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Role of strong spin-orbit coupling in the superconductivity of the hexagonal pnictide SrPtAs

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In clean inversion symmetric materials, spin-orbit coupling is not thought to have a pronounced effect on spin-singlet superconductivity. Here we show that, for the recently discovered pnictide superconductor SrPtAs, this is not the case. In particular, for spin-singlet superconductivity in SrPtAs, strong spin-orbit coupling leads to a significant enhancement of both the spin susceptibility and the paramagnetic limiting field with respect to that usually expected for spin-singlet superconductors. The underlying reason for this is that, while SrPtAs has a center of inversion symmetry, it contains weakly coupled As-Pt layers that do not have inversion symmetry. This local inversion-symmetry breaking allows for a form of spin-orbit coupling that dramatically effects superconductivity. These results indicate that caution should be used when interpreting measurements of the spin susceptibility and the paramagnetic limiting field if superconductivity resides in regions of locally broken inversion symmetry.

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Spin-orbit coupling (SOC) has emerged as a central interaction in condensed matter physics. It plays an important role in creating topological insulators[1] and in understanding spin transport.[2] Furthermore, it has been found to play a role in superconductivity in three contexts: SOC appears relevant to the recently observed increase of the superconducting T_c of thin films by a magnetic field[3] (the origin of this increase is not yet understood); impurity spin-orbit scattering has been shown to lead to a finite spin susceptibility in *dirty* spin-singlet superconductors at zero temperature;[4] and in materials that lack a center of inversion symmetry, SOC is important to understand the response of superconductors to magnetic fields. However, when inversion symmetry is present, the role of SOC on clean spin-singlet superconductors is not thought to be particularly noteworthy. In this Rapid Communication we show that this is not the case. In particular, we show that in the recently discovered inversion-symmetric pnictide SrPtAs,[5] SOC has a pronounced effect on the superconducting properties.

The superconducting pnictides[6] present a fascinating class of materials that highlight the interplay between electronic correlations, superconductivity, and magnetism in a multi-orbital system.[7] SrPtAs is a member of this family with a unique feature: The As-Pt atoms in a single layer form a honeycomb lattice (see Fig. 1). This is in contrast to previously studied pnictide superconductors that contain square lattices. Unlike the square lattice pnictides, the honeycomb lattice layers in SrPtAs do not have inversion symmetry. The broken inversion symmetry inherent to a single As-Pt layer has nontrivial consequences. In particular, assuming that SrPtAs is a spin-singlet superconductor, we predict a nonvanishing spin susceptibility at zero temperature with a magnitude that is a significant portion of the normal state spin susceptibility. We further show that it is likely to have a critical field that is larger than the paramagnetic limiting field. This behavior is not expected for tetragonal pnictide superconductors, for which the individual As-Fe layers contain a center of inversion symmetry. Our results highlight that such measurements are not sufficient to distinguish spin-triplet and spin-singlet pairing in materials for which superconductivity resides in regions that do not locally have inversion symmetry.

SrPtAs has a superconducting transition temperature $T_c = 2.4$ K and the resistivity shows metallic behavior.[5] The unit cell of SrPtAs contains two inequivalent As-Pt layers that are related by inversion symmetry (see Fig. 1). As mentioned above, a single As-Pt layer does not have a center of inversion symmetry. This allows for a particular form of SOC that exists in each layer. As shown below, this SOC is larger than the interlayer coupling. We therefore consider SrPtAs to be a superconductor with local inversion-symmetry breaking. We use this term to refer to the fact that physical properties usually associated with noncentrosymmetric superconductivity appear in SrPtAs, despite the presence of a center of inversion symmetry. For spin-singlet superconductors, these properties include an enhanced paramagnetic depairing field and a nonvanishing spin susceptibility at zero temperature.[8–18] In the following, we initially present the electronic structure of SrPtAs and then turn to an examination of the superconducting state in this material.

