

Pair-density wave states through spin-orbit coupling in multilayer superconductors

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Spin-singlet superconductors with quasi-two-dimensional multilayer structure are studied in high magnetic fields. Specifically, we concentrate on bi- and trilayer systems whose layers by symmetry are subject to Rashba-type spin-orbit coupling. The combination of magnetic field and spin-orbit coupling leads to a first-order phase transition between different states of layer-dependent superconducting order parameters upon raising the magnetic field. In this context, we distinguish the low-field Bardeen-Cooper-Schrieffer state where all layers have order parameters of the same sign and the high-field pair-density wave state where the layer-dependent order parameters change the sign at the center layer. We also show that progressive paramagnetic limiting effects yield additional features in the H - T phase diagram. As possible realizations of such unusual superconducting phases we consider artificial superlattices of CeCoIn₅ as well as some multilayer high- T_c cuprates.

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I. INTRODUCTION

Time reversal and inversion are two of the most important symmetries for the formation of Cooper pairs in a superconductor. Interesting new features of superconductivity arise when one or both of them are missing. One intriguing phase in the absence of time reversal symmetry is the so-called Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state—a spatially modulated superconducting condensate induced by the spin-splitting of the Fermi surface due to high magnetic fields.^{1,2} This phase has been discussed in many contexts, for condensed matter systems^{3,4} and ultra-cold atomic gases⁵ as well as in nuclear physics.⁶ The other interesting class is represented by superconductors without inversion center in the crystal lattice. These so-called noncentrosymmetric superconductors are also characterized by spin-splitting of the Fermi surface, in this case, due to antisymmetric spin-orbit coupling, and form Cooper pairs of mixed parity.⁷ Interestingly, both types of situations have been recently discussed in the context of Ce-based heavy-fermion superconductors. A phase observed in CeCoIn₅ at low temperatures and rather high magnetic fields is considered a possible candidate for an FFLO state.^{3,4,8,9} Most striking properties of unusually high upper critical fields have been reported for CeIrSi₃ and CeRhSi₃, which both have a noncentrosymmetric crystal lattice.⁷ In our study, we would like to consider a combined effect of spin splitting through magnetic fields and spin-orbit coupling in an artificially structured heavy-fermion superconductor based on CeCoIn₅.

Mizukami and his coworkers succeeded recently in fabricating superlattices of CeCoIn₅ and YbCoIn₅, the former a heavy-fermion and the latter an ordinary metal.¹⁰ Surprisingly, this system is superconducting, if the number of CeCoIn₅ layers within a unit cell exceeds two. In view of the fact that the upper critical field remains amazingly high despite the low transition temperature,¹⁰ the superconductivity in the superlattice is robust against the paramagnetic depairing effect. It was proposed that the superstructure of this material would have locally noncentrosymmetric features, while having global inversion centers and may lead to unusual properties of the superconducting phase.¹¹ Actually, the multilayered structure would induce a spatially dependent Rashba-type of spin-orbit

coupling,¹² which would influence the spin structure of the electronic bands and suppress the paramagnetic depairing effect.¹¹ A recent measurement of the angular variation of the upper critical field found evidence for such a behavior.¹³ In this study, we investigate an unusual form of the superconducting order parameter induced by the modulated Rashba spin-orbit coupling in the magnetic field. As it will turn out to involve a modulation on the length scale of lattice constant, the phase may be viewed as a pair-density wave (PDW) state. Here, we will restrict to models of bi- and trilayer systems as depicted schematically in Fig. 1.

II. FORMULATION

For our discussion, we analyze the following model of a multilayered system having the interlayer hopping and Rashba spin-orbit coupling:

$$\begin{aligned} H = & \sum_{\mathbf{k}, s, m} \xi(\mathbf{k}) c_{\mathbf{k}sm}^\dagger c_{\mathbf{k}sm} + t_\perp \sum_{\mathbf{k}, s, \langle m, m' \rangle} c_{\mathbf{k}sm}^\dagger c_{\mathbf{k}sm'} \\ & + \sum_{\mathbf{k}, \mathbf{k}', m} V(\mathbf{k}, \mathbf{k}') c_{\mathbf{k}\uparrow m}^\dagger c_{-\mathbf{k}\downarrow m}^\dagger c_{-\mathbf{k}'\downarrow m} c_{\mathbf{k}'\uparrow m} \\ & + \sum_{\mathbf{k}, s, s', m} \alpha_m \mathbf{g}(\mathbf{k}) \cdot \boldsymbol{\sigma}_{ss'} c_{\mathbf{k}sm}^\dagger c_{\mathbf{k}s'm} - \mu_B H \sum_{\mathbf{k}, s, m} s c_{\mathbf{k}sm}^\dagger c_{\mathbf{k}sm}, \end{aligned} \quad (1)$$

