

Paramagnetic Properties of Noncentrosymmetric Superconductors: Application to CePt₃Si

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In a noncentrosymmetric crystal, the Zeeman interaction of the band electrons with an external magnetic field is highly anisotropic in the momentum space, vanishing along some high-symmetry planes. One of the consequences is that the paramagnetic susceptibility in superconductors without inversion symmetry, such as CePt₃Si, shows an unusual temperature dependence.

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Electronic spin susceptibility measurements in the superconducting state provide one of the most useful tools for the identification of the pairing symmetry. Experimentally, the susceptibility $\chi(T)$ is measured by the Knight shift in the nuclear magnetic resonance frequency due to the hyperfine interaction of the conduction electrons with the nuclear magnetic moments. If the superconducting pairing occurs in the spin-singlet channel, then the Cooper pairs have spin $S = 0$ and do not contribute to the magnetization \mathbf{M} of the system, which is therefore entirely determined by the thermally excited quasiparticles. For a fully gapped order parameter one would see an exponentially decreasing $\chi(T)$ at low temperatures. In contrast, if the gap has zeros at the Fermi surface, then $\chi(T) \propto T^2$ for isolated point nodes, or $\chi(T) \propto T$ for line nodes. In the triplet case, when the Cooper pairs have spin $S = 1$ and the order parameter is a vector $\mathbf{d}(\mathbf{k})$ in the spin space (see, e.g., Ref. [1]), the susceptibility depends on the mutual orientation of \mathbf{d} and the external magnetic field \mathbf{B} . If $\mathbf{B} \parallel \mathbf{d}$, then the Cooper pairs do not contribute to \mathbf{M} , and $\chi(T)$ has the same temperature dependence as in the singlet case. If $\mathbf{B} \perp \mathbf{d}$, then both the pairs and the excitations contribute to \mathbf{M} , so that $\chi(T) = \chi_n$ —the normal-state susceptibility. The observation of a flat Knight shift in Sr₂RuO₄ for the field in the basal xy plane [2] has been used as proof of a spin-triplet pairing with $\mathbf{d} \parallel \hat{z}$.

The theoretical picture described above is valid only if the superconducting crystal has an inversion center. Although this is the case in the majority of superconductors, some exceptions have been known since the 1960s [3]. More recently, it was pointed out in Ref. [4] that the surface superconductors, e.g., Na-doped WO₃ [5], are intrinsically noncentrosymmetric simply because the two sides of the surface layer are manifestly nonequivalent. As for the bulk materials, the latest examples are CePt₃Si [6] and UIr [7]. Different models of superconductivity in CePt₃Si have been proposed in Refs. [8,9]. In this Letter, we calculate the suppression of the critical temperature and the paramagnetic susceptibility in superconductors without an inversion center. We focus on the tetragonal symmetry relevant for CePt₃Si, and show, in particular, that the

anisotropy of the temperature dependence of the susceptibility tensor $\chi_{ij}(T)$ is strikingly different from the centrosymmetric case.

The spin susceptibility in two-dimensional noncentrosymmetric superconductors has been previously studied in Refs. [4,10–12], where the inversion symmetry breaking in the presence of a nonzero spin-orbit (SO) coupling was introduced using the Rashba model [13]. Very recently, a three-dimensional generalization of the Rashba model was applied to CePt₃Si in Ref. [14]. Treating the SO band splitting as a perturbation, it was found that the order parameter becomes a mixture of a spin-singlet (even in \mathbf{k}) and a spin-triplet (odd in \mathbf{k}) component, which gives rise to a nonzero residual susceptibility at $T = 0$. In this Letter, we use a different approach based on the effective single-band Hamiltonian, which works for any crystal symmetry and arbitrary strength of the SO coupling. Guided by the fact that the SO band splitting is usually large compared to the superconducting energy scales, we consider both the Cooper pairing and the magnetic response independently in different bands. In contrast to the previous works, we construct the pairing interaction using the exact band states and explicitly take into account that all the pairing channels but one are suppressed [15].

