Paramagnetic intrinsic Meissner effect in layered superconductors

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Free energy of a layered superconductor with $\xi_\perp < d$ is calculated in a parallel magnetic field by means of the Gor'kov equations, where $\xi_\perp$ is a coherence length perpendicular to the layers and d is an interlayer distance. The free energy is shown to differ from that in the textbook Lawrence-Doniach model at high fields, where the Meissner currents are found to create an unexpected positive magnetic moment due to shrinking of the Cooper pair "sizes" by a magnetic field. This paramagnetic intrinsic Meissner effect in a bulk is suggested to detect, by measuring in-plane torque, the upper critical field and magnetization in layered organic and high-T_c superconductors, as well as in superconducting superlattices.

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The Meissner diamagnetic effect is known to be the most important property of a superconducting phase and is responsible for destruction of superconductivity both in type-I and type-II superconductors.[1] Meanwhile, as shown by us in Refs. 2–4 and as shown independently by Tesanovic *et al.*,[5] quantum effects of an electron motion in a magnetic field result in the appearance of a qualitatively different phenomenon—superconductivity surviving in high magnetic fields in layered[2–4] and isotropic three-dimensional (3D) (Ref. 5) type-II superconductors. In particular, it was shown[2–4,6] that in a layered conductor in a parallel magnetic field, where the Landau level quantization is impossible, some other quantum effects—the Bragg reflections—play an important role. These quantum effects result in a "two dimensionalization" (i.e., 3 D$\rightarrow$2 D crossover) of an open electron spectrum in an arbitrary weak parallel magnetic field. This is known[6,7] to cause the field-induced spin-density-wave (FISDW) and field-induced charge-density-wave (FICDW) instabilities in layered quasi-one-dimensional (Q1D) conductors. More complicated 3 D$\rightarrow$1 D$\rightarrow$2 D crossovers are shown[8,9] to be responsible for the experimentally observed nontrivial angular magnetic oscillations in a metallic phase of different layered organic conductors, including Lee-Naughton-Lebed[8] and Lebed magic angle[9] oscillations.

As shown in Refs. 2–4 and 7, the similar 3 D$\rightarrow$2 D crossovers have to be responsible for a stabilization of a superconducting phase in layered Q1D (Refs. 2 and 4) and quasi-two-dimensional (Q2D) (Ref. 3) conductors since two-dimensional (2D) superconductivity is not destroyed in a parallel magnetic field. More precisely, it is shown[2–4] that (i) the quantum effects make the upper critical field to be divergent, $H^{\parallel}_{c2}(T)\rightarrow\infty$ as $T\rightarrow 0$, and (ii) there is some critical field, H^*, above which superconducting temperature grows in an increasing magnetic field. Such superconducting phase with $dT_c/dH>0$ is called the re-entrant superconductivity (RS).[2–5] The original predictions[2–4] have been theoretically confirmed by a number of studies,[10–16] including Refs. 15 and 16, where paramagnetic Meissner effect was suggested for triplet Q1D superconductors. Despite the great success of 3 D$\rightarrow$1 D$\rightarrow$2 D and 3 D$\rightarrow$2 D crossover concepts in the explanations of magnetic properties in metallic,[7–9] the FISDW,[6,7] and the FICDW (Ref. 7) phases of organic conductors, so far there has been no evidence that superconducting temperature can grow in high magnetic fields due to the quantum 3 D$\rightarrow$2 D crossovers.[2–5,10–14] A possibility of the RS phase to exist was experimentally studied in Q1D layered organic superconductors $(TMTSF)_2X$ ($X=PF_6$ and $X=ClO_4$) by Lee *et al.*[17,18] and Jerome.[19] Their experiments gave hints on a possibility for superconductivity to significantly exceed the quasiclassical upper critical field $H^{\parallel}_{c2}(0)$—the effect predicted in Refs. 2–5 and 10–16. However, they were not able to confirm the appearance of the RS phase with $dT_c/dH>0$. Analogous experiments performed on a Q2D superconductor Sr_2RuO_4 (Ref. 20) did not detect any stabilization of superconductivity at $H>H^{\parallel}_{c2}(0)$.