First-principles calculations were performed using the highly precise full-potential linearized augmented plane wave (FLAPW) method.[19] We have used the experimental lattice constants $a = 4.24$ Å and $c = 8.98$ Å (Ref. 20) and a cutoff of 186 eV for basis functions. The local density approximation (LDA) is used for the exchange correlation as parametrized by Hedin and Lundqvist,[21] and SOC has been calculated using a second-variational treatment.[22] Figures 2 and 3 show the results of LDA calculations with and without SOC. Energy bands near the Fermi level originate from Pt $5d$ and As $4p$ orbitals. Specifically, the Fermi surface sheets labeled a and b in Fig. 3(a) stem from Pt d_{xy}, $d_{x^2-y^2}$, As p_x, and p_y orbitals while that labeled c stems from Pt d_{xz}, d_{yz} and As p_z orbitals. Our results without spin-orbit coupling agree with those of Ref. 23. Note the qualitative changes when SOC is added. In

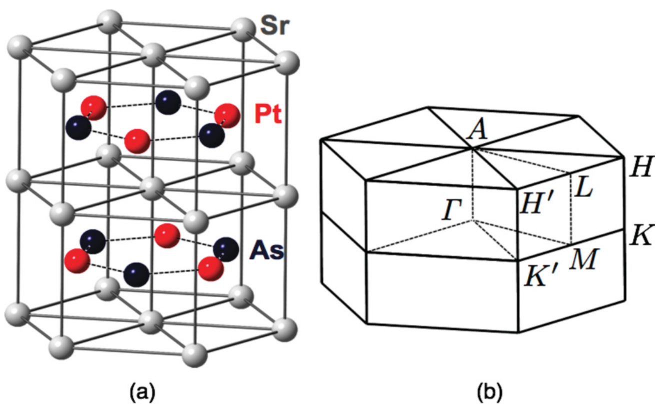

FIG. 1. (Color online) (a) Structure and (b) Brillouin zone in SrPtAs. Red, blue, and gray spheres denote Pt, As, and Sr atoms, respectively.

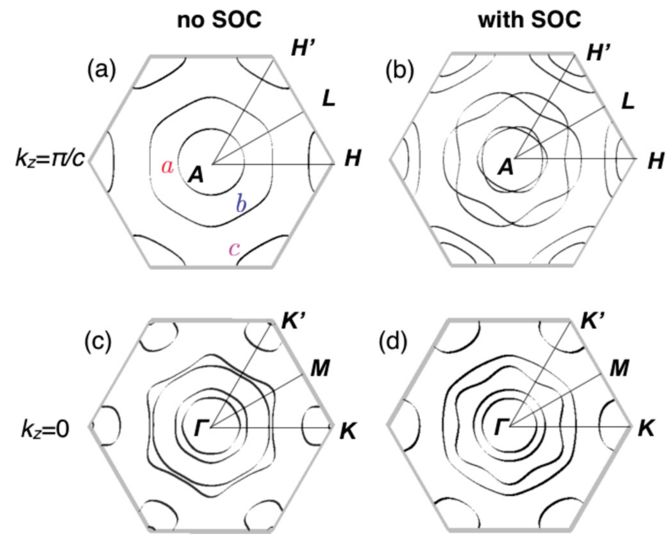

FIG. 3. (Color online) Cross sections of the Fermi surface of SrPtAs with and without SOC. (a) [(b)] is for $k_z = \pi/c$ and no SOC (with SOC), while (c) [(d)] is for $k_z = 0$ and no SOC (with SOC). Indices a, b, and c in (a) represent the three bands crossing the Fermi level.

particular, the spin-orbit coupling leads to appreciable changes in the band structure along the symmetry lines of H-A and L-H. Also of relevance is the difference between the bands along the symmetry lines H-A-L and K-Γ-M when there is no SOC (Fig. 2). This difference is due to interlayer coupling between the As-Pt layers. This coupling vanishes for symmetry reasons in the plane given by $k_z = \pi/c$. The band structure reveals that the band splittings due to SOC are comparable to or larger than those due to interlayer coupling. This fact plays an important role in the superconducting state.

To understand the bands stemming from the LDA calculations, it is useful to consider initially a single As-Pt layer. A key point is that this layer does not have a center of inversion and, therefore, a SOC of the form

$$\mathcal{H}_{\rm so}^{i} = \alpha_i \sum_{\boldsymbol{k},s,s'} \boldsymbol{g}_{\boldsymbol{k}} \cdot \boldsymbol{\sigma}_{ss'} c^{\dagger}_{\boldsymbol{k}si} c_{\boldsymbol{k}s'i} \qquad (1)$$

exists, where $c^{\dagger}_{\boldsymbol{k}si}$ ($c_{\boldsymbol{k}si}$) creates (annihilates) an electron with momentum $\boldsymbol{k}$ and pseudospin s in layer i, $\boldsymbol{\sigma}$ denote the Pauli matrices, and α_i is the layer i SOC energy. Time-reversal symmetry imposes $\boldsymbol{g}_{\boldsymbol{k}} = -\boldsymbol{g}_{-\boldsymbol{k}}$. Invariance of the Hamiltonian under the mirror symmetries with normals along the z axis and along the Pt-Pt bond imply that $\boldsymbol{g}(k_x,0,0) = \boldsymbol{g}(k_x,0,\pi/c) = 0$ (here a Pt-Pt bond is taken to be along the y axis). This reveals itself for bands along the A to L direction in the Brillouin zone, where there is no spin-orbit splitting [see Fig. 3(b)].