The starting point of our analysis is the observation that in a crystal lacking an inversion center the electron bands are nondegenerate almost everywhere, except along some high-symmetry lines in the Brillouin zone. Indeed, without the inversion operation I , one cannot construct two orthogonal degenerate Bloch states at the same \mathbf{k} . At zero SO coupling there is an additional symmetry in the system—the invariance with respect to arbitrary spin rotations, which restores the twofold spin degeneracy of the bands. Here we assume that the SO coupling is sufficiently strong, so that the bands are well split. The results of Ref. [8] show that this is indeed the case in CePt₃Si, where the SO band splitting can be as large as 50–200 meV depending on the band. Assuming that there is no disorder in the crystal, the Bloch wave vector \mathbf{k} is a good quantum number in zero field. The free electron Hamiltonian for a nondegenerate band can be written as $H_0 = \sum_{\mathbf{k}} \epsilon(\mathbf{k}) c_{\mathbf{k}}^\dagger c_{\mathbf{k}}$,

where $\sum_{\mathbf{k}}$ stands for the integration over the first Brillouin zone, and $\epsilon(\mathbf{k})$ is the quasiparticle dispersion, which takes into account the periodic lattice potential and the SO interaction.

If the time-reversal symmetry is not broken in the normal phase, then the states $|\mathbf{k}\rangle$ and $K|\mathbf{k}\rangle \sim |-\mathbf{k}\rangle$ are degenerate because of the Kramers theorem [16]. It is the coupling between those states that leads to the formation of the Cooper pairs and the superconductivity in the system. The large band splitting strongly suppresses the pairing of electrons from different bands. Then, considering just one band (i.e., neglecting the interband pair scattering) and assuming that the interaction has a generalized Bardeen-Cooper-Schrieffer form, we have

$$H_{\text{int}} = \frac{1}{2} \sum_{\mathbf{k}, \mathbf{k}'} V(\mathbf{k}, \mathbf{k}') c_{\mathbf{k}}^{\dagger} c_{-\mathbf{k}}^{\dagger} c_{-\mathbf{k}'} c_{\mathbf{k}'}. \quad (1)$$

The pairing potential can be written as $V(\mathbf{k}, \mathbf{k}') = \tilde{V}(\mathbf{k}, \mathbf{k}') t(\mathbf{k}) t^*(\mathbf{k}')$, where $\tilde{V}(\mathbf{k}, \mathbf{k}') = -V_{\Gamma} \sum_a \phi_a(\mathbf{k}) \phi_a^*(\mathbf{k}')$ is the part that transforms according to an irreducible representation Γ of the normal-state point group G , $\phi_a(\mathbf{k})$ are the scalar basis functions of Γ , which are non-zero only inside the energy shell of width ϵ_c near the Fermi surface, $V_{\Gamma} > 0$ is the coupling constant, and $t(\mathbf{k}) = -t(-\mathbf{k})$ are nontrivial phase factors in $K|\mathbf{k}\rangle = t(\mathbf{k})|-\mathbf{k}\rangle$ [17]. Although anticommutation of fermionic operators dictates that the mean-field order parameter $\Delta(\mathbf{k}) = t(\mathbf{k}) \sum_a \eta_a \phi_a(\mathbf{k})$ is odd in \mathbf{k} [8], its nodal structure is determined by the basis functions $\phi_a(\mathbf{k})$, which should be even because of the presence of $t(\mathbf{k})$ [18]. The focus of this Letter is on CePt₃Si, which has a noncentrosymmetric tetragonal crystal lattice described by the point group $G = \mathbf{C}_{4v}$. This group is generated by the rotations C_{4z} about the z axis by an angle $\pi/2$ and the reflections σ_x in the vertical plane (100) and has five irreducible representations: four 1D (A_1 , A_2 , B_1 , and B_2) and one 2D (E) [19]. Here are the examples of the even basis functions: $\phi_{A_1} \propto k_x^2 + k_y^2 + ck_z^2$, $\phi_{A_2} \propto k_x k_y (k_x^2 - k_y^2)$, $\phi_{B_1} \propto k_x^2 - k_y^2$, $\phi_{B_2} \propto k_x k_y$, and $(\phi_{E,1}, \phi_{E,2}) \propto (k_x k_z, k_y k_z)$. We consider a small sample, of a dimension $d \leq \xi < \delta$, where ξ is the superconducting correlation length and δ is the London penetration depth, which allows us to neglect the spatial variations of both the order parameter components η_a and the magnetic field.

