## 0- $\pi$ transition in superconductor-ferromagnet-superconductor junctions with strongly spin-dependent scattering

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We develop a theory of the critical current in superconductor-ferromagnetic alloy-superconductor trilayers, which takes into account the strong spin dependence of electron scattering on compositional disorder in a diluted ferromagnetic alloy. We show that in such a system the critical current oscillations as a function of the thickness of the ferromagnetic layer, with period of  $v_F/2I$ ,  $v_F$  and I being the Fermi velocity and exchange splitting, respectively, decay exponentially with a characteristic length of the order of the mean free path.

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The recent observation<sup>1-4</sup> of Josephson junctions with negative coupling,  $^{5,6}$  also known as  $\pi$  junctions, has attracted a lot of attention to hybrid superconductor-ferromagnetsuperconductor (SFS) structures. In contrast to conventional Josephson junctions, such as superconductor-normal metal systems, where the ground state corresponds to a superconducting phase difference  $\varphi$  of zero, the phase difference in a SFS trilayer can take both  $\varphi=0$  and  $\pi$  values, depending on the thickness of the ferromagnetic layer. Both 0 and  $\pi$  states in SFS trilayers have been deduced from the measurements of the density of states<sup>1</sup> and the critical current as a function of magnetic flux and temperature.<sup>2,4,7,8</sup> In particular, the critical current exhibits oscillations superimposed on the exponential decay as a function of the thickness of the ferromagnetic layer.<sup>3,9</sup> The decay length  $\xi_d$  and the period  $\xi_o$  of these oscillations have been measured, providing comparable yet unequal experimental values of these two parameters.

In ballistic SFS structures, the critical current is expected to oscillate with the period  $\xi_o = v_F/2I$ . For a ferromagnetic metal with a strong exchange splitting I, fluctuations of the width of the ferromagnetic layer suppress the appearance of the proximity effect, despite the fact that in ballistic structures Cooper pairs decay with the distance according to a power law rather than exponentially. Moreover, it has been shown<sup>10</sup> that, when the electron motion in a ferromagnetic film with large I is diffusive, the randomization of the oscillation phase over paths of different lengths leads to the exponential suppression of proximity at the scale of the mean free path:  $\xi_d \sim l$  for the case of  $I\tau \gg 1$  (where  $\tau$  is the electron mean free path). To enhance the proximity effect in SFS multilayers, one may want to use weakly ferromagnetic alloys, where the exchange field I is reduced by diluting the magnetic component. The analysis of diluted systems with  $I\tau \ll 1$  based upon modeling disorder in SFS junctions as spin-independent impurities has shown that the decay length may be expected<sup>6,11</sup> to extend beyond the mean free path range, such that  $\xi_d \sim \xi_o = \sqrt{D/I}$ , where  $D = v_F^2 \tau / 3$ .

In this paper, we show that the possibility to prolong the extent of the superconducting proximity effect in SFS structures by making them of diluted magnetic alloys is strongly limited. Following the theory of suppression of superconductivity by magnetic impurities, <sup>12</sup> earlier theories <sup>13,14</sup> took into account the effect of magnetic disorder by including in the Usadel equation a weak Cooper pair relaxation described by

a phenomenological spin relaxation rate  $\tau_s^{-1}$ . Keeping in mind that even in a weak ferromagnet electron spin flip is an inelastic process and should be accompanied by the excitation of a magnon, we attribute the pair breaking in a ferromagnetic alloy to a giant magnetoresistance (GMR) type effect. As noticed in earlier GMR studies, 15,16 a feature of ferromagnetic alloys is that elastic electron scattering in them is strongly spin dependent. Indeed, one scattering event off strongly spin-dependent disorder, seen differently by spin-up and spin-down electrons, is enough to break a singlet Cooper pair. In such a case, the decay length of a Cooper pair is of the order of the mean free path,  $\xi_d \sim l$ . Since, in this case, the use of Usadel equations adopted in the previous studies of disordered SFS junctions<sup>6,11</sup> does not hold, here we employ a nonlocal approach based on solution of the Eilenberger equation 6,10,13,17 to describe  $0-\pi$  Josephson oscillations as a function of the thickness of the diluted ferromagnetic alloy layer.

