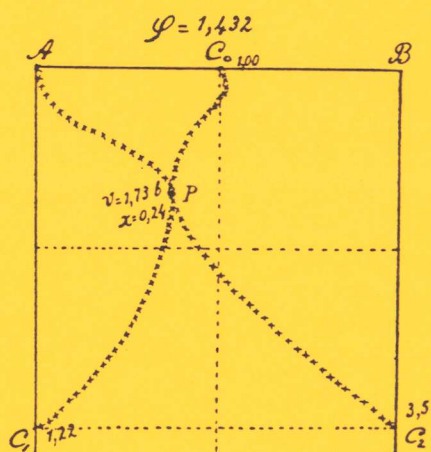


SOME MILESTONES IN THE LIFE AND WORK OF THE  
MATHEMATICAL CHEMIST J.J. VAN LAAR (1860-1938).

CALPHAD XIX (1990)



by

Everard Peter van Emmerik

June 1990

\*\*\*\*\*

SOME MILESTONES IN THE LIFE AND WORK OF THE  
MATHEMATICAL CHEMIST J.J. VAN LAAR (1860-1938).

CALPHAD XIX (1990)

Contents:

1. Introduction
2. Life
3. Recognition
4. Relations with Lorentz
5. Work
  - 5.1 Mathematical Chemistry
  - 5.2 Designer: Melting point line (1902)
  - 5.3 Arithmetician: Spinodal (1905)
  - 5.4 Maker of Predictions: Retrograde Solubility (1908)
6. Autumn of his life
7. Conclusion

Notes

by

Everard Peter van Emmerik

June 1990

\*\*\*\*\*

Abstract

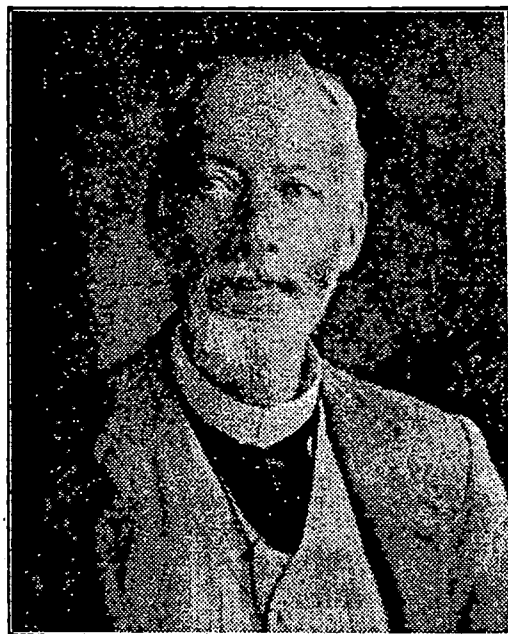
This lecture: 'Some Milestones in the Life and Work of Mathematical Chemist J.J. van Laar (1860-1938)' discusses his life and three important examples of his work.

Insufficient schooling meant that Van Laar was not permitted to take academic examinations and this lack of recognized qualifications led to problems in establishing his scientific career. These difficulties brought him close to mental collapse in 1896 and 1911. At the University of Amsterdam he was opposed by J.D. Van der Waals (1837-1923) but was highly appreciated by H.W. Bakhuis Roozeboom (1854-1907). This lecture focuses on some of his achievements and describes his recognition as a scientist. H.A. Lorentz (1853-1928) played an important part in Van Laar's development as a mathematical chemist. Van Laar is said to have endowed him with 'the father role' and it is certain that Lorentz encouraged and supported him.

The study of mathematical chemistry as Van Laar practiced it no longer exists. Together with Bakhuis Roozeboom, Van Laar is the founder of phase theory. Examples of his work are discussed in the sections of the melting point line of tin amalgam (1902), the spinodal (1905) and retrograde solubility (1908).

The lecture concludes with a description of the final years of his life and includes the kind letter of condolence by L.S. Ornstein (1880-1941). Throughout the lecture reference is made to fragments of letters and to photographs.

SOME MILESTONES IN THE LIFE AND WORK OF THE  
MATHEMATICAL CHEMIST J.J. VAN LAAR (1860-1938).



Van Laar in his sixties.

1. Introduction

Tonight, Ladies and Gentlemen, I would like to discuss Van Laar. Some of these facts about him are well known, others are more obscure and have lain untouched in archives for almost a century. For a more comprehensive study of Van Laar's life and work, I would like to refer you to my future thesis. I will begin with a short description of Van Laar's life. This will be followed by three examples of his work which are still relevant today: his work as a designer of the phase diagram for the melting point line (1902), as arithmetician of the spinodal (1905) and as the man who predicted retrograde solubility (1908). The lecture will conclude with a description of his later years and after that, I will do my utmost to answer any