The main obvious difficulty in the above-mentioned efforts to discover the RS phase is the Pauli spin-splitting destructive mechanism against superconductivity and the related Clogston paramagnetic limiting field, H_p.[1] It is absent only for some triplet superconducting phases, which are believed to exist in $(TMTSF)_2X$ (Refs. 4, 15, and 16) and Sr_2RuO_4 (Ref. 21) superconductors. On the other hand, recently there have appeared the NMR measurements[22] in favor of a singlet nature of superconductivity in $(TMTSF)_2ClO_4$ material, as well as some doubts[23] in a triplet nature of superconductivity in Sr_2RuO_4 one.

The goal of our Brief Report is a threefold one: First, we show that although in Q2D paramagnetically limited (singlet) superconductors the RS phase may not be characterized by $dT_c/dH>0$ feature,[3] nevertheless the RS phase reveals itself as another unique phenomenon—paramagnetic intrinsic Meissner effect (PIME). Second, we extend the microscopical theory[3] to describe the most important from an experimental point of view of d- and s-wave Q2D superconductors with $\xi_\perp<d$, where $\xi_\perp$ is a coherence length perpendicular to the conducting layers and d is an interlayer distance. Third, we suggest simple experimental methods to detect the PIME phenomenon in Q2D organic and high-T_c superconductors by using the in-plane torque, the upper critical field, and the magnetization measurements. In particular, we demonstrate that in-plane anisotropy due to anisotropic Ginzburg-Landau coherence lengths, which disappears in an intermediate region of magnetic fields (where the Lawrence-Doniach model is applicable), appears again in high magnetic fields as a consequence of the PIME phenomenon (see Figs. 1 and 2). We suggest to measure the in-plane anisotropy of the upper critical field and magnetization, as well as in-plane torque in high magnetic fields to discover the PIME and RS phenomena. For these purposes, we derive a free

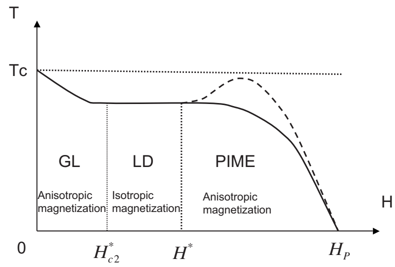

FIG. 1. Superconducting transition temperature in a parallel magnetic field for a paramagnetically limited Q2D superconductor is sketched. GL—area of applicability of the Ginzburg-Landau theory (Ref. 1). LD—area of applicability of the Lawrence-Doniach model (Refs. 27 and 28). PIME—area, where both the GL and LD descriptions are broken. In the latter case, which corresponds to the shrinking of the Cooper pair sizes by a magnetic field, our Eqs. (6)–(14) are still valid and the RS phase appears. The RS phase may reveal itself as an increase in the transition temperature in a magnetic field if the orbital effects of an electron motion are stronger than the Pauli spin-splitting effects (dashed line). The RS phase always reveals itself as a PIME, which results in unexpected in-plane anisotropy of the upper critical field and magnetization even in the case where the Pauli spin-splitting effects are strong, and thus, the area with $dT_c/dH>0$ is absent (solid line). We suggest to measure the in-plane torque, the upper critical field, and the magnetization to discover the RS phase.

energy of a Q2D superconductor with $\xi_\perp < d$ in a parallel magnetic field from the Gor'kov formulation[24–26] of the microscopic superconductivity theory. Our results coincide with that of the Lawrence-Doniach model[27,28] only at low enough magnetic fields, $H \ll H^*$, where the Meissner effect is diamagnetic. We show that at high magnetic fields, $H \sim H^* \leq H_p$, the field starts to shrink the Cooper pair sizes perpendicular to conducting layer directions due to 3 D $\rightarrow$2 D crossovers in a parallel magnetic field. The above-mentioned 3 D$\rightarrow$2 D crossovers of the Cooper pairs are not taken into account in the Lawrence-Doniach model and, as shown below, are responsible for the unique PIME phenomenon.

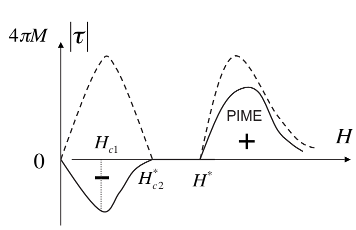

FIG. 2. Solid line: in-plane magnetization, $4\pi\mathbf{M}$, is sketched as a function of a magnetic field in the absence of the Pauli spin-splitting effects, where at high magnetic fields, $H \geq H^*$, the RS phase reveals itself as a PIME. Dashed line: an absolute value of in-plane torque, $|\tau|$, is sketched. It is important that the torque is independent of the Pauli spin-splitting effects since they are supposed to be isotropic. Therefore, even in the case where the destructive Pauli spin-splitting effects eliminate a positive sign of the Meissner effect in high magnetic fields, the PIME phenomenon and the RS phase can still be detected by the in-plane torque measurements.

