Isotropic s -wave superconductivity in the noncentrosymmetric charge density wave superconductor SrPt_2As_2

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SrPt_2As_2 undergoes a structural transition around 470 K (from a high-temperature tetragonal to a low-temperature orthorhombic phase) accompanied by a charge density wave transition and followed by a superconducting transition at much lower temperature (5 K). The superconducting gap symmetry of this noncentrosymmetric compound was investigated in detail by means of muon-spin rotation or relaxation (μSR) experiments in transverse-field (TF) configuration. The temperature-dependent magnetic penetration depth obtained from the TF- μSR spectra suggests an isotropic s -wave-type superconducting gap. No observable differences are seen in the asymmetry time spectra collected in zero field above and below the superconducting transition temperature T_c . This strongly suggests the absence of spontaneous magnetic fields in the superconducting state, hence pointing to a preserved time-reversal symmetry below T_c .

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I. INTRODUCTION

The unconventional nature of superconductivity (SC) often becomes apparent in the proximity of magnetic, charge, and valence instabilities [1–4]. Unconventional SC cannot be accounted for by the BCS theory, which is restricted to conventional superconductors. A variety of materials, including cuprates, heavy-fermion systems, and iron-based pnictide superconductors, belong to the class of unconventional superconductors [5–7]. The spin- or critical valence fluctuations are believed to play a key role in the formation of the superconducting condensate in such materials. Notably, in cuprates, spin fluctuations in the CuO_2 planes are postulated to participate in nodal d -wave SC with $d_{x^2-y^2}$ pairing symmetry [8]. In this context, iron-based pnictides have captured special attention, as they exhibit significantly high superconducting transition temperatures, even in the presence of antagonistic magnetic ordering, which mainly develops from a spin density wave (SDW) instability [7,9,10]. An extensive study carried out on 122 compounds, in particular on BaFe_2As_2 , which statically orders antiferromagnetically below 140 K, showed no traces of SC down to the lowest temperature [11–13]. The suppression of magnetic order by chemical or physical pressure leads to the emergence of SC with T_c as high as 38 K [14–17]. It is believed that the superconducting pairing mechanism is dominated by the magnetic fluctuations, thus resulting in an unconventional nature of SC. In iron pnictides, the nature of the superconducting gap symmetry still remains controversial, although different studies reported multiband

SC and also a sign reversal ($s\pm$)-type gap symmetry [18–20]. In order to gain further insight into the pairing mechanism of SC, it becomes important to identify the nature of gap symmetry and the role played by magnetic fluctuations associated with iron $3d$ orbitals.

For some years now, the systems which possess a charge density wave (CDW) instability, a nonmagnetic analog of SDW, have also attracted significant attention, considering that charge fluctuations associated with CDW are known to dramatically influence the superconducting ground state in cuprates [21]. Interestingly, CDW also develops in the tetragonal phase of $(\text{Ba,K})\text{Fe}_2\text{As}_2$ and $(\text{Ba,Na})\text{Fe}_2\text{As}_2$ for slightly underdoped samples, resulting in suppression of SC [22]. Both CDW and SC are often governed by the same fundamental electron-phonon interaction [23–26]. Hence, SC and CDW can compete with each other or may survive together. Although, individually, there is a general explanation of weakly coupled SC using the BCS theory [23–25] and of the CDW phenomenon using Fermi-surface nesting [27–29], the coexistence and/or the competition between the two is still under debate. Such interaction between CDW and SC can strongly influence the superconducting pairing mechanism. Hence, to understand the pairing mechanism in CDW superconductors, it is always desirable to look for and compare different systems manifesting the coexistence of SC and CDW.

Recently, in the quest to discover new CDW superconductors, a few nonmagnetic 122-type Pt-based arsenides, isostructural to 122 Fe-based arsenides, were studied in order to explore the coexistence phenomena of SC and CDW [30–37]. Among them, SrPt_2As_2 has received significant attention because of the possible coexistence of CDW and SC with $T_c \approx 5$ K, the latter being relatively higher than that of other Pt-based arsenides [38]. Interestingly, SrPt_2As_2

