I. INTRODUCTION

The noncentrosymmetric superconductors (NCSs) that allow mixing between spin-singlet and spin-triplet parity and exhibit exotic superconducting properties through the antisymmetric spin-orbit coupling (ASOC) are of great interest in current research activities on superconductivity [1]. These systems without inversion symmetry spin-triplet pairing is not permitted [1]. These contradicting situations are accounted for by mixed spin singlet-triplet states of the order parameter [6]. The irreducible representation point group for the tetragonal structure of CePt3Si is C4v, in which Rashba-type ASOC exists, which provides the key to understanding the intriguing superconducting behavior of CePt3Si [7–9]. The Ce-based NCSs CePt3Si many NCSs have been identified that present interesting superconducting properties, including Li2(Pd,Pt)3B [10,11], CeRhsi3 [12,13], CeIrSi3 [14], CeCoGe3 [15,16], CeIrGe3 [17], LaNiC2 [18–20], BaPtSi3 [21], (Rh, Ir)Ga5 [22,23], Mg2Ir10B16 [24], Mo3Al2C [25], LaRhSi3 [26], CaIrPtSi3 [27], Re3W [28], Nb1.18Re1.12 [29], Re2Zr [30], La(Pd,Pt)Si3 [31], Ca4Ir7Ge4 [32], etc. The Ce-based NCSs CeRhsi3, CeIrSi3, CeCoGe3, and CeIrGe3 crystallize with BaNiSn3-type tetragonal structure (space group P4mm), which lacks mirror plane symmetry along the c axis and belongs to the same point group, C4v, as CePt3Si. Thus, like CePt3Si, a Rashba-type ASOC is present in these CeMX3 compounds too, leading to an exotic superconducting ground state in them [12–17,23–35]. Like CePt3Si, they also exhibit heavy-fermion behavior and undergo a long-range antiferromagnetic ordering; however, they...
become superconducting only under the application of pressure [12–17,33–35]. The above-mentioned Ce-based NCSs are situated close to a magnetic quantum critical point, making it difficult to explore the effects of ASOC and inversion symmetry breaking on superconductivity. Therefore nonmagnetic Rashba-type NCSs are essential for understanding the effect and extent of ASOC on the superconducting properties of these Ce-based NCSs. The reported nonmagnetic AMX$_3$ NCSs with BaNiSn$_3$-type tetragonal structure include BaPtSi$_3$ ($T_c = 2.25$ K), LaRhSi$_3$ ($T_c = 2.16(8)$ K), CalIrSi$_3$ ($T_c = 3.6$ K), CaPdSi$_3$ ($T_c = 2.3$ K), LaPdSi$_3$ ($T_c = 2.65(5)$ K), and LaPtSi$_3$ ($T_c = 1.52(6)$ K) [21,26,27,31]. All these nonmagnetic NCSs behave like a conventional s-wave superconductor without any noticeable effect from the absence of inversion symmetry in their crystal structure. Nevertheless, being isostructural, they provide a direct comparison with CeM$_X$$_3$ NCSs and the investigations of these nonmagnetic NCSs connote the role of 4f moments in Ce NCSs. One important difference between the CeM$_X$$_3$ NCSs and these nonmagnetic NCSs is that CeM$_X$$_3$ exhibit superconductivity only at high pressures, whereas these nonmagnetic NCSs superconduct at ambient pressure. This difference may have its origin in magnetic pairing in Ce NCSs in contrast to phonon-mediated superconductivity in these nonmagnetic NCSs.

Theoretically, the Rashba-type ASOC has been studied extensively and is favored for investigations of NCSs; therefore compounds with tetragonal BaNiSn$_3$-type structure represent an important class of noncentrosymmetric materials. Continuing our work on BaNiSn$_3$-type structured materials, we have performed a comprehensive study of superconducting and normal-state properties of NCS LaIrSi$_3$ using heat capacity $C_p$ and electrical resistivity $\rho$ versus temperature $T$ measurements down to 0.35 K and muon spin relaxation and rotation ($\mu$SR) measurements down to 50 mK. The reported noncentrosymmetric body-centered tetragonal BaNiSn$_3$-type structure of LaIrSi$_3$ is confirmed by our room-temperature powder x-ray diffraction (XRD) and neutron diffraction. Superconductivity in LaIrSi$_3$ was reported about 30 years ago with $T_c$ between 1.9 and 2.7 K based on resistivity measurement [36,37]. In a recent study Okuda et al. reported a superconducting transition at $T_c = 0.77$ K from the heat-capacity measurement on LaIrSi$_3$ [33]. Okuda et al. also carried out a de Haas–van Alphen (dHvA) effect study and found that as a result of Rashba-type ASOC the Fermi surface of LaIrSi$_3$ splits into two Fermi surfaces (spin-up and spin-down energy bands), which are separated by about 1000 K [33]. In our recent investigations of superconducting properties of NCS LaRhSi$_3$ we found a conventional type-I superconductivity with preserved time-reversal symmetry, however, with an unusual exponential evolution of the Sommerfeld coefficient $\gamma$ with magnetic field which could be due to the reinforcement of ASOC with magnetic field [26]. Therefore in view of the unusual behavior of LaRhSi$_3$ and the strong effect of ASOC in LaIrSi$_3$ revealed by the dHvA effect study, it was felt necessary to investigate the superconducting properties of LaIrSi$_3$ in detail, which we present in this paper.