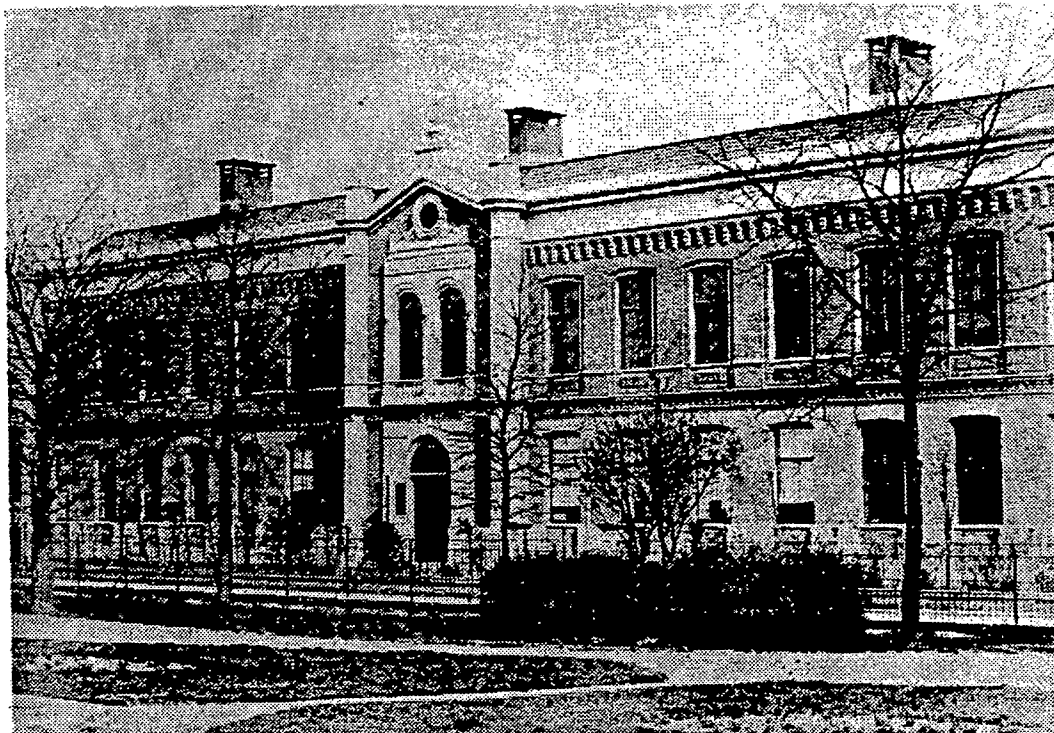
questions you may have. I must mention first of all that I am no specialist on phase theory. If you have questions on this subject, Dr. H.A.J. Oonk has kindly offered to help answer them.

## 2. Description of Van Laar's life

Van Laar's life can be divided into two parts, his Dutch period from 1860 until 1911 and his Swiss period from 1911 until his death in 1938.

He grew up in Haarlem and was orphaned at the age of twelve. An aunt subsequently took care of him and his sister Carolina and at the age of sixteen he was taken out of the HBS (the Dutch grammar school), and sent to the Royal Naval Institute. The reasons for this change have always remained obscure. He graduated from this Institute without any problems. He copied the mathematical notes from older fellow-students and in this way studied the mathematics that he would later apply with great ingenuity and determination. In 1880 he was appointed sub-lieutenant. However, naval life did not suit with him and he resigned in 1881. From 1881 until 1884 he studied chemistry with J.H. van 't Hoff (1852-1911) and J.D. van der Waals (1837-1923) in Amsterdam. His mathematical knowledge and his first scientific publication in 1892, formed the basis of his mathematical chemistry.

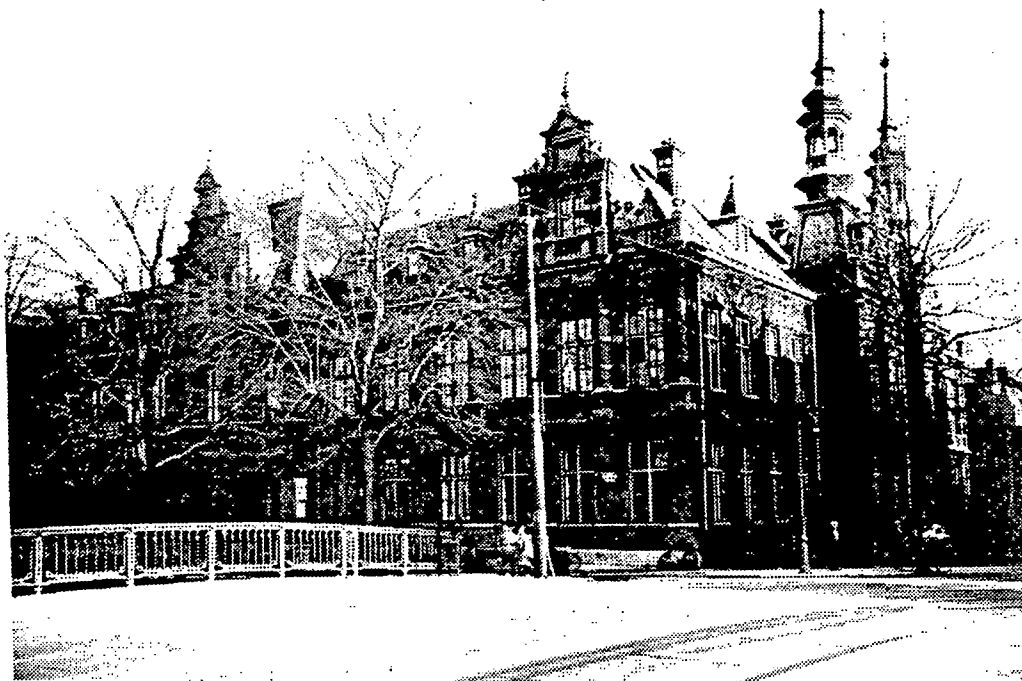
His inadequate preparatory education did not allow him to take academic exams but his naval rank did however allow him to teach mathematics at grammar schools. Thus, recommended by Van der Waals, he was appointed as a teacher in Middelburg in 1884, and recommended by H.A. Lorentz (1853-1928) in Utrecht in 1895. In 1896 he suffered a nervous breakdown which was probably caused by disappointment and disciplinary problems in the classroom. In 1897 he was given an honorary discharge for medical reasons. This term of office earned him a small annual pension of



Grammar school in Utrecht.<sup>1</sup>

fl.589,- a year. After having spoken to H.W. Bakhuis Roozeboom (1854-1907) and Van der Waals he was appointed as an unsalaried lecturer of mathematical chemistry in Amsterdam. In 1903 he became a research assistant to Bakhuis Roozeboom. He proved himself to be a good worker and got along very well with Bakhuis Roozeboom. However, he proved to be a man weak in social virtues and this often led to fierce polemics and emotional outbursts. Among these conflicts were arguments with W. Nernst (1864-1941) and W. Ostwald (1853-1932). After 1900 serious problems with Van der Waals began to surface when he tried to undermine Van Laar's position at the University. The archives show that Van der Waals did indeed try to do this. Whether this was caused by 'incompatibilité d'humeur' or Van Laar's 'mathematical chemistry', which Van der Waals regarded as his area, and which, he felt, was competitive, is not clear. The final result was that Van Laar was not appointed lecturer of mathematical