To describe a dilute ferromagnetic alloy, we use the following Hamiltonian (a  $2\times 2$  matrix in the spin space), adopted 15 in GMR theory:

$$\mathcal{H} = \hat{p}^2 / 2m + V(\mathbf{r}) + \boldsymbol{\sigma} \mathbf{J}(\mathbf{r}), \tag{1}$$

where V and  $\mathbf{J}$  describe magnetic atoms embedded into a normal metal, and  $\boldsymbol{\sigma}$  is the vector of Pauli matrices. The average  $\langle \mathbf{J} \rangle = \mathbf{e}_z I$  determines the exchange splitting for conduction band electrons, and  $\langle V \rangle = 0$ . Since every magnetic atom produces both scalar V and exchange  $\mathbf{J}$  potentials, we use the following correlation functions for magnetic and nonmagnetic disorder:  $\langle V(\mathbf{r})V(\mathbf{r}')\rangle = (2\pi\nu\tau_V)^{-1}\delta(\mathbf{r}-\mathbf{r}')$ ,  $\langle J_\alpha(\mathbf{r})J_\beta(\mathbf{r}')\rangle = (2\pi\nu\tau_J)^{-1}\delta_{\alpha\beta}\delta(\mathbf{r}-\mathbf{r}')$ , and  $\langle V(\mathbf{r})J_\alpha(\mathbf{r}')\rangle = (2\pi\nu\tau_{\mathrm{mix}})^{-1}\delta_{\alpha z}\delta(\mathbf{r}-\mathbf{r}')$ .

The starting point for a quantitative description is the Eilenberger equation for the retarded component of the semi-classical Green's function,

$$v_F \mathbf{n} \partial_{\mathbf{r}} \check{\mathbf{g}} + \left[ -i\omega \tau^3 + iI\sigma^z + i\check{\Sigma}, \check{\mathbf{g}} \right]_{-} = 0, \tag{2}$$

$$\check{g} \equiv \begin{pmatrix} g & f \\ f^+ & -g \end{pmatrix}, \quad f \equiv \begin{pmatrix} 0 & f_{\uparrow\downarrow} \\ f_{\downarrow\uparrow} & 0 \end{pmatrix}, \quad \check{g}^2 = 1, \tag{3}$$

where  $(f^+)_{\alpha\beta}(\mathbf{r},t;\mathbf{n},\omega) = -[f_{\alpha\beta}(\mathbf{r},t;-\mathbf{n},-\omega)]^*$ ,  $\tau^3$  acts in the Nambu space,  $\mathbf{n} = \mathbf{p}/p$ , the self-energy has the form

$$i\check{\Sigma} = \frac{1}{2\tau_V} \langle \check{g} \rangle + \frac{1}{2\tau_{\text{mix}}} [\sigma^z, \langle \check{g} \rangle]_+ + \frac{1}{2\tau_J} \sigma^z \langle \check{g} \rangle \sigma^z,$$

and  $\langle \check{g} \rangle = \int \check{g} \ d^2 \mathbf{n} / 4\pi$  is the Green's function averaged over the momentum direction. For the weak proximity effect, Eq. (2) can be linearized around the zero-order Green's function  $\check{g}_0 = \tau^3$ . Performing the expansion up to first order, we obtain

$$v_F \mathbf{n} \partial_{\mathbf{r}} f - 2i\omega f + 2iI\sigma_z f + (\tau_V^{-1} + \tau_J^{-1}) f - (\tau_V^{-1} - \tau_J^{-1}) \langle f \rangle = 0.$$
(4)

The linearization of the Eilenberger equation and subsequent analysis are based upon the assumption of weak coupling between superconductors and the ferromagnet, which is realized, for instance, if these are separated by an opaque barrier with low transparency  $\Theta \leq 1$ . The appropriate boundary conditions have been derived by Zaitsey, <sup>18</sup>

$$\check{g}_{S}^{a} = \check{g}_{F}^{a} \equiv \check{g}^{a}, \quad \check{g}^{a} \{ (1 - \Theta)[1 - (\check{g}^{a})^{2}] + \Theta(\check{g}_{-}^{s})^{2} \} = \Theta \check{g}_{-}^{s} \check{g}_{+}^{s},$$