Let us now turn on a uniform stationary magnetic field $\mathbf{B} = \text{curl} \mathbf{A}$. Assuming that the pairing interaction is field independent, \mathbf{B} can affect the system only through its coupling to the band states. At $\mathbf{B} \neq 0$, the band dispersion function $\epsilon(\mathbf{k})$ is replaced by an effective band Hamiltonian in the momentum space, which can be represented as a power series in \mathbf{B} : $\epsilon(\mathbf{k}) \rightarrow \mathcal{E}(\mathbf{k}, \mathbf{B}) = \epsilon(\mathbf{K}) + B_i \epsilon_{1,i}(\mathbf{K}) + \dots$, where $\mathbf{K} = \mathbf{k} + (e/\hbar c) \mathbf{A}(i\nabla_{\mathbf{k}})$ because of the requirements of gauge invariance [20]. The expansion coefficients must satisfy certain symmetry-imposed conditions; in particular, the zero-field band dispersion $\epsilon(\mathbf{k})$ must be invari-

ant under all operations from G . In addition, at $\mathbf{B} \neq 0$ the Hamiltonian is invariant with respect to time-reversal K only if the sign of \mathbf{B} (and of \mathbf{A}) is also changed, which imposes the following constraint on the function \mathcal{E} : $K^{\dagger} \mathcal{E}(-\mathbf{B}) K = \mathcal{E}(\mathbf{B})$.

In the analysis of the ‘‘paramagnetic’’ properties of superconductors, the orbital effect of the field is neglected, which is achieved by putting $\mathbf{A} = 0$ in the effective band Hamiltonian. Then, for a twofold degenerate band in the presence of inversion symmetry, \mathcal{E} is a 2×2 matrix, and the coupling to the magnetic field is described by a familiar Zeeman term: $\mathcal{E}_{\alpha\beta}(\mathbf{k}, \mathbf{B}) = \epsilon(\mathbf{k}) \delta_{\alpha\beta} - B_i \mu_{ij}(\mathbf{k}) \sigma_{j,\alpha\beta}$, with $\mu_{ij}(\mathbf{k}) = \mu_{ij}(-\mathbf{k})$ being the tensor generalization of the Bohr magneton μ_B for the case of band electrons. The indices $\alpha\beta$ here are pseudospin indices [21]. The Zeeman interaction splits the energies of the electrons forming the Cooper pairs and gives rise to the paramagnetic suppression of superconductivity [22].

If the inversion symmetry is absent and the bands are nondegenerate, then the Zeeman term should be modified. The effective single-band Hamiltonian describing the linear response of the system on external magnetic field can be written as

$$H_0 = \sum_{\mathbf{k}} [\epsilon(\mathbf{k}) - \mathbf{B} \boldsymbol{\lambda}(\mathbf{k})] c_{\mathbf{k}}^{\dagger} c_{\mathbf{k}}, \quad (2)$$

which is markedly different from the centrosymmetric case. Here $\boldsymbol{\lambda}(\mathbf{k})$ is a real pseudovector, which is invariant under all operations from the point group G . Because of the time-reversal symmetry, we also have $\epsilon(-\mathbf{k}) = \epsilon(\mathbf{k})$ and $\boldsymbol{\lambda}(-\mathbf{k}) = -\boldsymbol{\lambda}(\mathbf{k})$ [note that $\mathcal{E}(-\mathbf{k}, \mathbf{B}) \neq \mathcal{E}(\mathbf{k}, \mathbf{B})$, which is a consequence of the lack of inversion symmetry].