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crystallizes in a CaBe_2Ge_2 -type tetragonal structure which lacks inversion symmetry, as it has two inequivalent Pt and As sites due to stacking of alternate As-Pt-As and Pt-As-Pt layers along the c axis. Around 470 K, it undergoes a structural transition from a high-temperature tetragonal to a low-temperature orthorhombic structure, accompanied by a CDW transition [37,39,40]. The CDW modulation vector is reported to be $q_{\text{CDW}} = 0.62a^* = (0.62, 0, 0)$. In the SrPt_2As_2 case, it is believed that Fermi-surface nesting is responsible for the higher superconducting transition temperature T_c [38]. Moreover, a rigorous analysis of the temperature-dependent specific heat suggests an $(s + s)$ -wave gap symmetry at the Fermi surface and weak coupling, as in standard BCS superconductors [41]. Yet, recently, a single-gap s -wave symmetry was proposed based on the $1/T_1$ relaxation rate obtained from the ^{75}As nuclear quadrupole resonance (NQR), which shows a clear Hebel-Slichter peak below T_c and an exponential decrease at low temperatures [42]. To get more clarification on the superconducting order parameter, there is a clear call for a more comprehensive microscopic investigation. Motivated by these previous studies of SrPt_2As_2 , we performed detailed muon-spin rotation/relaxation (μSR) experiments on a polycrystalline sample of SrPt_2As_2 , grown using the solid-state reaction method. μSR is a powerful technique, able to provide the absolute value of the temperature-dependent magnetic penetration depth $\lambda(T)$, which in turn reflects the nature of the gap symmetry of a superconductor.

This paper is organized as follows: Sec. II describes the sample-preparation procedure, the results of x-ray and magnetization measurements, and the details of μSR experiments. In Sec. III, we report and discuss the results of transverse-field (TF) and zero-field (ZF) μSR experiments. Finally, Sec. IV contains our concluding remarks.

II. EXPERIMENTAL DETAILS

A. Sample preparation and characterization

The polycrystalline sample was prepared using a solid-state reaction method whose details are reported in Ref. [39]. Stoichiometric amounts of the elements were placed in a corundum crucible and sealed in a quartz tube under argon atmosphere. The mixture was heated to 980 °C, kept there for 24 h, and then cooled down to room temperature. The reaction products were homogenized and then annealed at 1050 °C several times. Figure 1 shows the powder x-ray diffraction (XRD) pattern recorded at room temperature. The Rietveld refinement of the recorded XRD pattern was performed using the TOPAS software [43]. All the peaks could be indexed well using an incommensurably modulated variant of the noncentrosymmetric CaBe_2Ge_2 -type structure with $q = 0.62a^*$, as reported in Ref. [39]. We did not observe traces of spurious phases, thus confirming that the grown samples are of high purity.

Magnetic susceptibility measurements were performed in the zero-field-cooled upon warming (ZFCW) and field-cooled upon cooling (FCC) modes in a 1.5-mT applied field using the vibrating sample magnetometer setup of a physical property measurement system (Quantum Design). Figure 2 shows the temperature-dependent volume magnetic

FIG. 1. X-ray diffraction pattern (black line) recorded at room temperature on a crushed polycrystalline sample of SrPt_2As_2 . The red line shows the theoretical curve obtained from the Rietveld refinement of the experimental pattern using the TOPAS software. The gray line is the difference between the experimentally observed and the theoretical spectra.

susceptibility $4\pi\chi_v(T)$. A sharp diamagnetic signal is seen in ZFCW susceptibility, which confirms the bulk nature of superconductivity in SrPt_2As_2 . The observed value of $4\pi\chi_v$ is smaller than the value of -1 expected for a sample showing a 100% superconducting volume fraction. The reasons for the smaller value of $4\pi\chi_v$ might be multiple: sample granularity, a remanent magnetic field, the demagnetization correction, etc. The superconducting transition temperature T_c is estimated to be 5 K from the cross point of two straight lines extrapolated from the normal and superconducting states.

FIG. 2. Temperature dependence of the volume magnetic susceptibility $4\pi\chi_v(T)$ collected in zero-field-cooled warming (ZFCW) and field-cooled cooling (FCC) modes in a 1.5-mT magnetic field.

B. Muon-spin rotation/relaxation technique

Muon-spin rotation/relaxation experiments were carried out at the Paul Scherrer Institute (PSI), Villigen, Switzerland. The measurements were performed in ZF and TF configurations on the Dolly and GPS spectrometers, situated respectively at the π E1 and π M3 beamlines. The experiments were carried out in the temperature range from 0.25 to 6 K in a 100-mT magnetic field for TF and in zero field for the ZF configuration. The sample was first cooled down from a temperature well above T_c to base temperature (1.5 K for the GPS spectrometer and 0.25 K for the Dolly spectrometer) in a 100-mT magnetic field, and then the μ SR spectra were collected upon warming. Finally, the data were analyzed with the help of the free software MUSRFIT [44].