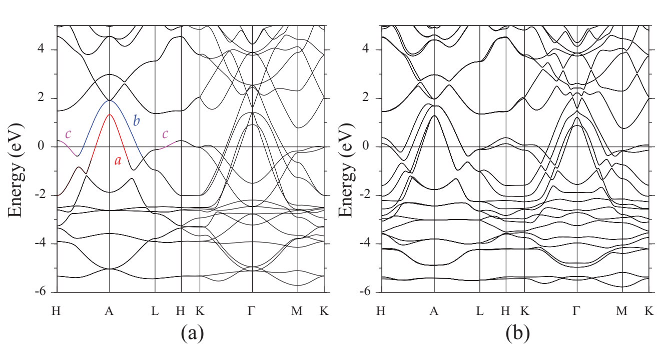

FIG. 2. (Color online) Energy bands of SrPtAs (a) without and (b) with SOC. Zero energy represents the Fermi level. Indices a, b, and c in (a) represent the three bands crossing the Fermi level.

Within a tight-binding approach, we find $\boldsymbol{g}_{\boldsymbol{k}} = \hat{z}\sum_i \sin(\boldsymbol{k}\cdot\boldsymbol{T}_i)$, where $\boldsymbol{T}_i$ are the translation vectors $\boldsymbol{T}_1 = (0,a,0)$, $\boldsymbol{T}_2 = (\sqrt{3}a/2,-a/2,0)$, and $\boldsymbol{T}_3 = (-\sqrt{3}a/2,-a/2,0)$. This form of SOC can be found for all bands stemming from Pt d orbitals by including hopping to neighboring As p orbitals and by including on-site SOC for both As and Pt sites. Symmetry also allows for g_x and g_y to be nonzero. However, these must be odd in k_z and, within a tight-binding analysis, are only found by including hopping along the z axis. Given the much weaker dispersion of the bands along k_z relative to the in-plane dispersion, we expect that g_x and g_y are much smaller than g_z and we will only include g_z in the following.

The analysis above applies to a single As-Pt layer. The two inequivalent As-Pt layers are related by inversion symmetry, and consequently α_i is of opposite sign for the two layers, i.e., $\alpha_i = (-1)^i\alpha$. To complete the description for the solid, a coupling between the two inequivalent layers is required. We take this to be $\epsilon_c(\boldsymbol{k})$ (symmetry requires this to vanish for $k_z = \pi/c$). Provided there are no band degeneracies other than spin and layer degeneracies, a generic Hamiltonian for SrPtAs is then

$$\mathcal{H}_0 = \sum_{\boldsymbol{k}} \Psi^{\dagger}(\boldsymbol{k})\{[\epsilon_1(\boldsymbol{k}) - \mu]\sigma_0\tau_0 + \alpha(\boldsymbol{k})\sigma_z\tau_z + \mathrm{Re}[\epsilon_c(\boldsymbol{k})]\sigma_0\tau_x + \mathrm{Im}[\epsilon_c(\boldsymbol{k})]\sigma_0\tau_y\}\Psi(\boldsymbol{k}), \qquad (2)$$

where $\Psi(\boldsymbol{k}) = (c_{\boldsymbol{k}\uparrow 1}, c_{\boldsymbol{k}\downarrow 1}, c_{\boldsymbol{k}\uparrow 2}, c_{\boldsymbol{k}\downarrow 2})^T$, σ_i (τ_i) are Pauli matrices that operate on the pseudospin (layer) space, and $\alpha(\boldsymbol{k}) = \alpha g_z(\boldsymbol{k})$. This Hamiltonian can be diagonalized with the resulting dispersion relations $\epsilon_{\pm}(\mathbf{k}) = \epsilon_1(\mathbf{k}) \pm \sqrt{|\epsilon_c(\mathbf{k})|^2 + \alpha^2(\mathbf{k})}$ and each state has a twofold Kramers degeneracy. To gain an intuition for the terms appearing in this Hamiltonian, we state the results for a simple tight-binding theory (note that below we keep these terms arbitrary). This yields $\epsilon_1(\mathbf{k}) = t_1(\cos\boldsymbol{k}\cdot\mathbf{T_1} + \cos\boldsymbol{k}\cdot$