Explicit expressions for $\boldsymbol{\lambda}(\mathbf{k})$ can be obtained only in some simple models. For example, in an isotropic two-dimensional electron gas in the xy plane with $G = \mathbf{C}_{\infty v}$, the combined effect of the SO coupling and the lack of inversion symmetry is described by an additional (Rashba) term in the single-particle Hamiltonian: $H_{\text{SO}} = \gamma \sum_{\mathbf{k}} \mathbf{n} \cdot (\boldsymbol{\sigma}_{\sigma\sigma'} \times \mathbf{k}) a_{\mathbf{k}\sigma}^{\dagger} a_{\mathbf{k}\sigma'}$, where \mathbf{n} is the normal vector to the plane [13]. Diagonalization of the Hamiltonian in zero field gives two nondegenerate Rashba bands: $\epsilon_{\pm}(\mathbf{k}) = \epsilon_0(\mathbf{k}) \pm \gamma |\mathbf{k}|$. At finite field, adding a usual Zeeman term $-\mu_B (\mathbf{B} \cdot \boldsymbol{\sigma})$ and expanding the eigenvalues of the Hamiltonian in powers of \mathbf{B} , we obtain $\mathcal{E}_{\pm}(\mathbf{k}, \mathbf{B}) = \epsilon_{\pm}(\mathbf{k}) - \boldsymbol{\lambda}_{\pm}(\mathbf{k}) \mathbf{B} + O(B^2)$, where $\boldsymbol{\lambda}_{\pm}(\mathbf{k}) = \pm \mu_B (\mathbf{k} \times \mathbf{n}) / |\mathbf{k}|$. Thus the coupling of the Rashba bands with the field is highly anisotropic; in particular, it vanishes for $\mathbf{B} \parallel \mathbf{n}$.

While a microscopic derivation of the effective single-band Hamiltonian (2) in more realistic systems can be done, at least in principle, using the procedures described in Ref. [20], it suffices for our purposes to work with a phenomenological expression for $\boldsymbol{\lambda}(\mathbf{k})$, which is compatible with all the symmetry constraints. We need an expression for $\boldsymbol{\lambda}(\mathbf{k})$, which satisfies (i) $\boldsymbol{\lambda}(-\mathbf{k}) = -\boldsymbol{\lambda}(\mathbf{k})$,

(ii) $(C_{4z}\boldsymbol{\lambda})(C_{4z}^{-1}\mathbf{k}) = \boldsymbol{\lambda}(\mathbf{k})$, and (iii) $(\sigma_x\boldsymbol{\lambda})(\sigma_x^{-1}\mathbf{k}) = \boldsymbol{\lambda}(\mathbf{k})$ (since $\boldsymbol{\lambda}$ is a pseudovector, we have $\sigma_x\boldsymbol{\lambda} \equiv IC_{2x}\boldsymbol{\lambda} = C_{2x}\boldsymbol{\lambda}$, where C_{2x} is a rotation by an angle π about the x axis). It is straightforward to check that the general expression for $\boldsymbol{\lambda}(\mathbf{k})$ is given by

$$\boldsymbol{\lambda}(\mathbf{k}) = \tilde{\phi}_{E,2}(\mathbf{k})\hat{x} - \tilde{\phi}_{E,1}(\mathbf{k})\hat{y} + \tilde{\phi}_{A_2}(\mathbf{k})\hat{z}, \quad (3)$$

where $\tilde{\phi}_{E,1(2)}(\mathbf{k})$ and $\tilde{\phi}_{A_2}(\mathbf{k})$ are real odd functions, which transform according to the representations E and A_2 , respectively, e.g., $\tilde{\phi}_{A_2} \propto k_x k_y k_z (k_x^2 - k_y^2)$ and $(\tilde{\phi}_{E,1}, \tilde{\phi}_{E,2}) \propto (k_x, k_y)$. We see that $\lambda_z = 0$ along the five nodal planes of the A_2 representation, while $\lambda_x = \lambda_y = 0$ along the z axis.