III. RESULTS AND DISCUSSION

A. TF- μ SR experiments

To elucidate the nature of the pairing symmetry of the superconducting gap, we tracked the time evolution of the muon-spin polarization $P(t)$ in the TF configuration of a polycrystalline SrPt_2As_2 sample. Measurements were done in standard way [45] in the temperature range $0.25 \text{ K} \leq T \leq 6 \text{ K}$ in the presence of a 100-mT magnetic field. In Fig. 3, we show some representative μ SR time spectra for SrPt_2As_2 , recorded above (6 K) and below (1.5 K) T_c . The two data sets were collected at the GPS and Dolly spectrometers in temperatures down to 1.6 and 0.25 K, respectively. The TF- μ SR spectra in the normal state show a very weak depolarization arising from the randomly oriented nuclear magnetic moments, while in the superconducting state, a strong muon-spin depolarization is observed, reflecting the broad field distribution originating from the vortex lattice formation. The obtained asymmetry spectra $A(t)$ could be fitted well with a sinusoidal decaying Gaussian function,

$$A(t) = A_0 P(t) = A_s \exp\left(-\sigma_{\text{tot}}^2/2\right) \cos(\gamma_\mu B_{\text{int}} t + \phi) + A_{\text{bg}} \exp\left(-\sigma_{\text{bg}}^2/2\right) \cos(\gamma_\mu B_{\text{bg}} t + \phi), \quad (1)$$

where the first term refers to the sample, while the second one is related to the background from the sample holder. A_0 is the initial asymmetry. A_s and A_{bg} are the initial asymmetries associated with the sample and the sample holder, respectively. $\gamma_\mu/2\pi = 135.5 \text{ MHz/T}$ is the muon gyromagnetic ratio, B_{int} and B_{bg} are the internal fields experienced by the muons in the sample and the sample holder, and ϕ is the initial phase of the muon-spin polarization. σ_{bg} is the Gaussian depolarization rate for the muons stopped in the background, while σ_{tot} represents the total depolarization rate of the sample. σ_{tot} is quadratically related to the contribution from the superconducting vortex lattice σ_{sc} and the nuclear magnetic moment contribution σ_{nm} , where the latter is expected to be constant across the whole temperature range (above and below T_c). The Gaussian depolarization rate σ_{sc} reflects the second moment of the field distribution due to the formation of the flux-line lattice in the mixed state of a type-II superconductor. σ_{tot} is related to σ_{sc} and σ_{nm} by the relation $\sigma_{\text{tot}} = \sqrt{\sigma_{\text{sc}}^2 + \sigma_{\text{nm}}^2}$. The contribution of the vortex lattice $\sigma_{\text{sc}}(T)$ is obtained by subtracting the nuclear magnetic moment contribution σ_{nm} from $\sigma_{\text{tot}}(T)$, where σ_{nm} is obtained by fitting the spectra taken

FIG. 3. (a) Transverse-field μ SR-time spectra of a polycrystalline SrPt_2As_2 sample collected well above (6 K) and well below (1.5 K) T_c in a 100-mT applied magnetic field. The solid lines correspond to fits using Eq. (1). (b) The power spectrum of the fast Fourier transform (FFT) of the corresponding spectra presented in (a), showing the broadening and the shift of the line at low temperatures.

above T_c . The power spectrum of fast Fourier transform of the asymmetry spectra presented in Fig. 3(b) is well described by a Gaussian function. Above T_c , a narrow signal centered around the applied magnetic field $\mu_0 H = 100 \text{ mT}$ is seen. A slightly shifted and broader signal is seen below T_c , reflecting the nonuniform local magnetic field resulting from the flux-line lattice in the Shubnikov phase.

Figure 4 shows the Gaussian depolarization rate σ_{sc} at 1.5 K as a function of the applied transverse magnetic field. Each data point was obtained by cooling the sample in selected field values from above T_c to 1.5 K. According to the model developed by Brandt [46], in the case of a single-gap *s*-wave superconductor in the framework of the Ginzburg-Landau theory, σ_{sc} can be expressed as a function of the reduced field $b = H/H_{c2}$ ($\mu_0 H_{c2}$ is the upper critical field) and the Ginzburg-Landau coefficient κ by the relation [46]

$$\sigma_{\text{sc}} [\mu\text{s}^{-1}] \approx 4.83 \times 10^4 (1 - b) \times [1 + 1.21(1 - \sqrt{b})^3] \lambda^{-2} [\text{nm}^{-2}], \quad (2)$$

FIG. 4. Field dependence of the muon-spin depolarization rate σ_{sc} at 1.5 K. The solid red line through the experimental data points represents the theoretical variation of σ_{sc} with field, as expected from the Brandt model for an isotropic s -wave superconductor [46]. Expectations for the d -wave model and a two-component model are also shown for comparison (for details, see text). The inset shows the temperature dependence of the upper critical field $\mu_0 H_{c2}(T)$. The data included in the graph are adapted from Refs. [40,41]. FS and TS respectively represent the $\mu_0 H_{c2}(T)$ values as obtained from field- and temperature-scan procedures.