where  $\check{g}_i^{s/a} = [\check{g}_i(n_z) \pm \check{g}_i(-n_z)]/2$  (i = S and F for a superconductor and a ferromagnetic alloy, respectively),  $n_z > 0$ , where  $n_z$  is the projection of  $\mathbf{n} = \mathbf{p}/p$  onto the direction normal to the SF interface, and  $\check{g}_{\pm}^s = (\check{g}_S^s \pm \check{g}_F^s)/2$ . In the case of low transparency  $\Theta \ll 1$ , we find that in the first order in  $\Theta$ ,

$$\check{g}^a = \frac{\Theta}{4} [\check{g}_S^{(0)}, \check{g}_F^{(0)}]_-,$$

where  $\check{g}_S^{(0)}$  and  $\check{g}_F^{(0)}$  are the Green's functions in the two materials when those are detached  $(\Theta=0)$ . Together with Eq. (4) this gives us a closed set of equations.

It is convenient to represent the semiclassical Green's function  $f(\mathbf{n},z)$  as a combination of two functions of a positive argument  $n_z > 0$ :  $f_1(n_z,z) \equiv f(n_z,z)$  and  $f_2(n_z,z) \equiv f(-n_z,z)$ . In this representation the boundary conditions take the form

$$f_{1}(n_{z},0) - f_{2}(n_{z},0) = a_{L},$$

$$f_{1}(n_{z},d_{F}) - f_{2}(n_{z},d_{F}) = -a_{R},$$

$$a_{L/R} = -\Theta \Delta \exp(i\phi_{L/R}) / \sqrt{\Delta^{2} - \omega^{2}},$$
(5)

where  $d_F$  is the thickness of the ferromagnetic layer. The equations for  $f_1$  and  $f_2$  take the forms

$$n_z \partial_z f_1(n_z, z) + \lambda f_1(n_z, z) - \alpha \langle f(z) \rangle = 0$$

$$-n_z \partial_z f_2(n_z, z) + \lambda f_2(n_z, z) - \alpha \langle f(z) \rangle = 0, \tag{6}$$

where  $n_z > 0$  and  $\alpha = (\tau_J - \tau_V)/(\tau_V + \tau_J)$ ,  $-1 \le \alpha \le 1$ . Also,

$$\lambda = 1 - 2i(\omega - I\sigma_z)\tau \tag{7}$$

is a  $2 \times 2$  matrix acting on the  $2 \times 2$  matrix f, and  $\tau^{-1} = \tau_V^{-1} + \tau_J^{-1}$ . The averaged Green's function equals

$$\langle f(z) \rangle = \int_0^1 dn_z [f_1(n_z, z) + f_2(n_z, z)].$$
 (8)

In the case of a thick ferromagnetic layer, such that  $e^{-d_F/l} \ll 1$ , where  $l = v_F \tau$  is the mean free path, one can write down the formal solution of Eqs. (6) as

$$f_{1}(n_{z},z) = a_{L}e^{-\lambda z/n_{z}l} + \frac{\alpha}{l} \int_{0}^{z} e^{\lambda(z'-z)/n_{z}l} \langle f(z') \rangle \frac{dz'}{n_{z}} + \frac{\alpha}{l} \int_{0}^{d_{F}} e^{-\lambda(z+z')/n_{z}l} \langle f(z') \rangle \frac{dz'}{n_{z}},$$
(9a)

$$f_{2}(n_{z},z) = a_{R}e^{\lambda(z-d_{F})/n_{z}l} + \frac{\alpha}{l} \int_{z}^{d_{F}} e^{\lambda(z-z')/n_{z}l} \langle f(z') \rangle \frac{dz'}{n_{z}} + \frac{\alpha}{l} \int_{0}^{d_{F}} e^{\lambda(z'+z-2d_{F})/n_{z}l} \langle f(z') \rangle \frac{dz'}{n_{z}}.$$
 (9b)

The subsequent algebra includes adding and averaging Eqs. (9), which leads to the integral equation for  $\langle f \rangle$ . Having presented  $\langle f(z) \rangle$  as the sum

$$\langle f(z)\rangle = a_I h(z) + a_R h(d_F - z), \tag{10}$$

we find that the (matrix) function h(z) satisfies a Fredholm equation of the second type,

$$2h(z) = K(\lambda z) + (\alpha/l) \int_{0}^{d_{F}} \left[ G(\lambda | z - z'|) + G(\lambda (z + z')) + G(\lambda (2d_{F} - z - z')) \right] h(z') dz',$$
(11)

where 
$$G(z) = \int_0^1 n_z^{-1} e^{-z/n_z l} dn_z$$
;  $K(z) = \int_0^1 e^{-z/n_z l} dn_z$ .