chemistry but was given the position of lecturer of mathematics



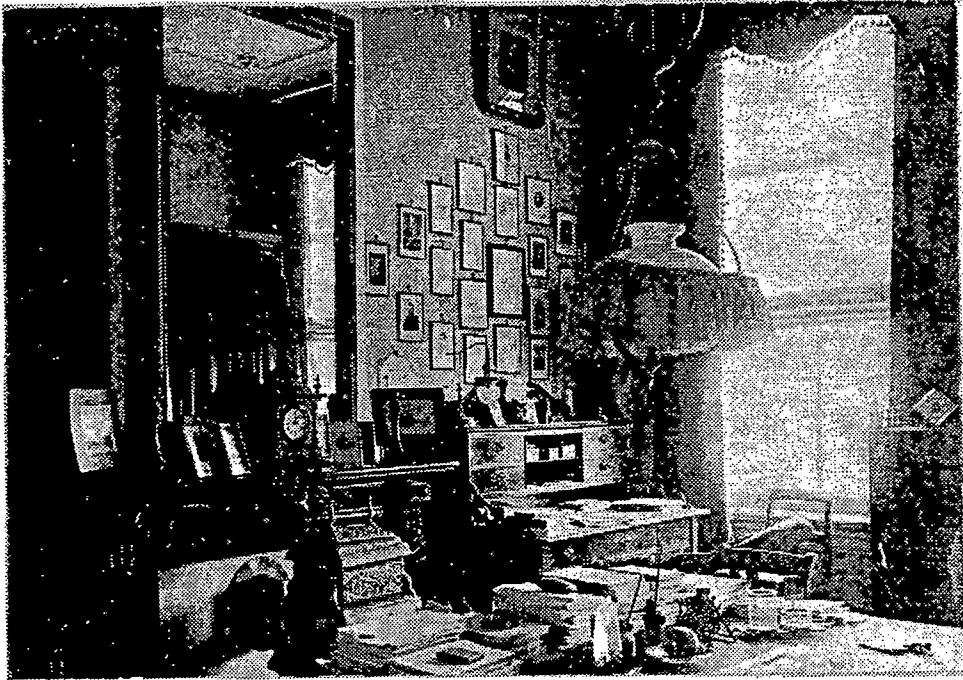
Chemistry laboratory where Van Laar worked from 1898 until 1912, demolished in 1988.<sup>2</sup>

especially for students in chemistry and biology. This not-adjusted situation again led to a nervous breakdown and in 1911 he left for Switzerland on sick leave. In 1912 he resigned on medical grounds and remained in Switzerland until his death, staying mainly in Tavel-sur-Clarens. After his resignation he put everything aside to work on scientific matters. Van Laar belonged to the margins of the group 'Tachtigers' a cultural group which started in the eighteen eighties and he was, like most members of this group, an agnostic.<sup>3</sup>

### 3. Recognition

In 1914 the University of Groningen awarded a number of honorary degrees to mark the tricentenary of its founding. The minutes of 24 January 1914 of the Faculty of Mathematics and Physics show

that Van Laar's name did not occur on the original list of those to be awarded honorary degrees. F.M. Jaeger (1877-1945)



Study of Bakhuis Roozeboom.<sup>4</sup>

nominated Van Laar at the meeting of 5 February 1914:

'A lengthy discussion arises on the nomination made by Jaeger of a candidate from our country, J.J. Van Laar to (crossed out). The decision is reserved to give Mr. Haga the opportunity to inquire in writing of Prof. Lorentz'.<sup>5</sup>

And the minutes of 10 February 1914:

'Mr. Haga reads the letter by Prof. Lorentz on the scientific merits of Mr. J.J. Van Laar. As a result of this, it is decided to nominate Mr. Van Laar for an honorary degree'.<sup>6</sup>

On 1 July 1914 Van Laar was promoted to Doctor Honoris Causa. Queen Wilhelmina (1880-1962) and Prince Consort Hendrik (1876-1934) attended the graduation ceremony in the 'Nieuwe



Kerk' in Groningen. Van Laar wrote letters to the Rector Magnificus and the Senate, and to the Faculty of Natural Sciences, about the invitation to come to Groningen. He regarded the promotion as a ray of light in his life and the first official recognition....<sup>7</sup> In his letter of 13 June 1914 to the editor of 'Chemisch Weekblad', W.P. Jorissen (1869-1959), Van Laar proved to be very pleased about his promotion:

'I would wish that, they, too had been lacking something, so that I could congratulate them on obtaining it! Lorentz and others were also very pleased; I also received a very kind letter from Kruyt, in which he even spoke of divine justice!'<sup>8</sup>

In 1929 he was awarded the Bakhuis Roozeboom medal and in 1931 he became correspondent of the Royal Academy and he was knighted into the Order of the Dutch Lion. In 1938 he became honorary member of the Chemical Society. He published twelve books and approximately 200 scientific articles.



Van Laar accepts the Bakhuis Roozeboom medal from F.A.H. Schreinemakers (1864-1945).<sup>9</sup>

#### 4. Relations with Lorentz

Contacts between Van Laar and Lorentz were established in 1893 when Lorentz wrote to Van Laar on the occasion of the publication of his first scientific textbook 'Thermodynamik in der Chemie'.<sup>10</sup> From the 72 letters Van Laar wrote to him we can see that Lorentz meant a lot to Van Laar. These letters give details of Van Laar's personal life and his scientific work. Lorentz was sometimes the father figure Van Laar apparently needed. It was Lorentz who brought about his departure from Middelburg and guided Van Laar's work on phase theory in the period between 1903 and 1907. The honorary doctorate Van Laar received in Groningen in 1914 was also made possible by the positive advice Lorentz submitted. Lorentz's celebration of 25 years Ph.D. in 1900 replied with a photograph which he sent to everyone who had congratulated him. When Van Laar received the photograph, he wrote, what was possibly his most beautiful letter.

Utrecht, 3 April, 1901.

Dear Professor,

May I thank you very much for sending me your portrait , with its great likeness, and the handwritten caption. It was a truly pleasant surprise.