where λ is the magnetic penetration depth. For type-II superconductors, the above expression is valid for $b \geq 0.25/\kappa^{1.3}$ and $\kappa \geq 5$, where $\kappa = \lambda/\xi$, with ξ being the Ginzburg-Landau coherence length. Following Ref. [38], the estimated value of ξ from the upper critical field at 0 K is 11.5 nm. Thus, we can use Eq. (2) provided $\lambda > 57.5$ nm. $\sigma_{sc}(B)$ was fitted using Eq. (2), with the upper critical field $\mu_0 H_{c2}$ and λ as fit parameters. We obtain an upper critical field $\mu_0 H_{c2} = 3.3(2)$ T at 1.5 K, which is in close agreement with the value reported previously from specific-heat and transport measurements [41]. From the fit, the magnetic penetration depth λ at 0 K is determined to be 384(2) nm. For comparison, we also analyzed $\sigma(B)$ data using a nodal d -wave model [$\sigma(B) \propto (1 - a\sqrt{B})$], and a two-component model proposed by Serventi *et al.* [47]. The latter model assumes independent contributions from the two bands, where each band is assigned a different coherence length (ξ_1, ξ_2). The second moment of field distribution inside the vortex lattice is determined within the framework of the modified London model with the following functional form:

$$\overline{\Delta B^2} = \left(\frac{\sigma_{sc}}{\gamma_\mu} \right)^2 = B^2 \sum_{q \neq 0} \left[w \frac{e^{-q^2 \xi_1^2 / 2(1-b_1)}}{1 + q^2 \lambda^2 / (1-b_1)} + (1-w) \frac{e^{-q^2 \xi_2^2 / 2(1-b_2)}}{1 + q^2 \lambda^2 / (1-b_2)} \right]^2, \quad (3)$$

where w is the contribution of the first band to the total superfluid density, $b_{1(2)} = B/B_{c2,1(2)}$ are the reduced fields within each band, and ξ_1 and ξ_2 are coherence lengths related

FIG. 5. Temperature-dependent muon-spin depolarization rate σ_{sc} , as measured at the GPS and Dolly spectrometers. The inset shows the variation of the internal magnetic field with temperature in an applied magnetic field of 100 mT.

to each band. q represents the reciprocal lattice vector for the hexagonal flux-line lattice. A unique set of parameters is difficult to obtain because of the strong correlation between the following four parameters: λ , w , ξ_1 , and ξ_2 . To overcome such uncertainties, it is important to fix some quantities obtained from other techniques. A correlation between ξ_1 and ξ_2 ($\xi_1/\xi_2 = \sqrt{3}$) was established by analyzing specific heat data [41]. ξ_1 is approximated using the obtained value of the upper critical field, i.e., 3.3 T [41]. Thus, we fix $\xi_1 = 5.76$ nm and $\xi_2 = 9.97$ nm from the literature. A good approximation of the experimental $\sigma(B)$ data is obtained for $\lambda = 381$ nm and $w = 0.5$. At first glance, all three models can reproduce $\sigma(B)$ data reasonably well. However, we can directly exclude the possibility of d -wave symmetry, as $\sigma(T)$ saturates at low temperature, which reflects the nodeless SC (explained later). It is difficult to distinguish between the single-gap and two-gap models, but it can be seen from the inset in Fig. 4 that the upper critical field obtained by fitting $\sigma(B)$ with the Brandt model falls on the experimentally observed $\mu_0 H_{c2}$ vs T curve adapted from the literature [41]. This shows that a *single-gap s-wave model* is sufficient to describe the $\sigma(B)$ data, and there is no need to introduce an extra gap.

Figure 5 shows the temperature-dependent $\sigma_{sc}(T)$ obtained from the analysis of TF spectra recorded at the Dolly and GPS spectrometers by using Eq. (1). The two data sets are in good agreement with each other. The inset in Fig. 5 displays the temperature-dependent internal field in an applied field of 100 mT. The internal field in the superconducting state is smaller than the applied field due to the diamagnetic shift expected for type-II superconductors. The temperature-dependent inverse-square London magnetic penetration depth $\lambda^{-2}(T)$ is derived from the depolarization rate $\sigma_{sc}(T)$ and the temperature-dependent upper critical field $\mu_0 H_{c2}(T)$ by means of Eq. (2) [48]. In the inset of Fig. 4, we show the temperature-dependent upper critical field adapted from Refs. [40,41]. To calculate $\lambda^{-2}(T)$, we used the $\mu_0 H_{c2}(T)$ data

FIG. 6. Evolution of the inverse-square magnetic penetration depth $\lambda^{-2}(T)$ with temperature. The solid line represents a fit with a single-gap *s*-wave model, as described in the text.

by Xu *et al.* [41]. The variation of the inverse-square magnetic penetration depth $\lambda^{-2}(T)$ is shown in Fig. 6.