where m is the index of layers. Note that we restrict here to one set of superconducting layers and ignore the normal metal part of the superlattice, assuming that the coupling between heavy-fermion and normal metal part is small. This is a reasonable approximation for the superlattice of CeCoIn₅, since the large mismatch in the Fermi velocities between CeCoIn₅ and YbCoIn₅ at the interface suppresses the proximity effect.¹⁴ For the band structure, we assume a simple nearest-neighbor hopping tight-binding form $\xi(\mathbf{k}) = -2t(\cos k_x + \cos k_y) - \mu$ on a square lattice with the chemical potential $\mu/t = 2$, and a small interlayer hopping $t_\perp/t = 0.1$. The symbol $\langle m, m' \rangle$ denotes the summation over nearest-neighbor layers. It is important to note that the following results do not qualitatively depend on the details of the band structure.

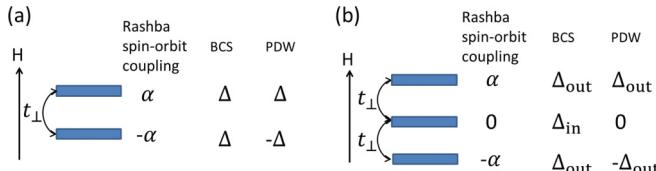


FIG. 1. (Color online) Schematic figures of (a) bi- and (b) trilayer systems. Thick bars describe the two-dimensional conducting planes that are coupled with each other through the interlayer coupling t_{\perp} . The structures of modulated Rashba spin-orbit coupling and superconducting order parameters in BCS and PDW states are shown in the right-hand side of each figure. See the text for details.

We also use an *s*-wave superconducting state by assuming $V(\mathbf{k}, \mathbf{k}') = -V$ (onsite attractive interaction) for simplicity. Although CeCoIn₅ is believed to be a *d*-wave superconductor and would have a small odd-parity component admixed with the local noncentrosymmetry, the features found below are independent of the pairing symmetry as long as spin-singlet pairing is predominant. In fact, we have confirmed that the model for *d*-wave superconductivity gives qualitatively same results (see Appendix). Hence, for the sake of numerical accuracy, we restrict ourselves to the simpler *s*-wave state. Our choice $V/t = 1.7$ gives rise to the critical temperature $k_B T_{c0}/t = 0.0255$ for both bilayer and trilayer systems with $\alpha = 0$, ignoring phase fluctuations.

For our purpose, it is sufficient to choose a *g* vector with Rashba spin-orbit structure, $\mathbf{g}(\mathbf{k}) = (-\sin k_y, \sin k_x, 0)$ without going into the microscopic details of the compound.¹⁵ The coupling constants α_m are layer dependent and antisymmetric with respect to reflection at the center of the multilayer structure. In this way, the existence of a global inversion center is guaranteed. For example, $(\alpha_1, \alpha_2) = (\alpha, -\alpha)$ for bilayers, and $(\alpha_1, \alpha_2, \alpha_3) = (\alpha, 0, -\alpha)$ for trilayers (see Fig. 1). Paramagnetic limiting and also eventually the PDW phase enter through the Zeeman coupling term, i.e., the last term in Eq. (1).

In our discussion, we restrict to magnetic fields along *c* axis and analyze the model on the basis of the Bogoliubov-de Gennes equation. While we include the layer dependence of order parameter $\Delta_m = -\sum_{\mathbf{k}} V \langle c_{-\mathbf{k}\downarrow m} c_{\mathbf{k}\uparrow m} \rangle$, we ignore the spatial modulation in the plane, such as the vortex state. This simplification is not crucial on a qualitative level, if we assume short coherence lengths and a large Ginzburg-Landau parameter κ , the ratio of coherence length and London penetration depth. Although phase fluctuations may play an important role for the critical temperature, we focus on the low-temperature and high-magnetic field phase and ignore this aspect.