Looking at the picture will not only bring one of the most excellent scientists to mind, but also an excellent man, who is always willing to spend time to support and help others in their scientific endeavours, and is always willing to encourage others, both in word written and spoken.

For the workers in the vineyard of science, despondency and misfortune often await, and a single encouraging word by you often works miracles. I myself have often benefited from them.

Again, thanking you deeply, greeting you, I remain, yours faithfully,

Yours obd.

J.J. van Laar.<sup>11</sup>

In 1903 Van der Waals proposed to the Physics Department of the Royal Academy of Science to allow two members to tender an article by a second party, instead of one.<sup>12</sup> Van der Waals supposedly tried on this occasion to hinder Van Laar in tendering his work. Bakhuis Roozeboom wrote a letter to Lorentz to this effect on 2 December 1904:

(...) About that submission (unreadable) I have then Van Der Waals (unreadable) now slowly begin to believe that Van der Waals has thought out this entire scheme, tendering articles by two people, to hurt Van Laar - instead of helping him, as is his duty (...).<sup>13</sup>

Schreinemakers in a letter of 10 May 1928 to F.G. Waller (1860-1935) in Delft wrote:

(...) many years ago they tried to prevent Van Laar from tendering his articles to the Academy: Lorentz as first tenderer and I did it then, and that was the case until Lorentz died.<sup>14</sup>

Van der Waals must have disliked Van Laar very much indeed!



Lorentz in 1901.<sup>15</sup>

## 5. Work

Van Laar deserves the credit for the development of phase theory, following the theory by Van der Waals for binary mixtures (1889 and 1900). Van Laar was the first to provide a theoretical quantitative thermodynamic discussion.<sup>16</sup> It examines the content of mathematical chemistry which is explained with the help of three examples: the melting point line of 1902, the spinodal of 1905 and the retrograde solubility of 1908.

### 5.1 Mathematical Chemistry.

Today, mathematical chemistry is no longer an independent subject of study, at least not under this name. In chemistry, experimentation is paramount and it is followed by the theory. However, one might say that around the turn of the century there was room for a primarily theoretical point of view because, at the time, the application of mathematics on chemistry was relatively new. Therefore, I would like to discuss Van Laar's views on mathematical chemistry briefly. In his biography on Van der Waals in 1900 Van Laar stated that it was Gibbs who provided an entirely new point of view with his thermodynamics of chemical transitions and equilibria and that the birth of mathematical chemistry was marked by this discussion.<sup>17</sup> Van Laar referred here to Gibbs's discussion 'On the Equilibrium of Heterogenous Substances' of 1876. In the 'Vorwort of Van Laar's 'Lehrebuch der Mathematischen Chemie' of 1901 he qualifies mathematical chemistry as the pendant of mathematical physics.<sup>18</sup> The older 'systematical chemistry' and the new 'physical or theoretical chemistry' in his view belong to experimental chemistry. The foundation of his mathematical chemistry is thermodynamics as developed by Gibbs, Planck and Duhem, and comprises all theoretical chemical problems in which mathematics as a supporting science is essential. This also includes the formulation of a theory on electrochemistry. Van Laar believed it to be a new science which at the time broke new ground. From

this discussion it will be clear that the formulation of a phase theory must be part of mathematical chemistry. In his textbook he first discusses Gibbs' thermodynamic potential ( $G$ ) and Planck's potential ( $-G/T$ ), in which he calculated different phase equilibria.

I would like to mention one special feature of this book. It contains an introduction by Bakhuis Roozeboom. From a postcard of 10 July 1899, and a letter of 16 August 1899, which Van Laar wrote to Lorentz it appears that publisher Sijthoff in Leyden wanted to publish the book, if 'a positive and encouraging word about it would be heard from Lorentz'.<sup>19</sup> Sijthoff sent the manuscript to Lorentz who, after some weeks, sent it on to Van Laar who wanted it back. Van Laar's letter on 16 August 1899 to Lorentz is long and sad.<sup>21</sup> He complains about direct opposition and 'hushing up'. Lorentz reacted immediately on 17 August 1899 and explained his side of the story.<sup>21</sup> He had reluctantly agreed to Sijthoff's request and had wanted to read the manuscript thoroughly. There were also some objections regarding content. Van Laar replied on 18 August 1899 and the tone of the letter was cheerful. Since his illness in 1896 Van Laar had regarded Lorentz lost to him and:

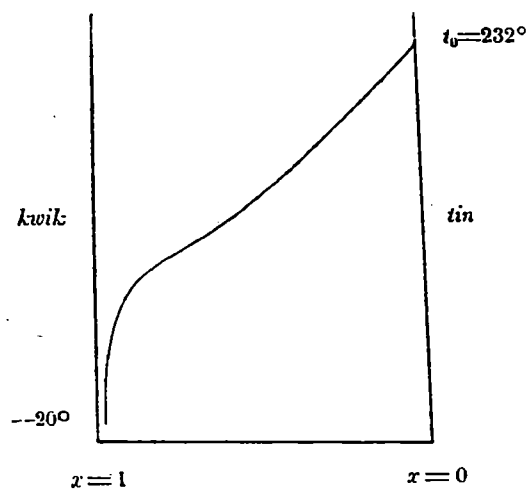
'In a pent up overflow of feeling, I uttered my grievances. If only I had heard from you, that you were too busy, that you would rather study my handwriting after the holidays, so that I could ask for it back if I wished to do so - I would immediately have resigned myself, and waited'.<sup>22</sup>

Lorentz did not write the introduction but fortunately Bakhuis Roozeboom was willing to do so instead.



## 5.2 Designer: Melting point line (1902)

I will first talk about an interesting article written by Van Laar in 1902: 'The course of the melting point lines of solid alloys or amalgams'.<sup>24</sup> In this article he describes how he designed the melting point line of tin amalgam in a way that is still in use to day.