To get further insight into the superconducting order parameter, the normalized inverse-square magnetic penetration depth or, equivalently, the normalized superfluid density (superfluid density $n_s \propto \lambda^{-2}$) was modeled with the temperature-dependent superconducting gap function [49]

$$\frac{\lambda_L^{-2}(T)}{\lambda_L^{-2}(0)} = 1 + \frac{1}{\pi} \int_0^{2\pi} \int_{\Delta_i}^{\infty} \frac{E}{\sqrt{E^2 - \Delta_k^2}} \frac{\partial f}{\partial E} dE d\phi, \quad (4)$$

where Δ_k is the angle-dependent superconducting gap and $f = (1 + e^{E/k_B T})^{-1}$ is the Fermi distribution function. The gap Δ_k is a function of temperature and the azimuthal angle ϕ and can be written as $\Delta_k = \Delta(T)g(\phi)$. The temperature dependence of the gap function $\Delta(T)$ can be taken in the form $\Delta(T) = \Delta_0 \tanh\{1.821[1.018(T_c/T - 1)^{0.51}]\}$, deduced by the BCS theory [50]. Here Δ_0 refers to the gap value at 0 K. $g(\phi)$ describes the angular dependence of the gap function: it is equal to 1, $\sin\phi$, and $\cos 2\phi$ for *s*-, *p*-, and *d*-wave models, respectively. For a fully gapped superconductor, $\lambda^{-2}(T)$ exponentially saturates at low temperature because thermal excitations are unable to disturb the superconducting condensate. Since we, indeed, observe a saturating behavior of the superfluid density at low temperature, this suggests nodeless superconductivity in the SrPt₂As₂ system. It can be seen from Fig. 6 that $\lambda^{-2}(T)$ in the superconducting state is well captured by a single-gap *s*-wave fit with $\Delta_0 = 0.799(7)$ meV, $\lambda^{-2}(0) = 7.02(3) \mu\text{m}^{-2}$, and $T_c = 4.90(2)$ K. From $\lambda^{-2}(0)$, one obtains $\lambda = 377$ nm, which is in quite good agreement with the 384 nm from $\sigma(B)$ data. The gap-to- T_c ratio, i.e., $2\Delta_0/k_B T_c = 3.76$, is comparable to the BCS value in the weak-coupling limit, 3.52. Our results are consistent with ⁷⁵As NQR measurements, where a clear Hebel-Slichter peak is seen below T_c and the relaxation rate decreases exponentially, indicating a conventional BCS-type superconductivity [42]. It is noteworthy that there is no need

to introduce an extra gap parameter to fit the $\lambda^{-2}(T)$ and $\sigma(B)$ data. Nevertheless, Xu *et al.* [41] showed that the low-temperature $\mu_0 H_{c2}(T)$ data could not be explained with a single-band Werthamer-Helfand-Hohenberg (WHH) theory; hence, a multiband nature of SC was suggested. Furthermore, the field-dependent Sommerfeld coefficient $\gamma(B)$ could be decomposed into two curves with different slopes, which, again, is indicative of two types of charge carriers contributing to SC. The electronic band structure of SrPt₂As₂ also suggests that multiple bands cross the Fermi surface, and a multigap nature of the SC is expected [51]. A similar situation was encountered in centrosymmetric SrPt₃P, where the temperature-dependent magnetic penetration depth $\lambda(T)$ was well described by a single-gap *s*-wave model [52]. However, the strong field dependence of λ and an upward curvature in the upper critical field at low temperatures were still suggestive of multiband SC. This apparent dilemma was resolved by suggesting for SrPt₃P a two-band superconductor with equal gaps, but with different coherence lengths within the two Fermi-surface sheets [53]. Coming back to the present case of SrPt₂As₂, a possible explanation for the single-gap nature of SC in this multiband system may lie in its strong interband coupling. Even though the superconducting gap opens in different bands, due to strong interband coupling, single-gap SC can still be realized. A detailed explanation of how this could occur is given in the Appendix.

In the end, it is also important to mention the difference in studying single-crystal and polycrystalline samples with the μ SR technique. In most of the traditional μ SR studies, the analysis of TF spectra in the superconducting state is performed by assuming a Gaussian field distribution due to the flux-line lattice. This is not true for perfect single crystals, where an asymmetric field distribution is expected due to the formation of a well-aligned vortex lattice. This asymmetric signal includes a cutoff at low field $B_{\min}$, a peak related to the saddle point located at the midpoint between the two adjacent vortices B_{sad} , and a long tail arising at high field caused by the vortex regions $B_{\max}$ (see Refs. [54,55] and references therein). However, for polycrystalline materials, this asymmetric field distribution could be masked by the effect of random pinning of vortices and fluctuations in the temperature or applied magnetic fields, which can result in an almost Gaussian field distribution. This masking can sometimes obfuscate the exact nature of the temperature-dependent magnetic penetration depth $\lambda(T)$; hence, it may give wrong information about the gap symmetry. One well-known example is YBa₂Cu₃O_{6.95}, a member of the cuprate family, where, initially, the *s*-wave nature of the superconducting gap symmetry was predicted by means of TF- μ SR experiments on a polycrystalline sample [56], but later on, a *d*-wave gap symmetry was realized through μ SR investigations on detwinned single crystals [57]. Thus, the exact determination of $\lambda(T)$ and hence the accurate description of the superconducting gap symmetry requires high-quality single crystals. Nevertheless, for polycrystalline SrPt₂As₂, the nodeless SC speculated through $\sigma(T)$ is further corroborated by the following comprehensive techniques: (1) The temperature evolution of the $1/T_1$ relaxation derived from ⁷⁵As NQR depicts a clear Hebel-Slichter peak below T_c , which decreases exponentially at low temperatures, indicating the opening of the isotropic gap around the Fermi surface [42].