III. SUPERCONDUCTING PHASES

A. Bilayer system

First, we address the results for the bilayer system. Figure 2 shows the T - H phase diagram for several values of α/t_{\perp} . For $\alpha/t_{\perp} = 0$ [see Fig. 2(a)], we obtain the conventional phase diagram of a superconductor subject to paramagnetic limiting.⁸ In this case, the order parameter is the same in both layers having the same phase, $(\Delta_1, \Delta_2) = (\Delta, \Delta)$. This state, which we call Bardeen-Cooper-Schrieffer (BCS) state, is stabilized basically by the interlayer ‘‘Josephson’’ coupling. The superconducting

phase transition turns first order at low temperatures because of the paramagnetic depairing effect. The in-plane FFLO phase with an inhomogeneous order parameter in the two-dimensional plane is supposed to appear around the first-order transition line in Fig. 2(a). However, in our approach we ignore the possibility of in-plane modulations as mentioned above.

We now turn to the role of the layer-dependent Rashba spin-orbit coupling. Interestingly, the order parameter configuration with a sign change, $(\Delta_1, \Delta_2) = (\Delta, -\Delta)$, becomes more stable at high magnetic fields for $\alpha/t_{\perp} \gtrsim 1$. This state is called the pair-density wave (PDW) state in Figs. 2(b)–2(d), since the order parameter modulates on length scales of the crystal lattice constant. The PDW state was proposed for the *Q*-phase in the bulk CeCoIn₅,^{16,17} but has not been established in microscopic models so far. With increasing spin-orbit coupling α the PDW state is stabilized in the large parameter range. Thus we here find that the PDW state may be realized in multilayer systems having ‘‘weak interlayer coupling’’ and ‘‘moderate spin-orbit coupling.’’ We would like to stress that the PDW state is stabilized in the purely superconducting multilayers, while a similar inhomogeneous superconducting state can be induced by the proximity effect in the ferromagnet-superconductor junction.¹⁸

The stability of the PDW state can be understood on the basis of the spin susceptibility and the band structure investigated in Ref. 11. In the absence of spin-orbit coupling the bilayer system forms two bands, the bonding band and antibonding band. Cooper pairs in the PDW state would form by interband pairing, in contrast to the BCS state, which is an intraband pairing state. Obviously, the PDW state would not be stable under these circumstances. On the other hand, the band structure is described by the two spin-split bands on each layer for large α/t_{\perp} . In this case, both BCS and PDW states occur as intraband Cooper pairing states. Although the zero-field transition temperature of the PDW state is lower than the onset of superconductivity (BCS state), a magnetic field along the *c* axis suppresses the BCS state more strongly through paramagnetic depairing. Indeed, the PDW state has a larger spin susceptibility than the BCS state.¹¹ Therefore the PDW state is more robust against paramagnetic limiting than the BCS state, as shown in Figs. 2(b)–2(d).

B. Trilayer system

New aspects can be found for the trilayer system. The order parameter has now three components, such that we distinguish the BCS state as $(\Delta_1, \Delta_2, \Delta_3) = (\Delta_{\text{out}}, \Delta_{\text{in}}, \Delta_{\text{out}})$ and the PDW state as $(\Delta_1, \Delta_2, \Delta_3) = (\Delta_{\text{out}}, 0, -\Delta_{\text{out}})$. For $\alpha/t_{\perp} \lesssim 2$, the phase diagram is qualitatively the same as shown in Fig. 2 for bilayer systems. The range of the PDW state is slightly reduced because of the complete suppression of order parameter in the inner layer. Increasing spin-orbit coupling to $\alpha/t_{\perp} = 3$ yields an intriguing modification of the phase diagram (see Fig. 3). A further first-order phase transition line appears within the BCS state, which is absent for bilayer systems.

In order to characterize the first-order phase transition in the BCS state, we show the magnetic field dependence of the order parameter in Fig. 4. At $H = 0$, the order parameter of the inner layer Δ_{in} is slightly larger than that of the outer layers Δ_{out} . Upon increasing the magnetic field for $T/T_{c0} = 0.04$