Up to this point, we could still reduce our equations to Usadel equations provided the diffusion approximation holds,  $(1-\alpha)\|\operatorname{Im}\lambda\| \ll 1$ . In the rest of the paper, we work outside this regime and consider the ballistic situation. For  $\alpha=0$ , the exact solution of Eq. (11) is  $h(z)=K(\lambda z)/2$ . Generalizing, we find that in the ballistic case the solution is determined by the behavior of functions K and G which at  $z \gg l$  are  $K(z) \approx G(z) \approx e^{-z/l} l/z$ . Assuming that solution falls off exponentially as  $e^{-\lambda z/l}$ , one can see that in Eq. (11) the last term in the integral can be neglected everywhere except for a small region near the boundary,  $z=d_F$ . This enables us to split the solution of Eq. (11) into two parts,

$$h(z) = e^{-\lambda z/l} h_L(z) + e^{-\lambda (2d_F - z)/l} h_R(d_F - z).$$
 (12)

The first term is relevant everywhere and is the main term of the solution, whereas the second one is only important close to the boundary  $d_F$ - $z\sim l$ , when the exponents become of the same order. Each of the matrix functions  $h_i$  (i=L,R) satisfies the equation

$$2h_{i}(z) = S_{i}(\lambda z) + \frac{\alpha}{l} \int_{0}^{z} \widetilde{G}(\lambda(z-z'))h_{i}(z')dz' + \frac{\alpha}{l} \int_{0}^{\infty} e^{-2\lambda z'} \left[\widetilde{G}(\lambda(z+z'))h_{i}(z') + \widetilde{G}(\lambda z')h_{i}(z+z')\right]dz', \tag{13}$$

where  $\tilde{G}(\lambda z) = G(\lambda z)e^{\lambda z/l}$  and  $S_L(\lambda z) = K(\lambda z)\exp(\lambda z)$ .

Far from the left boundary,  $z \ge l$ , we parametrize  $h_L(z) = A(z)l/\lambda z$ ,  $1/\lambda = \lambda^{-1}$ . Substituting it into Eq. (13) and keeping the leading order in l/z, we obtain the equation for the diagonal matrix A,

$$\left(2 - \frac{\alpha}{\lambda} \ln 2\right) A(z) = 1 + \frac{\alpha}{\lambda} \xi + \frac{\alpha}{\lambda} \int_0^z \frac{A(z')}{z'} dz' + \frac{\alpha}{\lambda l} \int_0^z A(z - z') \tilde{G}(\lambda z') dz', \quad (14)$$

where the matrix  $\xi = \lambda \int_0^\infty h_L(z) e^{-2\lambda z} dz/l$  does not depend on z. The last term in Eq. (14) in the leading order in  $\ln^{-1} z$  is  $A(z)[\gamma + \ln(\lambda z/l)]$  with  $\gamma$  being Euler's constant. Subsequently, we obtain a differential equation for the function  $\int_0^z A(z') dz'/z'$ . The solution far from the boundaries reads

$$h_L(z) = A(z) \frac{le^{-\lambda z/l}}{\lambda z}, \quad A(z) = \frac{\delta(\alpha, \lambda)}{\left[2 - (\alpha/\lambda)(\gamma + \ln 2\lambda z/l)\right]^2},$$
(15)

where a constant  $\delta(\alpha, \lambda)$  is of order one; at  $\alpha = 0$  the exact solution gives  $\delta(0, \lambda) = 2$ . Numerical calculations show that  $\delta(\alpha, \lambda)$  is still close to 2 even for  $\alpha = 1$ .

Having solved the equation for  $h_L$ , we use it to determine the matrix function  $S_R$ , according to

$$S_R(z) = \frac{\alpha}{l} \int_0^{d_F} \widetilde{G}(\lambda(z+z')) [h_L(d_F - z')$$
$$-e^{-2\lambda z'/l} h_L(d_F + z')] dz',$$

and find the solution for the function  $h_R(z)$ .