(2) The temperature-dependent electronic specific heat $C_e(T)$ could be replicated well with a two-gap model, with both gaps having s -wave symmetry [41]. (3) The field-dependent muon depolarization rate $\sigma(B)$ (this study) could be described well with the Brandt model accepted for the single-gap s -wave scenario. Thus, we can safely categorize SrPt_2As_2 as a fully gapped bulk superconductor.

B. ZF- μ SR experiments

The lack of inversion symmetry in SrPt_2As_2 , reflecting its noncentrosymmetric (NC) crystal structure, results in a nonuniform lattice potential. Potentially, this can give rise to an admixture of spin-singlet and spin-triplet Cooper pairs due to an asymmetric spin-orbit coupling. The possibility of symmetry-mixed Cooper pairs in a NC superconductor is reflected in time-reversal symmetry (TRS) breaking due to the appearance of small spontaneous magnetic fields in the superconducting state. ZF- μ SR turns out to be a very sensitive technique to detect such small fields (down to 10^{-4} T). Previously, it was successfully used to detect TRS breaking in the superconducting state of many compounds like Sr_2RuO_4 [58], UPt_3 [59], $\text{PrOs}_4\text{Sb}_{12}$ [60], $\text{PrPt}_4\text{Ge}_{12}$ [61], $\text{LaNi}(\text{C},\text{Ga})_2$ [62,63], La_7Ir_3 [64], $\text{Re}_{24}\text{Ti}_5$ [65], and $\text{Re}_{0.82}\text{Nb}_{0.18}$ [66], among others. In order to detect the presence of a tiny spontaneous magnetic field in the superconducting state of SrPt_2As_2 , we collected various ZF spectra at temperatures ranging from 1.6 to 8 K. To check the consistency, two sets of data were recorded [shown by red and black symbols in Fig. 7(b)]. Some representative spectra are shown in Fig. 7(a). Except for the 1.6 K data set, we could not detect significant differences between the spectra collected above and below T_c . The reason for the slightly enhanced relaxation at 1.6 K is not clear at this stage. As this change is not detected at T_c , it is most likely not related to the superconducting state of SrPt_2As_2 . For a quantitative analysis, the observed ZF spectra could be satisfactorily fitted using the following model:

$$A(t) = A_0 \exp(-\Lambda t) G_{\text{KT}}, \quad (5)$$

where G_{KT} is the Gaussian Kubo-Toyabe function [67], which describes the muon-spin depolarization rate associated with the nuclear magnetic moments, with the functional form

$$G_{\text{KT}}(t) = \frac{1}{3} + \frac{2}{3}(1 - \sigma_{\text{ZF}}^2 t^2) \exp\left(-\frac{\sigma_{\text{ZF}}^2 t^2}{2}\right), \quad (6)$$

where σ_{ZF} is the width of the nuclear dipolar field distribution experienced by the muon-spin ensemble and Λ represents the electronic-spin relaxation rate. For a nonmagnetic system, the depolarization of the muon-spin ensemble is due to static and randomly oriented nuclear moments, while the additional exponential-decay term is added to account for a possible field distribution due to electronic spins. The depolarization rate σ_{ZF} was fixed to a value of $0.0794 \mu\text{s}^{-1}$, obtained from fitting the spectra above T_c . The temperature-dependent electronic relaxation rate $\Lambda(T)$ is displayed in Fig. 7(b). No additional relaxation was observed across T_c , thus ruling out the possibility of spontaneous magnetic fields which might break TRS below T_c in this system. Hence, we conclude that TRS is preserved in SrPt_2As_2 upon entering the superconducting state. Generally, the lack of inversion symmetry does not necessarily

FIG. 7. (a) ZF- μ SR time spectra of polycrystalline SrPt_2As_2 measured at 1.6, 3, 5, and 7.5 K. (b) Electronic relaxation rate $\Lambda(T)$ as a function of temperature, obtained by fitting Eq. (6) to the corresponding data sets. Two sets of data (shown by red and black symbols) were collected to check consistency. No visible changes in the relaxation rate were observed across the superconducting transition T_c (shown by a vertical dashed line). The horizontal dashed line highlights the lack of change (within the error bars) in the relaxation rate across T_c .