$\mathbf{T_2} + \cos \boldsymbol{k} \cdot \mathbf{T_3}) + t_{c2}\cos(ck_z)$ and $\epsilon_c(\mathbf{k}) = t_c \cos(k_z c/2)(1 + e^{-i\mathbf{k}\cdot\mathbf{T}_3} + e^{i\mathbf{k}\cdot\mathbf{T}_2})$ with $\mathbf{T_i}$ and $\alpha(\boldsymbol{k})$ given above.

Now we turn to the superconducting state for which we show that the strong SOC is important. In the limit that the interlayer coupling vanishes, we have two uncoupled noncentrosymmetric systems. It is known that in noncentrosymmetric spin-singlet superconductors the spin susceptibility and the paramagnetic limiting field are significantly enhanced, if the SOC strength is much larger than the superconducting gap.[8–11,13,15] Given the large SOC compared to the interlayer coupling, it is conceivable that the behavior of superconducting SrPtAs resembles that of a noncentrosymmetric material. For this reason we calculate both the spin susceptibility and the limiting field assuming that SrPtAs is a spin-singlet superconductor (this is a reasonable assumption comparing with other pnictide superconductors).[7] To be concrete we assume intralayer s-wave pairing with an interaction

$$\mathcal{H}_{sc} = -V \sum_{\boldsymbol{k},\boldsymbol{k}',i,s,s'} c^{\dagger}_{\boldsymbol{k}si} c^{\dagger}_{-\boldsymbol{k}s'i} c_{-\boldsymbol{k}'s'i} c_{\boldsymbol{k}'si}, \tag{3}$$

where, as is usual in the weak-coupling limit, the sums over k and k' are restricted to electronic states within an energy range ω_c of the Fermi energy and V is determined by the observed transition temperature. Note that our results do not depend qualitatively on the choice of s-wave pairing.

For a system described by the Hamiltonians (2) and (3), the susceptibility in the superconducting and normal state can be calculated using[24]

$$\chi^s_{ij} = -\mu_B^2 T \sum_n \sum_{\boldsymbol{k}} \mathrm{tr}\big[\sigma_i G(\boldsymbol{k},\omega_n)\sigma_j G(\boldsymbol{k},\omega_n) - \sigma_i F(\boldsymbol{k},\omega_n)\sigma_j^T F^{\dagger}(\boldsymbol{k},\omega_n)\big], \tag{4}$$

with $G(\boldsymbol{k},\omega_n)$ and $F(\boldsymbol{k},\omega_n)$ the normal and anomalous Green's functions in the Matsubara formulation. Note that even for this "one-band" formulation, the Green's functions are 4×4 matrices, so that the trace runs over both layer and spin indices. In the notation of the Hamiltonian (2) there are three bands crossing the Fermi energy in SrPtAs [labeled a, b, and c in Figs. 2(a) and 3(a)] and we can generalize the above expression to

$$\chi_{ij} = \sum_{\nu} \chi_{ij}(\nu), \tag{5}$$

where the sum runs over the three orbital bands $\nu = a,b,c$. Below we calculate the susceptibility $\chi_{ij}(\nu)$ separately for each band ν using Eq. (4).

In the normal state, $F^{\nu}(\boldsymbol{k},\omega_n) = 0$, and we find for fields parallel to $\hat{z}$,

$$\chi^0_z(\nu) = 2\mu_B^2 \sum_{\boldsymbol{k},i=\pm} \frac{\partial n_F\big[\epsilon^{\nu}_i(\boldsymbol{k})\big]}{\partial \epsilon^{\nu}_i} = \sum_{\boldsymbol{k}} \chi^0_P(\boldsymbol{k},\nu), \tag{6}$$

where $n_F(\epsilon)$ is the Fermi distribution function as a function of energy ϵ and $\chi^0_p(\boldsymbol{k},\nu)$ denotes a Pauli susceptibility for band ν. This susceptibility describes intraband processes and at low temperatures is proportional to the density of states at the Fermi level. For fields in plane, we find

$$\chi^0_{\perp z}(\nu) = \sum_{\boldsymbol{k}} \left\{ \frac{\left|\epsilon^{\nu}_c(\boldsymbol{k})\right|^2 \chi^0_P(\boldsymbol{k},\nu) + [\alpha^{\nu}(\boldsymbol{k})]^2 \chi^0_{vV}(\boldsymbol{k},\nu)}{\left|\epsilon^{\nu}_c(\boldsymbol{k})\right|^2 + [\alpha^{\nu}(\boldsymbol{k})]^2} \right\}, \tag{7}$$