Let us consider a layered superconductor with a Q2D electron spectrum,

$$\epsilon(\mathbf{p}) = \epsilon_\parallel(p_x,p_y) + 2t_\perp \cos(p_z d), \quad t_\perp \ll \epsilon_F, \tag{1}$$

in a parallel magnetic field,

$$\mathbf{H} = (0,H,0), \quad \mathbf{A} = (0,0,-Hx), \tag{2}$$

where $\epsilon_\parallel(p_x,p_y) \sim \epsilon_F$ is an in-plane electron energy, $t_\perp$ is an overlapping integral of electron wave functions perpendicular to the conducting plane directions, and ϵ_F is the Fermi energy. Electron spectrum [Eq. (1)] can be linearized near the 2D Fermi surface (FS), $\epsilon_\parallel(p_x,p_y)=\epsilon_F$, in the following way:

$$\epsilon(\mathbf{p}) - \epsilon_F = v_x(p_y)[p_x - p_x(p_y)] + 2t_\perp \cos(p_z d), \tag{3}$$

where $v_x(p_y)=\partial\epsilon_\parallel(p_x,p_y)/\partial p_x$ is a velocity component and $p_x(p_y)$ is the Fermi momentum along the x axis.

In the gauge [Eq. (2)], electron Hamiltonian in a magnetic field can be obtained from Eq. (3) by means of the Peierls substitution method, $p_x \rightarrow -i(d/dx)$ and $p_z \rightarrow p_z + (e/c)Hx$.[6] Therefore, electron Green's functions in a magnetic field satisfy the following differential equation:

$$\left\{ i\omega_n - v_x(p_y)\left[-i\frac{d}{dx} - p_x(p_y)\right] + 2t_\perp \cos\left(p_z d + \frac{eHdx}{c}\right) + 2\mu_B H s \right\} G_{i\omega_n}(x,x_1;p_y,p_z;s) = \delta(x-x_1), \tag{4}$$

where ω_n is the Matsubara frequency,[24] μ_B is the Bohr magneton, and $s=\pm\frac{1}{2}$ is an electron-spin projection along the quantization y axis—$\hbar\equiv 1$. It is important that Eq. (4) can be solved analytically. As a result, we obtain

$$G_{i\omega_n}(x,x_1;p_y,p_z;s) = -i\frac{\mathrm{sgn}\,\omega_n}{v_x(p_y)}\exp\left[-\frac{\omega_n(x-x_1)}{v_x(p_y)}\right]\exp[ip_x(p_y) \times (x-x_1)]\exp\left[\frac{2i\mu_B s H(x-x_1)}{v_x(p_y)}\right] \times\exp\left\{\frac{i\lambda(p_y)}{2}\left[\sin\left(p_z d + \frac{eHdx}{c}\right) - \sin\left(p_z d + \frac{eHdx_1}{c}\right)\right]\right\}, \tag{5}$$

where $\lambda(p_y)=4t_\perp c/ev_x(p_y)Hd$.

Linearized gap equation determining superconducting transition temperature, $T_c(H)$, can be derived using Gor'kov equations for nonuniform superconductivity.[3,25,26] As a result, we obtain

$$\Delta(x) = V\oint \frac{dl}{v_\perp(l)}\int_{|x-x_1|>|v_x(l)|/\Omega}^{\infty} dx_1 \frac{2\pi T}{v_x(l)\sinh\left[\frac{2\pi T|x-x_1|}{v_x(l)}\right]} \times\cos\left[\frac{2\mu_B H(x-x_1)}{v_x(l)}\right] J_0\left\{2\lambda(l)\sin\left[\frac{eHd(x-x_1)}{2c}\right]\sin\left[\frac{eHd(x+x_1)}{2c}\right]\right\}\Delta(x_1), \tag{6}$$

where integration in Eq. (6) is made along the 2D contour, $\epsilon_{\parallel}(p_x,p_y)=\epsilon_F$, $v_{\perp}(l)$ is a velocity component perpendicular to the 2D FS, V is an effective electron-electron interaction constant, and Ω is a cut-off energy. [Note that although Eq. (6) is derived for singlet s-wave superconductors, it is also valid for d-wave superconductors[29] if we redefine properly anisotropic coherence lengths and the effective interaction constant V.]