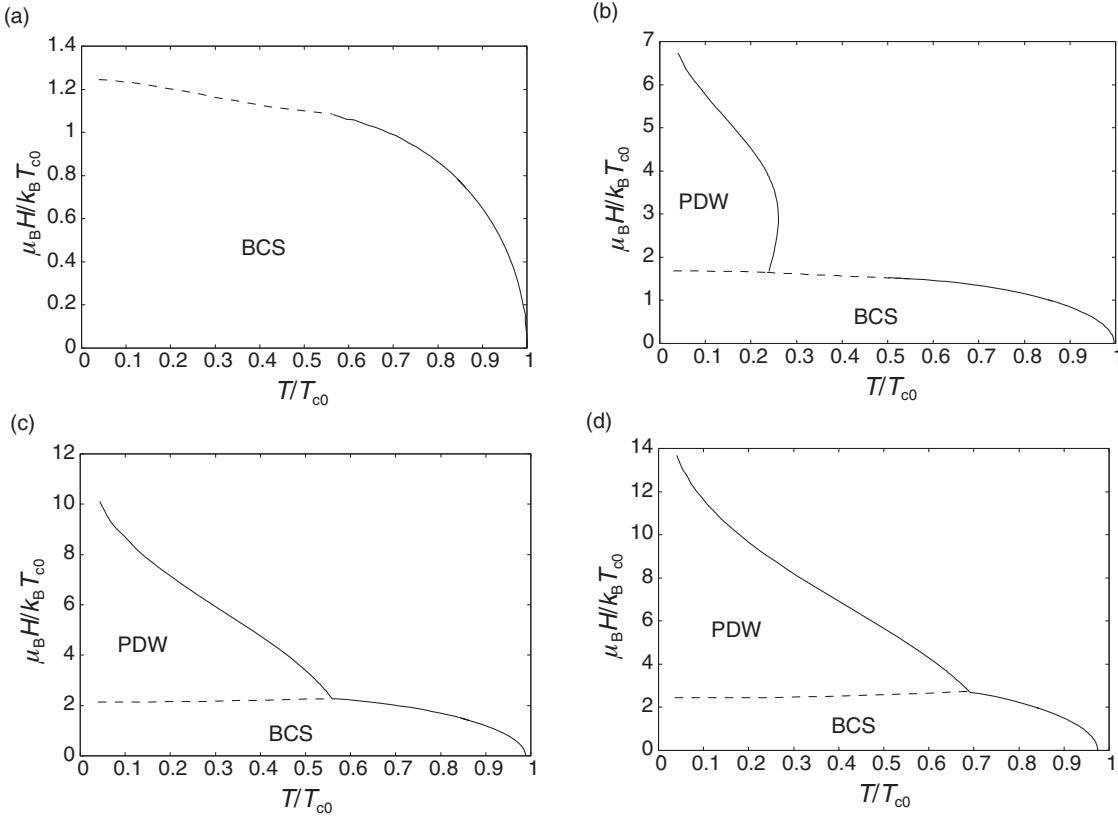


FIG. 2. T - H phase diagram of bilayer systems for (a) $\alpha/t_{\perp} = 0$, (b) $\alpha/t_{\perp} = 1$, (c) $\alpha/t_{\perp} = 2$, and (d) $\alpha/t_{\perp} = 3$. The solid and dashed lines show the second- and first-order phase transition lines, respectively. The “BCS” and “PDW” are explained in the text. The temperature T and magnetic field $\mu_B H$ in Fig. 2 are normalized by the critical temperature T_{c0} at $\alpha/t_{\perp} = 0$.

at $\mu_B H/k_B T_{c0} = 2.04$ an abrupt drop of Δ_{in} occurs, while Δ_{out} is only weakly affected [see Fig. 4(a)]. Indeed, with the layer-dependent spin-orbit coupling the paramagnetic limiting behavior is different for the inner and outer layers. The spin susceptibility in the superconducting phase is more strongly diminished for the inner than the outer layer when $\alpha/t_{\perp} > 1$, as the Rashba spin-orbit coupling vanishes by symmetry for the inner layer.¹¹ The high-field BCS state is stabilized by the more field resistant order parameter component Δ_{out} of the outer layer. Note that the interlayer hopping is responsible for

the fact that the order parameter components are not behaving independently. As the temperature is increased, the abrupt first-order phase transition in the BCS state turns into a crossover at the critical end point $T/T_{c0} \sim 0.12$. In this regime, the order parameter of the inner layer decreases continuously, as shown in Fig. 4(b).

The different phases also influence the quasiparticle spectrum, which we will discuss here looking at the density of states (DOS) layer-resolved for the trilayer system, as depicted in Fig. 5 (solid line for inner and dashed line for outer layers). The DOS of the outer layer always shows a gap at the Fermi energy. In the absence of a magnetic field, the DOS of both the inner and outer layers are essentially identical [see Fig. 5(a)]. Differences appear for finite fields. In Fig. 5(b), we show the DOS in the high-field BCS state, which is different for the inner and outer layers. The quasiparticle spectrum of the inner layer is gapless, although the order parameter Δ_{in} does not disappear. Surprisingly, a small gap on the inner layer opens up again when we enter the PDW state as revealed by Fig. 5(c), although the order parameter Δ_{in} vanishes in this case. The origin of this somewhat counterintuitive behavior lies in the interlayer hopping and the energy shifts due to the magnetic field leading to the pair breaking. The inner layer is obviously affected by paramagnetic limiting in the high-field BCS state yielding subgap quasiparticle states. On the other hand, in the PDW state the paramagnetic pair breaking effects almost disappear.¹¹ In this case, the intrinsic order parameter of the inner layer vanishes and a small gap appears in