Melting point line of tin amalgam.<sup>25</sup>

It was Van Laar who theoretically deduced the course of this melting point line by equating the thermodynamic potentials of solid tin  $\mu = u - cT$  and liquid amalgam  $\mu = u_1 - c_1T + RT \log(1-x)$ :

$$T = T_0 / (1 - RT_0/q \cdot \log(1-x)) \quad (1)$$

$T_0$  is the melting temperature of pure tin and  $q$  the melting heat of solid tin at the transition into amalgam. Equation (1) represents the experimentally deduced melting point line qualitatively correct. In the same article Van Laar, with the

help of Van der Waals' equation of state, deduces a stricter relation. For the thermodynamic potentials he takes  $\mu = u_1 - c_1T - (ax^2 + bx^3 + cx^4) + RT \log(1 - x)$  for solid tin, and  $\mu = u_1 - c_1T - (ax^2 + bx^3 + cx^4) + RT \log(1 - x)$  for liquid amalgam. After equation the result is:

$$T = T_0 (1 - (ax^2 + bx^3 + cx^4)) / (1 - RT_0/q \cdot \log(1-x)) \quad (2)$$

This formula is still correct.<sup>26</sup> For a he calculated: -0.325, for b: 1.11 and c: -1.33. With  $RT_0/q = 0.4$  and substitution in equation (2) he founds a particularly good quantitative correspondence! See the table below:

$x$	$x^2$	$x^3$	$x^4$	$-\log(1-x)$	Teller	Noemer	$T - 273$ berek.	$T - 273$ gev.	$\Delta$
0,1005	0,01010	0,001015	0,0001021	0,1059	1,0023	1,0424	212,4	211,6	0,8
0,1716	0,02945	0,005053	0,0008673	0,1883	1,0051	1,0753	198,9	198,6	0,3
0,2338	0,05466	0,01278	0,002988	0,2663	1,0076	1,1065	186,7	183,7?	3,0?
0,2969	0,08815	0,02617	0,007779	0,3523	1,0099	1,1409	173,8	173,0	0,8
0,3856	0,1487	0,05733	0,02211	0,4872	1,0141	1,1949	155,4	155,2	0,2
0,5001	0,2501	0,1251	0,06255	0,6933	1,0256	1,2773	132,3	133,4	-1,1
0,5973	0,3568	0,2131	0,1273	0,9095	1,0488	1,3638	115,2	115,2	0,0
0,6467	0,4182	0,2705	0,1749	1,0404	1,0682	1,4161	107,7	107,4	0,3
0,6754	0,4562	0,3081	0,2081	1,1252	1,0830	1,4501	104,0	103,4	0,6
0,6813	0,4642	0,3162	0,2155	1,1435	1,0866	1,4574	103,3	102,4	0,9
0,7104	0,5047	0,3585	0,2547	1,2393	1,1047	1,4957	99,8	99,0	0,8
0,7155	0,5119	0,3663	0,2620	1,2570	1,1083	1,5028	99,2	98,8	0,4
0,7477	0,5591	0,4180	0,3126	1,3772	1,1335	1,5509	95,9	95,4	0,5
0,7547	0,5696	0,4299	0,3244	1,4053	1,1393	1,5621	95,1	94,0	1,1
0,7963	0,6341	0,5049	0,4021	1,5912	1,1805	1,6365	91,1	90,0	1,1
0,8189	0,6706	0,5492	0,4497	1,7087	1,2064	1,6835	88,7	88,4	0,3
0,8921	0,7958	0,7100	0,6333	2,2266	1,3128	1,8906	77,5	79,7	-2,2
0,9483	0,8993	0,8528	0,8087	2,9623	1,4212	2,1849	55,3	65,2	-9,9

Van Laar's table.<sup>27</sup>



In 1903 the formula was improved and included in Bakhuis Roozeboom's book 'Die heterogene Gleichgewichte' (page 293) of 1904.<sup>28</sup>

### 5.3 Arithmetician: Spinodal (1905)

In his article in 1905 for the Academy: 'An exact expression for the course of spinodal curves and their plait-points for all temperatures, in the case of mixtures of normal substances' Van Laar found, with the help of Van der Waals' state equation, the equation for the projection on the v,x-plane of the spinodal for a binary mixture which consists of so-called 'normal' substances (that is to say: without association or polarity):

$$RT = \frac{2}{v^3} \left[ x(1-x) \left\{ (b_1 \sqrt{a_2} - b_2 \sqrt{a_1}) + \alpha(v-b) \right\}^2 + \alpha(v-b)^2 \right] \quad (3)$$

$$\alpha = \sqrt{a_2} - \sqrt{a_1}$$

and for the projection on the v,x-plane of the plait point curve (the geometrical position of the plait points at changing temperatures):

$$x(1-x)\theta^2 \left[ (1-2x)v - 3x(1-x)\beta \right] + \sqrt{a}(v-b)^2 \left[ 3x(1-x)\theta(\theta - \beta\sqrt{a}) + \right. \\ \left. + a(v-b)(v-3b) \right] = 0 \quad (4)$$

$\theta = \pi + \alpha(v-b)$ ,  $\pi = b_1\sqrt{a_2} - b_2\sqrt{a_1}$ ,  $\alpha = \sqrt{a_2} - \sqrt{a_1}$  and  $\beta = b_2 - b_1$ , in which it is assumed that  $b_{12} = \frac{1}{2}(b_1 + b_2)$  and  $a_{12} = \sqrt{a_1 a_2}$ .<sup>29</sup>

Van Laar's letter to Lorentz of 20 March 1905 shows that Van Laar had a lot of trouble reaching this conclusion:

Hilversum 20 March, 1905.

Dear Professor,

I have read your letter with great interest. I thank you again for your trouble and your corrections. no. 1 - 12.

I will rewrite my article and use the corrections and appropriate fragments on page 5 ( $df/dx + df/dv(dv/dx)_{p,T} = 0$ ) in the way you suggested.