Within the approximations used in the above analysis of the Eilenberger equation for the anomalous Green's function f, the Josephson current density in the SFS structure can be represented as

$$\mathbf{j} = -\pi e \nu v_F \int n(\omega) \operatorname{Re} \mathcal{I} \frac{d\omega}{2\pi}, \quad \mathcal{I} = \langle \mathbf{n} \operatorname{tr}(\hat{f}\hat{f}^{\dagger}) \rangle.$$
 (16)

Here,  $n(\omega)$  is the Fermi distribution function. Substituting the expressions for h(z) into Eq. (10) and Eqs. (9), we find

$$\mathcal{I} = \frac{\Theta^2 \Delta^2 \sin(\phi_L - \phi_R)}{\Delta^2 - \omega^2} \operatorname{tr} \frac{le^{-\lambda d_F/l}}{\lambda d_F} Z(\alpha, \lambda, d_F), \tag{17}$$

where the matrix function Z depends on  $d_F$  logarithmically and for  $\alpha=0$  equals  $Z(0,\lambda,d_F)=1$ . Generally, in the leading order in  $d_F^{-1}$  it becomes

$$Z = \frac{\alpha}{\lambda} \xi \left( 1 + \frac{\alpha}{\lambda} \xi \right) + \frac{\alpha^2}{\lambda} \zeta + \left( 2 - \frac{\alpha}{\lambda} (1 + \ln 2) \right) A(d_F),$$
(18)

where  $\xi = \lambda \int_0^\infty h_L(z) e^{-2\lambda z} dz/l$  and  $\zeta = \alpha^{-1} \lambda^2 d_F \int_0^\infty h_R(z) K(\lambda z) \times dz/l$ . For  $d_F \gg l$  the quantity  $\xi$  is constant, and A depends on it logarithmically. The quantity  $\zeta$  depends on  $d_F$  in the same way as A. For  $\Delta \tau \ll ||\lambda|| \sim 1$ , the calculation of the frequency integral leads to an expression that is most conve-

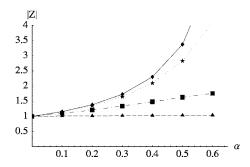


FIG. 1. Dependence of the factor |Z| determined in Eq. (18) on  $\alpha = (\tau_J - \tau_V)/(\tau_V + \tau_J)$  for various values of  $I\tau$ . From top to bottom:  $I\tau = 0, 0.3, 1, 10$ . The results show that outside the diffusive regime,  $I\tau \ll 1$  and  $\tau_J \gg \tau_V$ , Z is a smooth function of  $\alpha$  of order 1.

niently represented as the sum over Matsubara frequencies  $\omega_n = 2\pi T(n+1/2)$ . This is equivalent to the replacement  $i\omega \to \omega_n$  in the above expressions involving the matrix  $\lambda$ . As a result,  $\lambda$  becomes a diagonal matrix with two complex conjugate eigenvalues, so that in the Matsubara representation Z has the same property and can be written down as

$$Z = |Z| \exp(i\sigma_z \varphi_Z)$$
.

Note that for  $\omega_n \tau \lesssim \Delta \tau \lesssim 1$ , |Z| and  $\varphi_Z$  are two parameters of a structure independent of the Matsubara frequency. For  $\alpha = 0$ , one finds Z = 1. The dependence of |Z| on the value of the parameter  $\alpha$  is plotted in Fig. 1. Although the parameters |Z| and  $\varphi_Z$  depend on the quantities  $\alpha = (\tau_J - \tau_V)/(\tau_V + \tau_J)$ ,  $I\tau$ , and  $d_F/I$ , this fact does not qualitatively affect the results. Outside the regimes of  $I\tau \ll 1$  and  $\tau_J \ll \tau_V$  (where our results are not applicable) Z is a smooth function of  $d_F$  of order 1 that does not contain any dependence on scales of the order of  $\xi_d \sim l$  or  $\xi_o = v_F/2I$ .