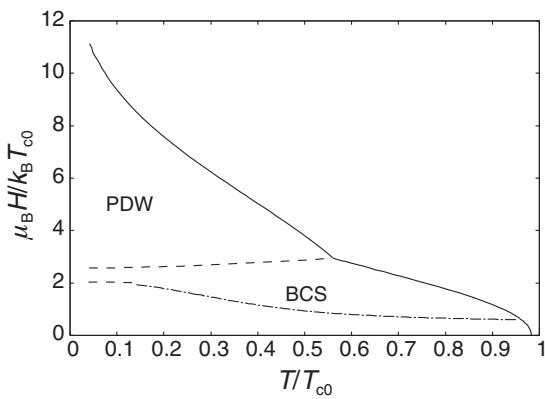


FIG. 3. T - H phase diagram of trilayer systems for $\alpha/t_{\perp} = 3$. The dash-dotted line shows the crossover field determined by the criterion $\Delta_{in} = \Delta_{out}$.

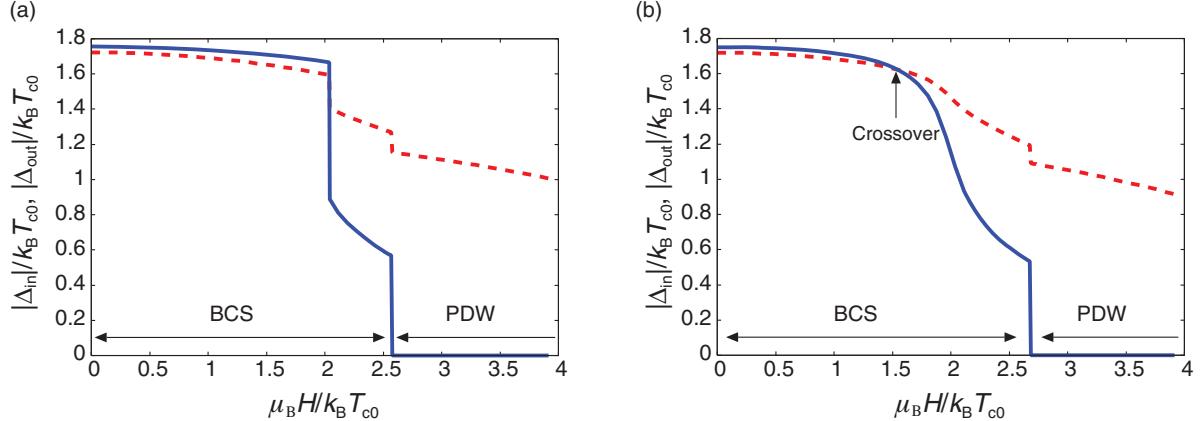


FIG. 4. (Color online) Magnetic field dependence of the order parameters in the inner layer Δ_{in} (solid lines) and outer layers Δ_{out} (dashed lines) for $\alpha/t_{\perp} = 3$. (a) $T/T_{c0} = 0.04$ and (b) $T/T_{c0} = 0.28$.

the quasiparticle excitations because the quasiparticles are extended over all layers due to the interlayer hopping.

IV. SUMMARY AND DISCUSSION

As mentioned earlier, possible realizations of the situation discussed here could be found in artificially grown superlattices of CeCoIn₅¹⁰ as well as in multilayer high- T_c

cuprates.^{19,20} These multilayer systems are quasi-two-dimensional superconductors and likely near the Pauli limit as they are rather robust against orbital depairing due to the very short coherence lengths.^{10,21} Consequently, they satisfy the condition for the phenomenology analyzed in our study. Indeed, the reduced paramagnetic limiting due to the Rashba spin-orbit coupling of the outer layers has been confirmed in recent experiments for superlattices CeCoIn₅.¹³