mean that time-reversal symmetry is broken. Indeed, there are many examples of NC superconductors, e.g., $\text{Mo}_3\text{Al}_2\text{C}$ [68], LaRhSi_3 [69], $\text{Mg}_{10}\text{Ir}_{19}\text{B}_{16}$ [70], and LaPt_2Si_2 [34], which did not exhibit TRS breaking in the superconducting state. The TRS breaking in NC superconductors is mostly related to unconventional pairing mechanisms. For a multiband system with fully opened superconducting gaps, TRS breaking in the superconducting state can be realized in two circumstances. The first scenario is when the Coulomb repulsion between two Fermi surface pockets dominates, hence resulting in a nonzero relative phase difference between two pockets where the gap appears. Such a superconducting state (an $s + is$ state) will break TRS. The second scenario is suggested for the well-renowned TRS breaking superconductors LaNiC_2 and LaNiGa_2 [71], where SC is dominated by the interband pairing. A nonunitary triplet SC with pairing between the same spins from two orbitals is proposed, which results in breaking of TRS despite singlet s -wave pairing for both bands. Coming to the present case, we did not see any visible change

in the relaxation rate across T_c , which rules out the TRSB in SrPt_2As_2 . This indicates the fact that none of the above-discussed scenarios is realized for SrPt_2As_2 , hence preserving the time-reversal symmetry in SrPt_2As_2 .

IV. CONCLUSION

We investigated the superconducting properties of the SrPt_2As_2 noncentrosymmetric superconductor by means of zero-field and transverse-field μSR techniques. The temperature evolution of the superfluid density, as obtained from TF- μSR experiments, could be reproduced well with an isotropic single-gap s -wave model. The single-gap nature of superconductivity was further corroborated by the field dependence of the Gaussian depolarization $\sigma_{\text{sc}}(B)$ in the superconducting state, which is well described by the Brandt model for an isotropic s -wave superconductor. The existing reports on the electronic-band structure of SrPt_2As_2 show that the multiband nature of superconductivity is expected when multiple bands cross the Fermi surface. The unusual realization of single-gap superconductivity in a multiband system can probably be explained by taking into account the interband coupling. Finally, the relaxation rate obtained from ZF- μSR experiments did not evidence changes across

T_c , hence suggesting that time-reversal symmetry is preserved in the superconducting phase of SrPt_2As_2 , despite its noncentrosymmetric structure.

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APPENDIX: A SELF-CONSISTENT APPROACH FOR A TWO-BAND SUPERCONDUCTOR

In this Appendix, we show that single-gap superconductivity can still be realized in a multiband superconductor with strong interband coupling. For this, we use the self-consistent coupled gap equations within the quasiclassical Eilenberger weak-coupling formalism to obtain the temperature dependence of the gaps, instead of the phenomenological α model [50,53–55,72–76], which considers a similar BCS-type temperature dependence for both gaps. The coupled gap equations as introduced by Kogan are [77]

$$\delta_1 = \frac{\Delta_1}{T_c} \frac{1}{2\pi t} = n_1 \lambda_{11} \delta_1 \sum_{n=0}^{\infty} \left[S + \ln\left(\frac{1}{t}\right) - \left(\frac{1}{n+0.5} - \frac{1}{\sqrt{\delta_1^2 + (n+0.5)^2}} \right) \right] + n_2 \lambda_{12} \delta_2 \sum_{n=0}^{\infty} \left[S + \ln\left(\frac{1}{t}\right) - \left(\frac{1}{n+0.5} - \frac{1}{\sqrt{\delta_2^2 + (n+0.5)^2}} \right) \right], \quad (\text{A1})$$

$$\delta_2 = \frac{\Delta_2}{T_c} \frac{1}{2\pi t} = n_1 \lambda_{21} \delta_1 \sum_{n=0}^{\infty} \left[S + \ln\left(\frac{1}{t}\right) - \left(\frac{1}{n+0.5} - \frac{1}{\sqrt{\delta_1^2 + (n+0.5)^2}} \right) \right] + n_2 \lambda_{22} \delta_2 \sum_{n=0}^{\infty} \left[S + \ln\left(\frac{1}{t}\right) - \left(\frac{1}{n+0.5} - \frac{1}{\sqrt{\delta_2^2 + (n+0.5)^2}} \right) \right]. \quad (\text{A2})$$