Now, we calculate the free energy \mathcal{F} for the Hamiltonian $H = H_0 + H_{\text{int}}$, defined by Eqs. (2) and (1). We use the effective field theory in terms of the bosonic Matsubara fields $\eta_a(\tau)$, which can be introduced in a standard fashion by decoupling the pairing interaction (1). The effective action for a uniform stationary order parameter in the mean-field approximation reads

$$S_{\text{eff}} = \frac{\beta\mathcal{V}}{2V_\Gamma} \sum_a |\eta_a|^2 - \frac{1}{2} \text{Tr} \ln G^{-1}, \quad (4)$$

where

$$G^{-1}(\mathbf{k}, \omega_n) = \begin{pmatrix} i\omega_n - \epsilon(\mathbf{k}) + \mathbf{B}\boldsymbol{\lambda}(\mathbf{k}) & -\Delta(\mathbf{k}) \\ -\Delta^*(\mathbf{k}) & i\omega_n + \epsilon(\mathbf{k}) + \mathbf{B}\boldsymbol{\lambda}(\mathbf{k}) \end{pmatrix} \quad (5)$$

is the 2×2 inverse Gor'kov Green function, $\omega_n = (2n + 1)\pi T$ is the fermionic Matsubara frequency, and \mathcal{V} is the system volume. The mean-field free energy is related to the saddle-point action (4): $\mathcal{F} = (1/\beta)S_{\text{eff}}$, and the magnetization density is $\mathbf{M} = -\mathcal{V}^{-1}(\partial\mathcal{F}/\partial\mathbf{B})$. The saddle-point condition yields the self-consistency equation

$$\frac{1}{V_\Gamma} \eta_a + T \sum_n \sum_{\mathbf{k}} t^*(\mathbf{k}) \phi_a^*(\mathbf{k}) G_{12}(\mathbf{k}, \omega_n) = 0, \quad (6)$$

which determines the temperature and field dependence of the order parameter components. Substituting here the Green function (5), one can see that the phase factors $t(\mathbf{k})$ drop out of the gap equation.

Let us find how the critical temperature is suppressed by the field. The equation for $T_c(\mathbf{B})$ is obtained by linearizing Eq. (6) and can be written in the form $\det\|K_{ab}\| = 0$, where

$$K_{ab} = \left[\ln \frac{T_{c0}}{T_c} + \psi\left(\frac{1}{2}\right) \right] \delta_{ab} - \left\langle \phi_a^*(\mathbf{k}) \phi_b(\mathbf{k}) \text{Re} \psi\left(\frac{1}{2} - i \frac{\mathbf{B}\boldsymbol{\lambda}(\mathbf{k})}{2\pi T_c}\right) \right\rangle_{\text{FS}}, \quad (7)$$

where $\psi(x)$ is the digamma function, $T_{c0} \approx 1.13\epsilon_c \exp(1/N_F V_\Gamma)$ is the critical temperature in zero field (N_F is the density of states at the Fermi level), and the angular brackets denote the average over the Fermi surface.

Next, differentiating the effective action (4) with respect to \mathbf{B} , we find the magnetization density

$$\mathbf{M} = \frac{1}{2} T \sum_n \sum_{\mathbf{k}} \boldsymbol{\lambda}(\mathbf{k}) [G_{11}(\mathbf{k}, \omega_n) + G_{22}(\mathbf{k}, \omega_n)]. \quad (8)$$

The uniform susceptibility tensor is defined in the usual manner as $\chi_{ij} = \partial M_i / \partial B_j |_{\mathbf{B}=0}$. One can easily check using Eq. (6) that the corrections to the order parameter components η_a in a weak magnetic field are quadratic in \mathbf{B} , which means that in the calculation of χ_{ij} one can neglect the field dependence of $\Delta(\mathbf{k})$, to obtain

$$\chi_{ij}(T) = \frac{1}{4T} \sum_{\mathbf{k}} \frac{\lambda_i(\mathbf{k}) \lambda_j(\mathbf{k})}{\cosh^2[E(\mathbf{k})/2T]}, \quad (9)$$

where $E(\mathbf{k}) = \sqrt{\epsilon^2(\mathbf{k}) + |\Delta(\mathbf{k})|^2}$.

We now apply the general theory to CePt₃Si. The pseudovector $\boldsymbol{\lambda}(\mathbf{k})$ in this case is given by Eq. (3). Since all three components of $\boldsymbol{\lambda}$ are in general nonzero, we expect the superconducting critical temperature to be suppressed for all orientations of the magnetic field. According to Eq. (7), the magnitude of the suppression depends on many factors: the shape of the Fermi surface, the symmetry of the order parameter, and also the explicit form of the functions $\tilde{\phi}$ in Eq. (3). The Fermi surface of CePt₃Si is quite complicated and consists of six sheets [8]. It is not known which one (or ones) of them are superconducting. The order parameter symmetry is not known either, although the observation of a linear temperature dependence of the specific heat [6] probably indicates that the order parameter has lines of nodes. In view of all this uncertainty, it seems to be premature to discuss the paramagnetic suppression in CePt₃Si quantitatively.