I regret that I cannot agree to your expectation of the calculation of the  $\psi$ -function, because your equation (6)

$$\frac{\partial^3 \psi}{\partial x^3} \left( \frac{\partial^2 \psi}{\partial v^2} \right)^2 - 3 \frac{\partial^3 \psi}{\partial x^2 \partial v} \frac{\partial^2 \psi}{\partial x \partial v} \frac{\partial^2 \psi}{\partial v^2} + 3 \frac{\partial^3 \psi}{\partial x \partial v^2} \left( \frac{\partial^2 \psi}{\partial x \partial v} \right)^2 - \frac{\partial^3 \psi}{\partial v^3} \frac{\partial^2 \psi}{\partial x^2} \frac{\partial^2 \psi}{\partial x \partial v} = 0$$

will become simpler.

I had already tried this, but without success. The differential quotients you suggest are simple, but this is among others no longer the case for  $d^2\psi/dx^2$ . But the third differential quotient becomes very ugly because also  $\theta = \pi + (1-b)$  is a function of both  $v$  (and because of  $b$ ) and  $x$  so that all these quotients get more terms. At the time I did not succeed in omitting terms so that the result becomes simple. Of course, this should be the case after further calculations, and the result will also appear in this way but that calculation will be much longer than mine.

Van der Waals says somewhere that the calculation by  $(d^2\psi/dx^2) \cdot (d^2\psi/dv^2) - (d^2\psi/dxdv)^2$ , etc. becomes so complicated that it is unfeasible. (See also Cont.II. page 137 line 11 ex.) Korteweg gave, with a lot of trouble, an approximation formula at the edge ( $x=0$ ).

The only merit of my derivation is that I succeeded, by a special method of term-summarizing, which only came to light accidentally because of my special method, to find the relatively simple expression. Already, the equation (6) for the projection of the spinodal line on the  $v, x$ -plane

$$RT = \frac{2}{v^3} [x(1-x)\theta^2 + a(v-b)^2]$$

has not been deduced by anybody so far. You were right when you wrote: "Of course, it doesn't belong to the things of which one might say: I could have done that". Still, a restriction has to be made here (unreadable) can test the calculation and start, but then one would find, like Van der Waals and Korteweg, who surely must have often tried this, such complicated forms that one gives in. I was no more fortunate, but I still tried to make something out of it, but always in vain, until I succeeded in this way.

I have elaborately indicated the steps in deductions (8) and (6), and this could be abbreviated if one were not to mention everything. But that would of course only be a sham abbreviation. I have even tried to get the result in many different ways.

Should I leave (6) in the form

$$RT = \frac{2}{v^3} [x(1-x)\theta^2 + a(v-l)^2]$$

it does not work. But if I write

$$RT = \frac{2}{v^3} [x(1-x)(\pi^2 + 2\alpha(v-l)) + (v-l)^2(a_1 + x(a_2 - a_1))]$$

it does work!

$$\theta = \pi + \alpha(v-l)$$

You should see the paper on which I made the calculations which failed!

That is why I will leave my calculations (unreadable), as I finally found them after a lot of trouble; and you - after correction of all the rest, and after the corrections you submitted - I will return it in a few days after I have copied it. If you would in the meantime ask Professor Bakhuis Roozeboom to be the second person tendering the article, he will certainly agree. Thus the article could be tendered on Saturday and I can quietly deduce the consequences of my equation, which I so far have only done in principle because my head was tired and I have had to take some rest.

I am certain you will be able to agree. Thank you again, I remain, after kind greetings, Yours sincerely,

yours obd.

J.J. van Laar.<sup>30</sup>

That Van Laar was a real arithmetician is shown again. Assuming that the covolumes of both components are equal ( $b_1=b_2$ ) and assuming the validity of the empirical relation of Berthelot  $a_{12}=\sqrt{a_1a_2}$  he simplified the equations (3) and (4) into two new equations and thus discovered, after solving the seventh power equation, the double point. This point is, because of Meijer's publication, again topical.<sup>31</sup> Van Laar calculated more plait-point lines for different temperatures. Note the figure below. In figure 4 we see two plait point lines that cross at point P, the double point, which Meijer called the Van Laar point.

J. J. VAN LAAR. „Over het verloop der plooiingslijnen bij mengels van normale stoffen.”

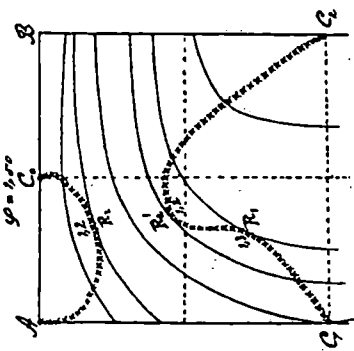


Fig. 3.

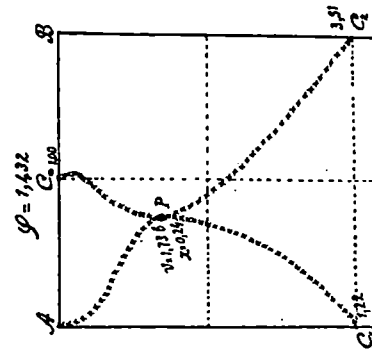


Fig. 4.

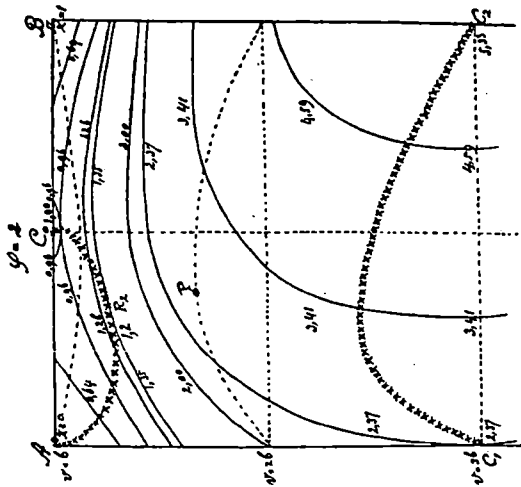


Fig. 2.