Finally, we arrive at the expression for the critical current density  $[in j=j_c \sin(\phi_L-\phi_R)]$  which has the form

$$j_c = 2\pi e \nu v_F \Theta^2 |Z| \frac{e^{-d_F/l}}{d_F/l} T \sum_{\omega_n > 0} \frac{e^{-2\omega_n d_F/v_F}}{1 + \omega_n^2/\Delta^2} \times \frac{\cos[2Id_F/v_F + \arctan(2I\tau) - \varphi_Z]}{\sqrt{1 + (2I\tau)^2}}.$$
 (19)

In the limiting cases, the summation over Matsubara frequencies  $\omega_n$  can be calculated explicitly. For a ferromagnetic layer with thickness much greater than the coherence length in the superconductor,  $d_F \gg v_F/\Delta$ , the sum equals  $2T \sinh^{-1}(2\pi T d_F/v_F)$ . In the opposite case of a thin layer,  $d_F \ll v_F/\Delta$ , one obtains  $(\Delta/4) \tanh(\Delta/2T)$ . At zero temperature, the sum can be converted into an integral which equals  $\mathrm{Ci}(a)\sin(a)+[\pi/2-\mathrm{Si}(a)]\cos(a)$ , where  $a=2\Delta d_F/v_F$ , and the functions Si and Ci are sine and cosine integrals, respectively. For high temperature,  $d_F \gg v_F/T$ , only the lowest frequency is important, and the sum equals  $T(1+\pi^2T^2/\Delta^2)^{-1}\exp(-2\pi T d_F/v_F)$ .

The critical current dependence on the ferromagnetic layer thickness described in Eq. (19) for a weakly ferromagnetic layer with  $d_F > l$  is shown in Fig. 2. Even when dilution of the ferromagnetic layer is such that the exchange energy

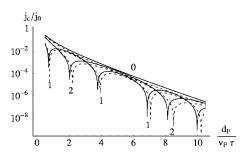


FIG. 2. Dependence of the critical current in a SFS trilayer on ferromagnetic layer thickness. Here, we normalize the current using a notional  $j_0=2\pi e v v_F \Theta^2 \Delta$ , and show it for T=0,  $\Delta \tau=0.1$  and several values of parameters  $\alpha$  and  $I\tau$ .  $\alpha=0$  (dashed lines) and 0.3 (solid lines). Dips in the value of  $j_c$  indicate positions where it disappears and changes sign, thus resulting in a sequence of  $0-\pi$  transitions. Neighboring dips always correspond to the same value of  $I\tau$ , demonstrating only weak dependence of the results on the parameter  $\alpha$ . For comparison, we also show the decay of the Josephson proximity effect in a SNS structure heavily doped by magnetic scatterers ( $I\tau=0$ ).

in it is weak,  $I\tau \ll 1$ , oscillations of  $j_c$  as a function of the layer thickness, with the period of  $\xi_o = v_F/2I$ , decay exponentially at the length scale of the mean free path,  $\xi_d = l$ —similarly to what happens in a disordered ferromagnetic layer with a strong exchange  $I(I\tau \gg 1)$ . Our results for  $I\tau \gg 1$  coincide with those of Ref. 10: For strong fields, the phase randomization of the order parameter is effective irrespective of the nature of scatterers. The dependence of the critical current on the thickness of the ferromagnetic layer in

Eq. (19) resembles the experimentally observed suppression of the proximity effect, at a length scale comparable to the mean free path measured in the same material.<sup>2</sup> Note that theories involving generation of the triplet order parameter due to nonuniform (spiral) magnetization in the ferromagnet<sup>19</sup> end up with the opposite conclusion, predicting weaker decay of the order parameter.

In conclusion, we developed a theory of the proximity effect in a superconductor-weakly ferromagnetic GMR alloy-superconductor trilayers, which takes into account the strong spin dependence of electron scattering on compositional disorder. The result, Eq. (19), describes the  $0-\pi$  transition for the Josephson effect as a function of the thickness of the ferromagnetic layer  $d_F$ : Oscillations occur with the period of  $\xi_0 = v_F/2I$  and exponential decay with the characteristic length  $\xi_d = l$  of the order of the mean free path, even in the regime when  $I\tau \ll 1$ . This result complements previous of the spin-singlet proximity effect superconductor-ferromagnet hybrid structures performed for ballistic and diffusive systems with spin-independent scattering<sup>6,10,13</sup> as well as theories of the suppression of the order parameter oscillations caused by interfaces.<sup>20</sup>

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