We point out that Eq. (6) is the most general one among the existing equations to determine the parallel upper critical field in a layered superconductor. In particular, it takes into account the Bragg reflections and related 3 D→2 D crossovers of electrons, which move in the extended Brillouin zone in a parallel magnetic field. As shown in Ref. 7, the above-mentioned quantum effects result in a momentum quantization law for an electron momentum component along the x axis. This is the reason why the kernel of the integral Eq. (6) is periodic[2,3] with respect to variables x and x_1. In the case where the destructive Pauli spin-splitting effects against superconductivity are absent [i.e., at $\mu_B=0$ in Eq. (6)], Eq. (6) possesses a periodic solution for $\Delta(x)$ at any magnetic field. In this case, superconductivity is stable in an arbitrary strong magnetic field and exists at high fields in the form of the RS phase with $dT_c/dH>0$. In the case of a singlet superconductivity, which is considered in the Brief Report, the Pauli spin-splitting effects may eliminate the superconductivity with $dT_c/dH>0$. Nevertheless, in the latter case, the RS phase reveals itself as an unusual anisotropy of the upper critical field and magnetization in high magnetic fields, $H \geq H^* \sim (t_{\perp}/T_c)^{1/2}\phi_0/\xi_x d \ll H_p$ (see Figs. 1 and 2).

As the most general equation, Eq. (6) contains the Ginzburg-Landau and Lawrence-Doniach descriptions as its limiting cases at low enough magnetic fields, $H \ll H^*$. For the so-called Josephson coupled layered superconductors with $\xi_{\perp}<d$,[27,28] Eq. (6) may be simplified and rewritten in the following differential form:

$$\left[\frac{T_c-T}{T_c} - 2.1\left(\frac{\mu_B H}{\pi T_c}\right)^2 + \xi_x^2\frac{d^2}{dx^2} - A(H) + B(H)\cos\left(\frac{2x\omega_c}{v_F}\right)\right]\Delta(x)=0, \tag{7}$$

with

$$A(H)=\frac{8t_{\perp}^2}{\omega_c^2}\left\langle\left[\frac{v_F}{v_x(l)}\right]^2\int_0^{\infty}\frac{dz}{\sinh(z)}\sin^2\left[\frac{\omega_c}{4\pi T_c}\frac{v_x(l)}{v_F}z\right]\right\rangle, \tag{8}$$

and

$$B(H)=\frac{8t_{\perp}^2}{\omega_c^2}\left\langle\left[\frac{v_F}{v_x(l)}\right]^2\int_0^{\infty}\frac{dz}{\sinh(z)}\sin^2\left[\frac{\omega_c}{4\pi T_c}\frac{v_x(l)}{v_F}z\right]\times\cos\left[\frac{\omega_c}{4\pi T_c}\frac{v_x(l)}{v_F}z\right]\right\rangle, \tag{9}$$

where

$$\langle\ldots\rangle=\oint\frac{dl}{v_{\perp}(l)}(\ldots)\Big/\oint\frac{dl}{v_{\perp}(l)}. \tag{10}$$

[Here, $\omega_c=eHv_Fd/c$ is a characteristic frequency of an electron motion along open FS [Eq. (1) and Ref. 3] and $\xi_x=\sqrt{7\zeta(3)}\langle v_x^2(l)\rangle^{1/2}/4\pi T_c$ is an in-plane anisotropic Ginzburg-Landau coherence length, $\mu_B H\simeq\omega_c(H)\ll\pi T_c$.]

Note that Eqs. (7)–(10) extend the Lawrence-Doniach model[27,28] to the case of strong magnetic fields and can be called the extended Lawrence-Doniach equations. In contrast to the traditional Lawrence-Doniach equations, the coefficients $A(H)$ and $B(H)$ in Eqs. (7)–(10) depend on a magnetic field, which means that the probability for the Cooper pair to jump from one conducting layer to another depends on the field. This important feature of Eqs. (7)–(10) is a consequence of the shrinking of the Cooper pair sizes due to 3 D→2 D crossover in a parallel magnetic field.[2,3,7]