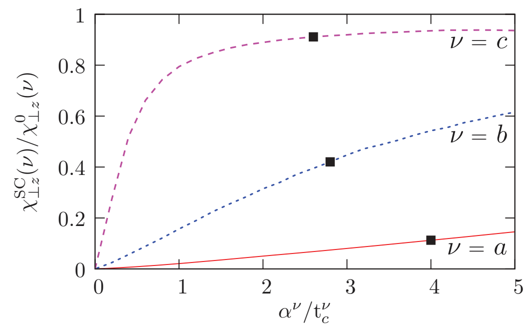

FIG. 4. (Color online) Spin susceptibility at $T = 0$ in the superconducting state normalized with respect to the normal state susceptibility for the three bands crossing the Fermi surface as a function of α^{ν}/t^{ν}_c. The black squares denote approximate values for these three bands. The tight-binding parameters used are (t_1, t_c, t_{c2}, μ): Band a (1.25,0.1,0.05,0.5); band b (1,0.1,0.05,2.5); and band c (−0.48,0.075,−0.03,0.6).

where we introduced

$$\chi^0_{vV}(\boldsymbol{k},\nu) = 2\mu_B^2 \left\{ \frac{n_F[\epsilon^{\nu}_{+}(\boldsymbol{k})] - n_F[\epsilon^{\nu}_{-}(\boldsymbol{k})]}{\sqrt{\left|\epsilon^{\nu}_c(\boldsymbol{k})\right|^2 + [\alpha^{\nu}(\boldsymbol{k})]^2}} \right\}. \tag{8}$$

This contribution describes processes between the two pseudospin and layer bands stemming from a single orbital band (labeled by ν) and is thus referred to as the van Vleck susceptibility. For the superconducting states in the limit $\sqrt{|\epsilon^{\nu}_c(\boldsymbol{k})|^2 + [\alpha^{\nu}(\boldsymbol{k})]^2} \gg \Delta^{\nu}$, we recover the expressions given in Eqs. (6) and (7), where for the Pauli susceptibility, we have to replace $\epsilon^{\nu}_{\pm}(\boldsymbol{k})$ with $E^{\nu}_{\pm}(\boldsymbol{k}) = \sqrt{[\epsilon^{\nu}_{\pm}(\boldsymbol{k})]^2 + [\Delta^{\nu}]^2}$. The Pauli susceptibility contribution thus vanishes due to the opening of the superconducting gap, while the van Vleck susceptibility is unchanged by superconductivity, even at $T = 0$. Consequently, χ^{SC}_z will behave as that expected for a conventional spin-singlet superconductor while $\chi^{\mathrm{SC}}_{\perp z}$ will have a large spin susceptibility, even at $T = 0$. To demonstrate this, Fig. 4 shows the ratio of $\chi^{\mathrm{SC}}_{\perp z}(\nu)$ in the superconducting phase at $T = 0$ to the normal state in-plane susceptibility $\chi^0_{\perp z}(\nu)$ as a function of α^{ν}/t^{ν}_c for the three bands $\nu = a,b,c$ (where α^{ν} is the spin-orbit strength and t^{ν}_c is the interlayer coupling strength). These values were determined using simple tight-binding calculations for the three Fermi surface sheets a,b,c. Estimating the ratios α^{ν}/t^{ν}_c from the band structure, we find values of $\chi^{\mathrm{SC}}_{\perp z}(\nu)/\chi^0_{\perp z}(\nu) = 0.11, 0.42$, and 0.91 for the Fermi surface sheet $\nu = a$, b, and c, respectively. Consequently, we expect that a sizable portion of the normal state susceptibility will exist in the limit $T \to 0$ for in-plane magnetic fields. We note that related behavior has recently been predicted for multilayer systems.[25]