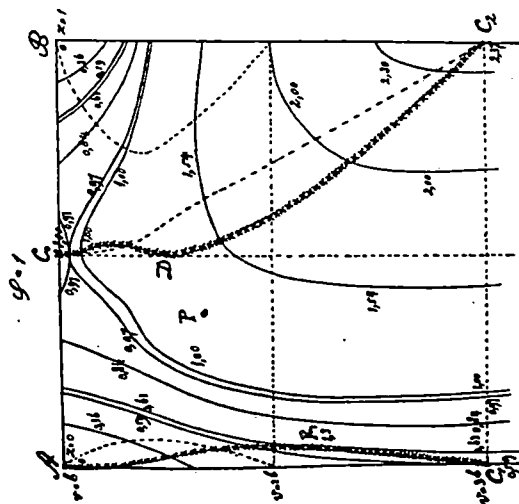


Fig. 1.

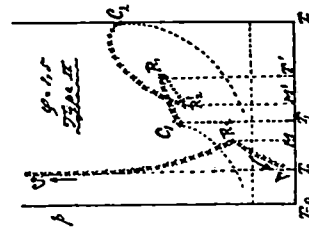


Fig. 3a.

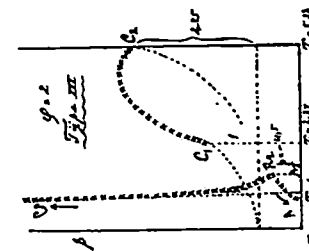


Fig. 2a.

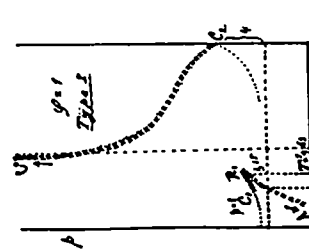
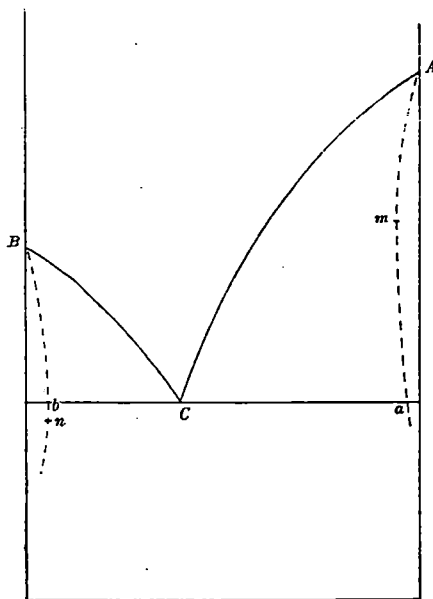


Fig. 1a.

Verlag der Abteilung Naturk. DI. XIV. A. 1905, II.

#### 5.4 Maker of predictions: Retrograde Solubility (1908)

In his publications in 1903 in Reports of the Royal Academy of Sciences: 'On the possible forms of the melting point curve for binary mixtures of isomorphous substances I and II' (expanded in Archives du Musée Teyler, 1905) and at later at Bakhuis Roozeboom's request included in 'Zeitschrift für physikalische Chemie' in 1908, Van Laar predicted the form of the melting point curve which belongs to retrograde solubility.<sup>33</sup> In these articles Van Laar deduced among others, a formula for  $T=f(x')$  in which  $x$  is the molfraction and the accent refers to the solid state. He showed that these melting point lines have a maximum (turning point), and with an example he calculated that these maxima (turning point) were close to the vertical axis. In the article of 1908 he called these maxima (turning points): 'Merkwürdig'!



Retrograde solidus by Van Laar in 1903.<sup>34</sup>

During his research into the alloys of cadmium and zinc, in 1926, Jenkins found the experimental confirmation of Van Laar's prediction.<sup>35</sup>

#### 8. Autumn of his life

After his second wife's death in 1934, Van Laar had a Dutch housekeeper probably named Mrs. Koker, who looked after him until his own death. He only felt really at home in his study and had relatively few outings, although he took walks in the surrounding areas.<sup>36</sup> Sudden thoughts were jotted down on his cuffs or were written in the sand with his cane.<sup>37</sup>



Van Laar and probably Mrs. Koker botanizing  
in an Alpine pasture.<sup>38</sup>

On 22 April 1938 Van Laar wrote one of his last postcards to Jorissen:

'(...) You can see from all this, dear Jorissen, that the old Van Laar is played out. From now on I must live on a lower heart level and always take the greatest care and precaution. Until 1937 I was still 'young' (I was called a 'young man'!) - and now ... I won't say the word. It only happens to ships! Best wishes, (...)'.<sup>39</sup>

He became ill and died six month later on 9 December 1938 at

Maison 'Les Tilleuls' in Tavel-sur-Clarens, in his second mother country. His death was reported in some of the Dutch daily papers. After his children and neighbours had paid their last respects, he was laid to rest in the 'Cimetiere de Clarens'. The letter of condolence of 14 December 1938 from Professor L.S. Ornstein (1880-1941) of Utrecht was exceptionally kind:

'Herewith I express my deepest sympathy at the death of your honoured father. I have always had great respect for his person and his work. He was a truly great man, who has long been misunderstood, but who bore this proudly. I feel gratitude for the many signs of sympathy I received from him. Our connection began in 1908 when I received a very angry letter from him about one of the propositions of my thesis. After my answer, however, this grew into a good friendship. When in 1911 in Utrecht I was passed over, he wrote me a letter which greatly encouraged me and after my appointment in 1914 in Utrecht he was among the first to congratulate me in his own kind fashion. His memory will, because of his work, live on for a long time; his friends, among whom I count myself, will keep it alive for as long as they live. Believe me, Yours faithfully, L.S. Ornstein'.<sup>40</sup>



L.S. Ornstein.<sup>41</sup>

### Conclusion

Ladies and Gentlemen, I have now reached the conclusion of my lecture.