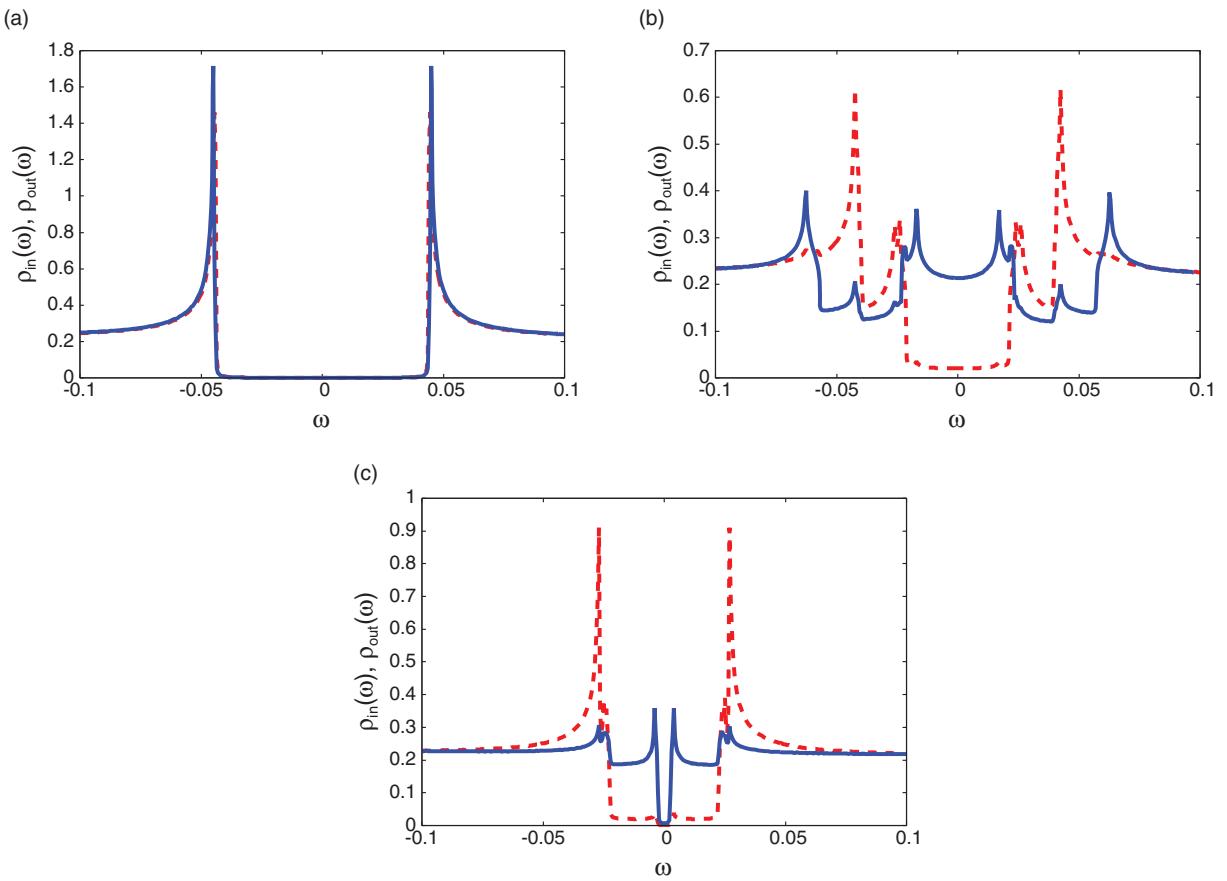


FIG. 5. (Color online) DOS in the inner layer $\rho_{\text{in}}(\omega)$ (solid lines) and outer layers $\rho_{\text{out}}(\omega)$ (dashed lines) at $T/T_{c0} = 0.04$. (a) $\mu_B H/k_B T_{c0} = 0$ (low-field BCS state), (b) $\mu_B H/k_B T_{c0} = 2.25$ (high-field BCS state), and (c) $\mu_B H/k_B T_{c0} = 3$ (PDW state), respectively. The other parameters are the same as Fig. 3.

The experiment is approaching to the FFLO state for the field along ab plane.¹³ Our study shows that the PDW state may be stabilized in the magnetic field along c axis. While this lies in the measurable range for the “low- T_c ” system CeCoIn₅, fields revealing the high-field BCS or PDW state are likely beyond 100 T in high- T_c cuprates.