More interesting qualitative conclusions can be drawn from the analysis of the susceptibility $\chi_{ij}(T)$. It follows from Eq. (9) that the main contribution to the susceptibility tensor at low temperatures comes from the thermally excited nodal quasiparticles. If the order parameter $\Delta(\mathbf{k})$ has no zeros at the Fermi surface (e.g., for the A_1 representation), the susceptibilities are exponentially small in all directions. On the other hand, for the 1D order parameters corresponding to the representations A_2 , B_1 , or B_2 the lines of nodes are imposed by symmetry. In this case, the components of the susceptibility tensor are given by $\chi_{xx} = \chi_{yy} = \chi_{\parallel}$ and $\chi_{zz} = \chi_{\perp}$, where

$$\chi_{\parallel}(T) = \frac{1}{8T} \sum_{\mathbf{k}} \frac{\tilde{\phi}_{E,1}^2(\mathbf{k}) + \tilde{\phi}_{E,2}^2(\mathbf{k})}{\cosh^2[E(\mathbf{k})/2T]}, \quad (10)$$

$$\chi_{\perp}(T) = \frac{1}{4T} \sum_{\mathbf{k}} \frac{\tilde{\phi}_{A_2}^2(\mathbf{k})}{\cosh^2[E(\mathbf{k})/2T]}. \quad (11)$$

It is straightforward to show that for the field in the basal plane, $\chi_{\parallel}(T) \propto T$, since $\tilde{\phi}_{E,1}^2(\mathbf{k}) + \tilde{\phi}_{E,2}^2(\mathbf{k})$ is nonzero everywhere, except the poles of the Fermi surface. This

behavior is characteristic of the systems with lines of nodes. In contrast, for the field orientation along the z axis, we have from Eq. (11) $\chi_{\perp}(T) \propto T^3$, since $\phi_{A_2}(\mathbf{k})$ vanishes at the nodal lines for all the 1D order parameters. Such temperature dependence is never seen in the centrosymmetric case. For a two-component order parameter $\Delta(\mathbf{k}) \sim \eta_1 k_x k_z + \eta_2 k_y k_z$ with a horizontal line of zeros at $k_z = 0$, one also has a T^3 behavior of $\chi_{\perp}(T)$. On the other hand, the anisotropy and the temperature dependence of $\chi_{\parallel}(T)$ is determined by (η_1, η_2) .

We note that, in contrast to Refs. [4,14], our approach does not yield a finite value of the susceptibility at $T = 0$. The explanation is that the residual susceptibility in the systems described by the Rashba model comes from the interband transitions between the Rashba bands (Van Vleck susceptibility), which are not affected by the transition into the superconducting state [12]. In our model, the quasiparticles in each band respond to the external field independently of the other bands. If to take the interband transitions into account, then the observed susceptibility will be $\chi_{\text{tot}}(T) = \chi_0 + \chi(T)$, where the first term is the temperature-independent background that comes, e.g., from the Van Vleck processes, and the second term is the single-band contribution (9). Other mechanisms that might affect the residual susceptibility include the contributions from the unpaired sheets of the Fermi surface, or spin-reversing scattering at impurities or surface imperfections [23].

In conclusion, we have shown that the paramagnetic responses of superconductors with and without inversion center are qualitatively different. The most important feature is that, in the latter case, the coupling of the non-degenerate band electrons with the external field is strongly momentum dependent and vanishes, for symmetry reasons, along some high-symmetry planes in the Brillouin zone. This results in a high anisotropy of the susceptibility in the superconducting state with lines of nodes, from $\chi_{\parallel}(T) \propto T$ to $\chi_{\perp}(T) \propto T^3$, which can be used as a clearcut experimental test of our theory.

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