In the above equations,

$$S = \frac{n_1 \lambda_{11} + n_2 \lambda_{22} - \sqrt{(n_1 \lambda_{11} - n_2 \lambda_{22})^2 + 4n_1 n_2 \lambda_{12}^2}}{2n_1 n_2 (\lambda_{11} \lambda_{22} - \lambda_{12}^2)}, \quad (\text{A3})$$

and $t = T/T_c$ is the reduced temperature. Here n_1 and n_2 are the partial densities of states for the corresponding bands at the Fermi level. λ_{11} (λ_{22}) and λ_{12} (λ_{21}) are the intraband and the interband coupling strengths, respectively. In the notation of Kogan, the two interband coupling strengths are equal, i.e., $\lambda_{12} = \lambda_{21}$. The temperature variation of $\Delta_1(T)$ and $\Delta_2(T)$ obtained from Eqs. (6) and (A1) is then used to evaluate the temperature variation of the normalized inverse-square magnetic penetration depth $\lambda_v^{-2}(T)/\lambda_v^{-2}(0)$, which directly gives the superfluid densities $[\rho_v(T)]$ for the two bands, by using

the expression

$$\rho_v(T) = \frac{\lambda_v(T)^{-2}}{\lambda_v(0)^{-2}} = \delta_v^2 \sum_{n=0}^{\infty} [\delta_v^2 + (n+0.5)^2]^{-\frac{3}{2}}, \quad (\text{A4})$$

where $v = 1, 2$ are the band indices.

With the known temperature variation of $\rho_1(T)$ and $\rho_2(T)$, the total superfluid density $\rho(T)$ is then obtained using the following equation:

$$\rho(T) = \gamma \rho_1(T) + (1 - \gamma) \rho_2(T), \quad (\text{A5})$$

where γ is the weight factor for the contribution of superconducting states with the larger gap Δ_1 . The main panels of Fig. 8 show the temperature-dependent total superfluid density $\rho(T)$, and the insets present the temperature variation of gaps $\Delta_1(T)$ and $\Delta_2(T)$, estimated using coupled gap equations. For simplicity we assumed $\gamma = 0.5$, $n_1 = 0.5$, and

FIG. 8. (a)–(c) The superfluid density $\rho(T)$ (shown by the dashed green line) as a function of temperature for $\lambda_{11} = 0.32$, $\lambda_{22} = 0.3$, and $\lambda_{12} = 0, 0.01$, and 0.2 respectively. (d) $\rho(T)$ for $\lambda_{11} = \lambda_{22} = 0.3$ and $\lambda_{12} = 0.01$. The insets of all graphs show the temperature-dependent gaps $\Delta_1(T)$ and $\Delta_2(T)$ obtained using coupled gap equations. Black and red solid lines in all graphs show the contributions of Δ_1 and Δ_2 to the total superfluid density.

$n_2 = 0.5$. In order to see the variation of the total superfluid density as a function of temperature for different coupling strengths, we considered two cases: (1) Intraband coupling is different for the two bands, and (2) intraband coupling is the same for the two bands. The first scenario is depicted in Figs. 8(a)–8(c), where the intraband coupling constants were fixed to $\lambda_{11} = 0.32$ and $\lambda_{22} = 0.3$, and different interband coupling strengths (0, 0.01, and 0.2) were considered. As shown in the inset of Fig. 8(a), in the case of zero coupling between the bands, the temperature dependence of the $\Delta_1(T)$ and $\Delta_2(T)$ gaps is different, and the gaps vanish at two different temperatures. An inflection point in $\rho(T)$ can also be seen at the temperature where the smaller gap vanishes. We can say that a clear two-gap behavior is seen for this case. Experimentally, similar kinks have been observed in the temperature-dependent inverse-square magnetic penetration depth in the basal plane, i.e., $\lambda_{ab}^{-2}(T)$ of $\text{FeSe}_{0.94}$. The reason was attributed to an extremely small interband coupling $\lambda_{12} = 0.0005$ [78]. However, the gaps start to close at the same temperature, as we increase the

interband coupling to 0.01 [see Fig. 8(b)], finally approaching an almost similar temperature variation in the case of rather strong interband coupling, $\lambda_{12} = 0.2$. Thus, it is fair to say that in the two-band model with nonzero coupling, both gaps open practically at the same T_c . This observation suggests that for nonzero interband coupling, a single-gap model is enough to describe the temperature variation of the superfluid density. Next, for the second case, we assumed the intraband coupling constants are the same, i.e., $\lambda_{11} = \lambda_{22} = 0.3$. The temperature variation of the gaps remained identical [see Fig. 8(d)], *independent of the interband coupling constant*. This is the reason why we show the temperature-dependent superfluid density only for $\lambda_{12} = 0.01$. It is also clear that an experimentally obtained superfluid density in this situation can be described also with a single-gap model. In conclusion, we can say that even though the superconducting gap opens at two different Fermi surfaces, the temperature variation of the superfluid density as a whole can still be described well with a single-gap model due to the high coupling between the electronic bands.

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