Finally, we would like to comment on several points of our results. First, we restricted to pairing interaction in the spin-singlet channel. Spin-singlet pairing is essential for the observation of the phase diagram including different BCS states and the PDW state. If spin-triplet pairing compatible with the Rashba-spin-orbit coupling would be dominant, paramagnetic limiting would not play a role at all, since the spin susceptibility would keep the full value of the normal-state Pauli spin susceptibility for magnetic fields along the c axis.¹¹ Therefore no change of phase would be induced by a magnetic field. For the intermediate situation, if spin-singlet and spin-triplet pairings are of comparable strength, an intriguing mixing of the two channels is possible leading to a complex phase structure involving spontaneous time reversal symmetry breaking. This situation will be discussed elsewhere. Note, however, that the pairing interactions in the spin-triplet channel are expected to be weak in the bulk/superlattice of CeCoIn₅ and high- T_c cuprates. Even then, weak interactions in the spin-triplet channel gives rise to the uniform spin-triplet Cooper pairs in the PDW state.

Second, we focused on the Pauli-limited superconductor with large Maki parameter, neglecting orbital depairing effects, in particular, the aspect of a mixed phase including flux lines. Although orbital depairing may play quantitatively important roles for the field along c -axis, the PDW state survives moderate orbital depairing effects (moderate Maki parameters) because the spatial modulation of vortex state along the ab plane is not affecting the π -phase shift along c axis profoundly. The situation is quite different for the helical superconducting state in Rashba-type noncentrosymmetric superconductors, which is obscured even by the weak orbital depairing effect.²²

Third, the PDW state is stabilized not only in the bi- and trilayers but also in systems with more layers. In such systems, the layers towards the center are subject to paramagnetic limiting and it is expected that within the BCS state we encounter a progressive demolishing of superconductivity in these layers upon increasing field. Eventually, at high enough magnetic field, the superconducting system behaves like the bilayer system in the PDW state. Due to the fact that these outer layers are well separated the condition for the appearance of the PDW state is less stringent than in the real bilayer system. These views have been confirmed by us numerically for the four-layer system.

In conclusion, we have studied the superconducting state in multilayer systems including layer-dependent antisymmetric spin-orbit coupling due to the coordination of the layers. We found that in a magnetic field along the c -axis spin-orbit coupling influences the phase of the order parameters of different layers. We find a first-order phase transition separating two states different by symmetry: the high-field BCS state, which is even under reflection at the center of the multilayer system, and the PDW state, which is odd under reflection. The latter phase allows for a considerable

enhancement of the upper critical field and generates the uniform order parameter for the spin-triplet superconductivity. This phenomenology relies on the fact that orbital depairing is weak, i.e., the coherence length of the superconductor is very short, and that the dominant pairing is in a spin-singlet channel. This may apply to the artificially grown multilayer version of CeCoIn₅ and some multilayered high- T_c cuprates.

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APPENDIX: CASE OF THE d -WAVE SUPERCONDUCTIVITY

Here, we show the phase diagram for d -wave superconductivity using a linearized gap equation. We define the superconducting susceptibility $\chi_{m_1 m_2}^{\text{sc}}(q)$ as following:

$$\chi_{m_1 m_2}^{\text{sc}}(q) = \int_0^\beta d\tau e^{i\Omega_n \tau} \langle B_{qm_1}(\tau) B_{qm_2}(0) \rangle, \quad (\text{A1})$$

where $q = (\mathbf{q}, i\Omega_n)$, $\Omega_n = 2\pi n/\beta$ with an integer n is a boson Matsubara frequency, $\beta = 1/k_B T$, and $B_{qm}(\tau) = e^{H\tau} B_{qm} e^{-H\tau}$. $B_{qm} = \sum_{\mathbf{k}} c_{\mathbf{k}+\mathbf{q}\uparrow m} c_{-\mathbf{k}\downarrow m} \phi(\mathbf{k})$ is the creation operator of spin-singlet Cooper pairs with a center-of-mass momentum \mathbf{q} on the layer m , and $\phi(\mathbf{k}) = \cos k_x - \cos k_y$ shows the momentum dependence of the order parameter with d -wave symmetry. Taking into account the attractive interaction in the d -wave channel V_d , we obtain the following expression:

$$\hat{\chi}^{\text{sc}}(q) = \frac{\hat{\chi}_0^{\text{sc}}(q)}{\hat{1} - V_d \hat{\chi}_0^{\text{sc}}(q)}, \quad (\text{A2})$$

where $\hat{\chi}^{\text{sc}}(q)$ and $\hat{\chi}_0^{\text{sc}}(q)$ are the $M \times M$ matrix representation for the reducible and irreducible superconducting susceptibilities, respectively (M is the number of layers). The bare superconducting susceptibility $\hat{\chi}_0^{\text{sc}}(q)$ is obtained as