Van Laar did not have an easy life. Inadequate preparatory education made it impossible for him to take academic exams. He was a very nervous man and in 1896 and 1911 he came close to mental collapse. During his life he met with opposition as well as appreciation. Van der Waals's theory on binary mixtures in 1889 and 1900 was a startingpoint for Van Laar to develop a part of phase theory and must be given the full credit for it. He was the first to provide a quantitative thermodynamic discussion of these binary mixtures. Together with Bakhuis Roozeboom he has to be seen as co-founder of phase theory. Examples of his work are remarkable, the 'melting point line of tin amalgam' (1902), the 'spinodal' (1905) and the 'retrograde solubility' (1908). Van Laar, as the theoretical designer of phase theory proved himself to be a dedicated arithmetician and scientist who was ahead of his time. It is a pity that Van Laar did not have better social skills and that Van der Waals never helped him. Credit and admiration are due to Lorentz, who always supported Van Laar. That Van Laar also experienced sympathy and friendship from others is illustrated by Ornstein's letter of condolence.

Thank you for your attention.



Notes

The following abbreviations have been used in the notes:

AHC Archives Historical Commission 'Koninklijke Nederlandse Chemische Vereniging'  
AL Archives H.A. Lorentz in the General State Archives, The Hague  
2.21.203, No.45.  
ARAG General State Archives, The Hague  
BML Boerhaave Museum Leiden  
GAU Town Archives, Utrecht  
IMG Letterkundig Museum The Hague  
RAH State Archives, Haarlem  
RAG State Archives, Groningen  
UMA University Museum Amsterdam

1. Photograph (GAU).
2. Photograph (UMA).
3. Letter J.J. van Laar to W. Kloos of 4 June 1935 (IMG); Information Mrs. L. van Laar and Mrs. E.W van Laar Amsterdam.
4. Photograph: Bakhuis Roozeboom Institute IJmuiden.
5. Minutes of Faculty in Groningen of 24 January 1914 (No.623 RAG).
6. Minutes of Faculty in Groningen of 5 February 1914 (No.623 RAG).
7. Letter J.J. van Laar to Senate in Groningen of 11 April 1914 (No.369 RAG) and to Faculty in Groningen of 15 April 1914 (No.629 RAG).
8. Letter J.J. van Laar to W.P. Jorissen, 13 June 1914 (AHC).
9. Photograph (Mrs L. van Laar, Amsterdam).
10. Letter J.J. van Laar to H.A. Lorentz, 3 April 1901 (AL).
11. Letter J.J. van Laar to H.A. Lorentz, 18 August 1899 (AL).
12. Minutes of the meeting of the Royal Academy of Science (Department of Physics) of 19 December 1903 (64 No.11 RAH).
13. Letter H.W. Bakhuis Roozeboom to H.A. Lorentz, 2 December 1904 (AL).
14. Letter F.A.H. Schreinemakers to F.G. Waller of 10 May 1928 (Archives Department of Education, Arts and Sciences 2.14.20, Nr.345, ARAG).
15. Photograph (BML).
16. J. de Boer, Van der Waals in his time and the present revival opening address, *Physica* 73 (1974) 1-27.
17. J.J. van Laar, J.D. van der Waals, in: J. Kalff, *Mannen en Vrouwen van Betekenis in onze Dagen*, Haarlem: Tjeenk Willink (1900).
18. J.J. van Laar, *Lehrbuch der mathematischen Chemie*. Leipzig: Barth (1901) VII.
19. Postcard J.J. van Laar to H.A. Lorentz of 10 July 1899 (AL).
20. Letter J.J. van Laar to H.A. Lorentz of 16 August 1899 (AL).
21. Concept of H.A. Lorentz to J.J. van Laar of 17 August 1899 (AL).
22. Letter J.J. van Laar to H.A. Lorentz of 18 August 1899 (AL).
23. Ibid., 21.
24. The course of the meltingpoint-lines of solid alloys or amalgams, *Proceedings Royal Academy* 5 (1902/03) 424-430;
25. Ibid.
26. H.A.J. Oonk, *Phase Theory*, Amsterdam: Elsevier (1981), p. 170-174.
27. Ibid. 24.
28. H.W. Bakhuis Roozeboom, *Die heterogenen Gleichgewichte*, Braunschweig: Vieweg (1904).
29. J.J. van Laar, An exact expression for the course of spinodal curves and their plait-points for all temperatures, in the case of mixtures of normal substances, *Proceedings Royal Academy* 7 (1904/05) 646-657; On the shape of the plaitpoint-curve for mixtures of normal substances, Ibid. 8 (1905/06) 33-48 and *Les courbes de plissement et leur point double chez les mélanges de substances normales, dans le cas que les volumes moléculaires sont inégaux*, *Archives du Musée Teyler* (2) X (1905) 19-44.
30. Letter J.J. van Laar to H.A. Lorentz, 26 March 1905 (AL).
31. P.H.E. Meijer, The van der Waals equation of state around the van Laar point, *J. Chem. Phys.* 90 (1988) 448-456.
32. Ibid. 27.

33. J.J. van Laar, On the possible forms of the meltingpoint-curve for binary mixtures of isomorphous substances I and II, Proceedings Royal Academy 6 (1903/04) 151-169 and 244-259; Sur les allures possibles de la courbe de fusion de mélanges binaires de substances isomorphes, Archives du Musée Teyler (2) VIII (1904) 517-580 and Die Schmelz- oder Erstarrungskurven bei binären Systemen, wenn die feste Phase ein Gemisch (amorphe feste Lösung oder Mischkrystalle) der beiden Komponenten ist, Zeitschrift für physikalische Chemie 63 (1908) 216-253.
  34. Sur les allures possibles de la courbe de fusion de mélanges binaires de substances isomorphes, Archives du Musée Teyler (2) VIII (1904) 545.
  35. C.H.M. Jenkins, The constitution and the physical properties of the alloys of Cadmium and Zinc, Journal Inst. Metals 36 (1926) 63-93.
  36. Ibid. 3.
  37. Honderd jaar geleden werd een groot Hagenaar geboren, Het Vaderland van 14 July 1960 and Information Mrs. L. van Laar and Mrs. E.W. van Laar, Amsterdam.
  38. Photograph (Mrs.L. van Laar, Amsterdam).
  39. Postcard J.J. van Laar to W.P. Jorissen of 22 April 1938. (BML).
  40. Letter L.S. Ornstein, Utrecht of 14 December, 1938 (Mrs.E.W. van Laar, Amsterdam).
  41. Photograph (MBL).
-