen. De meest waarschijnlijke ontwikkelingen die het aantal Ampere-windingen kunnen doen toenemen zijn een hogere maximale stroomdichtheid, of het beschikbaar komen van films met een dikte groter dan de huidige bovengrens van $0.5\text{ }\mu\text{m}$. Een andere mogelijkheid is het toepassen van bulk materiaal dat de laatste tijd enorm is verbeterd. Indien men echter kleine ijzerloze lenzen wil vervaardigen met behulp van bulk materiaal zal het een probleem worden om de windingen voldoende nauwkeurig te positioneren.

Indien de hoge temperatuur supergeleiders verder kunnen worden verbeterd, in die zin dat voldoende Ampere-windingen worden verkregen met een dunne film lens, zijn er enige interessante potentiële toepassingen te verwachten. Deze omvatten, naast kleine ijzerloze lenzen, dunne film afbuigers en stigmatoren, welke geïntegreerd kunnen worden met een dunne film lens. Een meer geavanceerde optie is het vervaardigen van een kleine scannende elektronen microscoop die kan worden gebruikt om processen die zich onder vacuüm afspelen in-situ te bekijken, zoals bijvoorbeeld Moleculaire Bundel Epitaxi (MBE).

Aangezien de optische kwaliteitslimiet, bepaald door de eigenschappen van het ferromagnetisch circuit, bij de huidige poolschoen lenzen reeds bereikt is, kunnen significante verbeteringen slechts verwacht worden van kleine ijzerloze lenzen met een hoge stroomdichtheid, waarvoor de hoge temperatuur supergeleiders een goed uitgangsmateriaal vormen.

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Curriculum Vitae

Jan-Peter Adriaanse werd geboren op 15 november 1963, te Zierikzee. Na zijn lagere school in Dreischor doorliep hij, van 1976 tot 1981, het HAVO en van 1981 tot 1983 het VWO aan de Rijksscholengemeenschap "Professor Zeeman" te Zierikzee.

Wellicht geïnspireerd door de naam van zijn school, startte hij in 1983 met de opleiding Technische Natuurkunde aan de Technische Hogeschool Delft. Zijn afstudeerwerk verrichtte hij in de vakgroep Deeltesoptica, onder leiding van Prof. van der Mast. Tijdens deze afstudeerperiode deed hij een stage bij het "Institut Jozef Stefan", te Ljubljana, Joegoslavië, waar hij meewerkte aan het ontwikkelen van software voor een klimaatbeheersingssysteem.

Na zijn afstuderen, in 1987, begon hij zijn promotiewerk. Gedurende deze periode was hij in dienst bij TNO, organisatorisch ondergebracht bij de Technisch Fysische Dienst TNO-TU Delft en werkzaam aan de TU Delft, hetgeen vaak niet zonder meer werd begrepen. Het eerste jaar werkte hij mee aan het ontwikkelen van software voor het optimaliseren van elektrostatische lenzen. Daarna startte hij met zijn eigenlijke promotiewerk, namelijk een haalbaarheidsonderzoek naar het gebruik van hoge temperatuur supergeleiders voor toepassingen in de deeltjesoptica. Hij begon dit werk in de vakgroep supergeleiding, onder supervisie van Prof. Mooij en werkte gedurende één jaar mee aan het vervaardigen van supergeleidende dunne films met een hoge kritische temperatuur. In de resterende jaren spitste hij zijn onderzoek toe op het ontwikkelen van een prototype voor een magnetische lens, gebaseerd op supergeleidende dunne films, hetgeen beschreven is in dit proefschrift. In deze periode ook, was hij actief betrokken bij de oprichting van de aan de universiteit gelieerde Stichting Deeltjes Optica Delft, waarin hij zich later als bestuurslid onder meer bezig hield met het coördineren van de verkoop van software aan diverse bedrijven en instellingen.

Momenteel is hij, als ontwerper van elektronen optiek voor beeldbuis-toepassingen, werkzaam bij Philips Components te Eindhoven.

Appendix A: Recipe for fabrication of a stack element.

The photoresist that has been used in the patterning process is AZ 5214E, from Hoechst. This photoresist can either be used in the positive or in the negative mode. When used in the negative mode, the resist needs a reverse bake and a flood exposure, following the contact exposure, in order to first polymerize the exposed area and then expose the rest of the resist. When the photoresist has been processed according to this scheme, the polymerized areas will not be removed during development, so the negative pattern is obtained. This procedure just described is known as image reversal.

I

- Start with a SrTiO_3 substrate of 0.5 mm thickness covered with a 0.5 μm film of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ made by laser ablation
- Spin on AZ 5214E from Hoechst at 5700 rpm during 30 seconds. Thickness of the resist: $\pm 1.2 \mu\text{m}$.
- Prebake of the photoresist on a hotplate at 100° C during 2.5 minutes.
- Contact exposure with the first mask, in a mask aligner, during 14 seconds (power density: 1.75 mW/cm²).
- Reverse bake on a hotplate at 125° C during 2.5 minutes.
- Flood exposure during 45 seconds (power density: 1.75 mW/cm²).
- Develop the photoresist in PLSI developer from Hunt in a 1:1 solution with deionized water during 60 seconds
- Deposit 2 nm Cr + 200 nm Ag
- Lift-off in Acetone during 30 minutes using an ultrasonic cleaner
- Etch the $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ in hydrochloric acid (37 %) and deionized water (1:800 solution) during 30 seconds

II

- Spin on AZ 5214E from Hoechst at 5700 rpm during 30 seconds. Thickness of the resist: $\pm 1.2 \mu\text{m}$.
- Prebake of the photoresist on a hotplate at 100° C during 2.5 minutes.

- Contact exposure with the second mask, in a mask aligner, during 14 seconds (power density: 1.75 mW/cm^2).
- Develop the photoresist in PLSI developer from Hunt in a 1:1 solution with deionized water during 60 seconds
- Deposit 800 nm Al_2O_3
- Lift-off in Acetone during 30 minutes using an ultrasonic cleaner

III

- Spin on AZ 5214E from Hoechst at 5700 rpm during 30 seconds. Thickness of the resist: $\pm 1.2 \mu\text{m}$.
- Prebake of the photoresist on a hotplate at 100°C during 2.5 minutes.
- Contact exposure with the third mask, in a mask aligner, during 14 seconds (power density: 1.75 mW/cm^2).
- Develop the photoresist in PLSI developer from Hunt in a 1:1 solution with deionized water during 60 seconds
- Deposit 900 nm Ag and 100 nm Au
- Lift-off in Acetone during 45 minutes using an ultrasonic cleaner