The enhanced susceptibility suggests that the Pauli limiting field will also be enhanced. To calculate this, we include the Zeeman field $\mathcal{H}_Z = \sum_{\boldsymbol{k},s,s',i} g\mu_B \mathbf{H}\cdot\boldsymbol{\sigma}_{s,s'} c^{\dagger}_{\boldsymbol{k}si} c_{\boldsymbol{k}s'i}$ and orient the field in the basal plane. Within weak-coupling theory and assuming $\sqrt{|\epsilon_c(\boldsymbol{k})|^2 + \alpha^2(\boldsymbol{k})} \gg g\mu_B|\mathbf{H}|$ (which is well supported by LDA results), we find the following expression for T_c as a function of $h = g\mu_B|\mathbf{H}|$:

$$\ln\left(\frac{T_c}{T_{c0}}\right) = -\Psi\left(\frac{1}{2}\right) + \mathrm{Re}\left\{\frac{1}{2}\left\langle \Psi\left(\frac{1}{2} + ih(\boldsymbol{k})\right)\right\rangle_{\boldsymbol{k}}\right\}, \tag{9}$$

where

$$h(\boldsymbol{k}) = \frac{\frac{h|\epsilon_c(\boldsymbol{k})|}{2\pi k_B T_c}}{\sqrt{|\epsilon_c(\boldsymbol{k})|^2 + \alpha^2(\boldsymbol{k})}}, \tag{10}$$

T_{c0} is the transition temperature for $h = 0$, $\langle f \rangle_{\boldsymbol{k}}$ means an average of f over the Fermi surface, Re means real part, and $\Psi(x)$ is the digamma function. The band index ν is omitted for brevity. In the limit where $\epsilon_c = 0$, we find that T_c is independent of h. This is in agreement with the result predicted and observed for noncentrosymmetric superconductors in the limit of large SOC.[10,15] At $T_c = 0$, using Eq. (9), we find that the Pauli limiting field is given by

$$\Psi\left(\frac{1}{2}\right) = \left\langle \ln \left| \frac{h|\epsilon_c(\boldsymbol{k})|}{2\pi k_B T_{c0}\sqrt{|\epsilon_c(\boldsymbol{k})|^2 + \alpha^2(\boldsymbol{k})}} \right| \right\rangle_{\boldsymbol{k}}. \tag{11}$$

Using tight-binding calculations, we estimate that the enhancement of the Pauli limiting field h_P/h_{P0} (where h_{P0} is the limiting field when $\alpha = 0$) takes the values 1.1, 1.8, and 7.4 for Fermi sheets a, b, and c, respectively. Provided that the orbital upper critical field is sufficiently large, an enhanced Pauli limiting field should be observable. For fields along the z axis, a usual Pauli suppression is expected.

We point out that, in addition to a spin-singlet pairing order parameter, a spin-triplet order parameter component will also appear.[26] In particular, in a given layer a spin-triplet component with $\boldsymbol{d}(\boldsymbol{k})$ along the direction of $\boldsymbol{g}(\boldsymbol{k})$ exists, such that it has an opposite sign in the two inequivalent layers of SrPtAs. Using the results of Ref. 26, we estimate that the size of the spin-triplet order parameter component is a factor α/W smaller than the spin-singlet order parameter (W is the bandwidth). Consequently, this spin-triplet pairing will not qualitatively change the results given above. We also note that the Cooper pairs will in general have both spin-singlet and spin-triplet parts even without any spin-triplet order parameter component. This spin mixing of the Cooper pairs is included in the theory presented above.

In the calculation above, we focused on a spin-singlet order parameter. If the instability would occur in the spin-triplet channel, the SOC would force the $\boldsymbol{d}(\boldsymbol{k})$ vector to lie parallel to $\boldsymbol{g}(\boldsymbol{k})$.[26] A magnetic field in the $\hat{z}$ direction would therefore be pair breaking, similar to the spin-singlet case. Thus, as is also the case in noncentrosymmetric superconductors,[11] measurements of spin susceptibility and the critical field of SrPtAs do not suffice to distinguish spin-triplet and spin-singlet pairing.

In conclusion, we have shown that the unique structure in the pnictide SrPtAs has nontrivial effects on superconductivity. In particular, the lack of an inversion center in the As-Pt honeycomb lattice layers, combined with strong spin-orbit coupling, allows a significant enhancement of both the Pauli limiting field and the spin susceptibility for spin-singlet superconductivity. SrPtAs provides an ideal example of a material with inversion symmetry for which SOC can make a pronounced effect on spin-singlet superconductivity.

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