$$\begin{aligned} \chi_{0m_1 m_2}^{\text{sc}}(q) &= \frac{1}{\beta} \sum_{\mathbf{k}} \sum_l [G_{m_1 m_2}^{\uparrow\uparrow}(\mathbf{k} + \mathbf{q}, i\omega_l) \\ &\quad \times G_{m_1 m_2}^{\downarrow\downarrow}(-\mathbf{k}, i\Omega_n - i\omega_l) \phi(\mathbf{k}) \phi^*(\mathbf{k}) \\ &\quad - G_{m_1 m_2}^{\uparrow\downarrow}(\mathbf{k} + \mathbf{q}, i\omega_l) G_{m_1 m_2}^{\downarrow\uparrow}(-\mathbf{k}, i\Omega_n - i\omega_l) \\ &\quad \times \phi(\mathbf{k}) \phi^*(-\mathbf{k} - \mathbf{q})], \end{aligned} \quad (\text{A3})$$

where $\omega_l = \pi(2l + 1)/\beta$ is a fermion Matsubara frequency and $G_{m_1 m_2}^{ss'}(\mathbf{k}, i\omega_l)$ is the noninteracting Green function. The superconducting critical temperature T_c is determined by the divergence of the superconducting susceptibility $\hat{\chi}^{\text{sc}}(\mathbf{0}, 0)$, namely, $\hat{1} - V_d \hat{\chi}_0^{\text{sc}}(\mathbf{0}, 0) = 0$ (Thouless criterion).

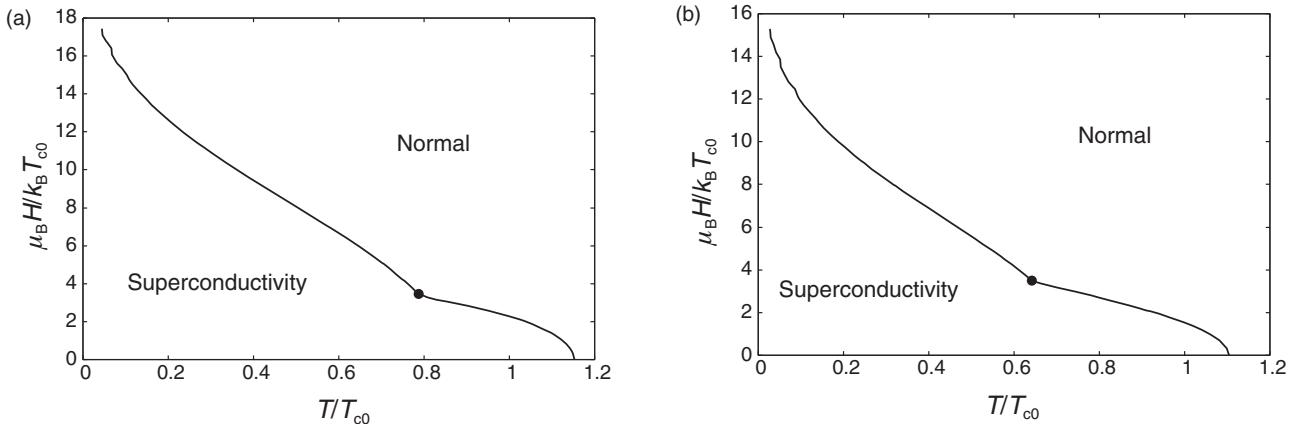


FIG. 6. T - H phase diagram for (a) bi- and (b) trilayer systems, with attractive interaction in the d -wave symmetry, $V(\mathbf{k}, \mathbf{k}') = -V_d(\cos k_x - \cos k_y)(\cos k'_x - \cos k'_y)$. We assume $V_d/t = 2.2$ and $\alpha/t_\perp = 3$. The critical temperature is $k_B T_{c0}/t = 0.0201$ for bilayer system and $k_B T_{c0}/t = 0.0202$ for trilayer system for $\alpha = 0$. Other parameters are the same as in Fig. 2. The points show the end point of first-order transition line between the BCS and the PDW states.

In Fig. 6, we show the phase diagram for bi- and trilayer systems. Although we cannot determine the first-order phase transition line between the BCS and PDW states within the linearized equation, the superconducting

transition lines in Figs. 6(a) and 6(b) resemble those in Figs. 2(d) and 3, respectively. Hence our results in this paper are qualitatively valid for the d -wave superconducting state.

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