Below, we are interested in the descriptions of the RS and PIME phenomena, therefore, we consider Eqs. (7)–(10) at high magnetic fields. It is possible to show that at $H\geq H^*$ the solution of Eq. (7) can be represented as $\Delta(x)=\Delta=\text{const}$, which corresponds to the RS phase.[2,3] In this case, the corresponding second-order term of a free energy with respect to the order parameter Δ can be written in the following simple form:

$$F^2(T,H)=-N(\epsilon_F)\left[\frac{T_c(H)-T}{T_c}\right]\Delta^2, \tag{11}$$

with

$$T_c(H)=T_c-2.1\frac{(\mu_B H)^2}{\pi^2 T_c}-2.1\frac{t_{\perp}^2}{\pi^2 T_c}+0.95\frac{t_{\perp}^2}{\pi^2 T_c}\left(\frac{eHd}{c}\right)^2\xi_x^2, \tag{12}$$

where $N(\epsilon_F)$ is a density of states per one electron-spin projection at $\epsilon=\epsilon_F$.

Note that the first term in Eq. (12) describes the destruction of a singlet superconductivity by the Pauli spin-splitting effects, whereas the last term in Eq. (12) is responsible for the restoration of superconductivity at high magnetic fields and for the appearance of the RS phase and the PIME phenomenon. If we take into account that the fourth-order term of a free energy with respect to the order parameter Δ can be calculated at $H\geq H^*$ in a standard manner[25] $F^4=7\zeta(3)N(\epsilon_F)\Delta^4/16\pi^2T_c^2$, then we can minimize the total free energy and find that

$$\delta F(T,H)=-\frac{4\pi^2}{7\zeta(3)}N(\epsilon_F)[T_c(H)-T]^2. \tag{13}$$

Magnetization can be found by a differentiation of the free energy with respect to the magnetic field,

$$M(T,H)=\frac{8}{7\zeta(3)}N(\epsilon_F)\left(\frac{T_c-T}{T_c}\right)\left[-4.2\mu_B^2+1.9\left(\frac{et_{\perp}d\xi_x}{c}\right)^2\right]H + M_0, \tag{14}$$

where M_0 is a magnetization in a metallic phase. Equations (12)–(14), which are valid at $H^*\leq H\leq H_p$, are the main results of the Brief Report. Note that in Eq. (14), the first term corresponds to a destruction of superconductivity due to the

Pauli spin-splitting effects, whereas the second term represents unusual paramagnetic orbital contribution to a magnetic moment (i.e., the PIME phenomenon). It is important that ξ_x in Eqs. (12)–(14) is anisotropic and depends on a direction of a magnetic field since it is an in-plane component of a coherence length perpendicular to the field. Therefore, the RS and PIME effects in Eqs. (12)–(14) can be detected by measuring a torque, provided that spin-splitting effects are isotropic.

In conclusion, we discuss possible experiments to discover the PIME and RS phenomena. The most direct method is to create such layered superconducting superlattice, where $\omega_c(H) \gg \mu_B H$.[30] The latter condition means that the orbital effects are more important than the Pauli spin-splitting ones. Therefore, in the above-mentioned case, the increase in transition temperature [Eq. (12)] and the paramagnetic Meissner effect [Eq. (14)] can be directly observed. Nevertheless, in most real physical compounds with $\xi_\perp < d$ and $\omega_c(H) \simeq \mu_B H$, the PIME [Eq. (14)] and RS [Eq. (12)] phenomena can only be observed indirectly—by measurements of anisotropies of the in-plane upper critical field [Eq. (12)] and magnetization [Eq. (14)], as well as by measurements of in-plane torque. In our opinion, the most perspective superconductors for indirect observations of the PIME phenomenon in steady magnetic fields are organic compounds α-$(ET)_2NH_4Hg(SCN)_4$, κ-$(ET)_2Cu(NCS)_2$, κ-$(ET)_2Cu[N(CN)_2]X$, α-$(ET)_2KHg(SCN)_4$, and λ-$(BETS)_2FeCl_4$.[31] The above-mentioned studies of the in-plane anisotropies can also be performed in high-temperature superconductor $Y_1Ba_2C_3O_7$, but it will require ultrahigh pulsed magnetic fields.

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[29] In this Brief Report, we calculate the paramagnetic Meissner moment and its two-fold anisotropy. We disregard a weak four-fold anisotropy due to a d-wave gap since it is $(t_\perp / T_c)^2 \ll 1$times smaller than the above calculated two-fold one.

[30] This idea belongs to P. M. Chaikin (private communication).

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