Our $C_p(T)$ data confirm bulk superconductivity in LaIrSi$_3$ below $T_c = 0.72(1)$ K, in agreement with the report by Okuda et al. [33]. However, $\rho(T)$ exhibits superconductivity at a higher $T_c = 1.45$ K, apparently due to filamentary nonbulk superconductivity. The normal-state $\rho$ is metallic and well described by the Bloch-Grüneisen model of resistivity for $T \geq 1.6$ K. The low-$T$ normal-state $C_p(T)$ gives an electronic coefficient $\gamma_e = 4.60(2)$ mJ/mol K$^2$ and a density of states at Fermi energy $D(E_F) = 1.95(1)$ states/eV f.u. for both spin directions, where f.u. stands for formula unit. A sharp jump is observed at $T_c = 0.72(1)$ K in electronic heat capacity $C_e(T)$, with $\Delta C_e/\gamma_e T_c = 1.09(3)$, which is smaller than the BCS expected value of 1.43. Within the single-band picture the reduced value of $\Delta C_e/\gamma_e T_c$ can be attributed to an anisotropic energy gap in LaIrSi$_3$. We have analyzed the superconducting-state electronic heat-capacity data using the $\omega$ model of BCS superconductivity [38–40], which suggests that the s-wave order parameter of LaIrSi$_3$ is anisotropic in momentum space, with an energy gap $\Delta(0)/k_B T_c = 1.54(2)$. We have estimated various normal- and superconducting-state parameters that indicate a weak-coupling type-I BCS superconductivity in the dirty limit. Our $\mu$SR investigations also confirm type-I superconductivity in LaIrSi$_3$. Further, $\mu$SR results also show that the time-reversal symmetry is preserved as is expected for a conventional s-wave singlet pairing superconductivity. No evidence of parity mixing is observed as one would have expected in view of splitting of spin-up and spin-down energy bands revealed by dHvA study [33].

II. EXPERIMENTAL DETAILS

A polycrystalline sample of LaIrSi$_3$ was prepared by the standard arc melting of a stoichiometric mixture of high-purity elements (La: 99.9%, Ir: 99.99%, Si: 99.999%) on a water-cooled copper hearth under the titanium-gettered inert argon atmosphere with several flips to ensure homogeneity. The arc-melted sample was further heat treated at 900 °C for a week under the dynamic vacuum. The crystal structure was determined by the powder XRD using Cu $K_\alpha$ radiation. The heat-capacity measurements were performed by the relaxation method with a physical properties measurement system (PPMS, Quantum Design, Inc.). The electrical resistivity measurements were performed by the standard four-probe ac technique using the PPMS. Temperatures down to 0.35 K were attained using a $^3$He attachment to the PPMS.

Powder neutron diffraction (ND) measurement was performed at room temperature using the ROTAX diffractometer at the ISIS facility of the Rutherford Appleton Laboratory, Didcot, United Kingdom. The $\mu$SR measurements were carried out using the MuSR spectrometer at the ISIS facility with the detectors in both longitudinal and transverse configurations. A high-purity-silver plate was used to mount the sample, which gives only a nonrelaxing muon signal. The powdered sample was mounted on silver plate using diluted GE varnish that was covered with Kapton film. Temperatures down to 50 mK were achieved by cooling the sample in a dilution refrigerator. Correction coils were used to counter the effect of the stray fields at the sample position to within 1 $\mu$T.

III. CRYSTALLOGRAPHY

The room-temperature powder XRD data were analyzed by structural Rietveld refinement using the program FULLPROF [41]. The refinement confirmed the reported BaNiSn$_3$-
LaIrSi$_3$ recorded at room temperature. The solid line through the experimental points is the Rietveld refinement profile calculated for the noncentrosymmetric body-centered tetragonal BaNiSn$_3$-type structure (space group $I4mm$). The short vertical bars indicate the Bragg peak positions. The lowermost curve represents the difference between the experimental and calculated intensities.

The BaNiSn$_3$-type body-centered tetragonal structure (space group $I4mm$) of LaIrSi$_3$ and revealed the single-phase nature of the sample without any trace of the impurity phase. The single-phase nature of the whole bulk of the sample is further inferred from the Rietveld refinement of room-temperature powder neutron diffraction data that was performed using the program GSAS [42]. The neutron diffraction pattern and refinement profile for the noncentrosymmetric body-centered tetragonal BaNiSn$_3$-type structure are shown in Fig. 1. While refining, no improvement in the fit quality was observed upon refining the occupancies of atomic positions, and within the error bars the atomic occupancies were found to be unity; therefore in the final refinement we fixed the occupancies to unity. The crystallographic parameters obtained from the refinement of powder neutron diffraction are listed in Table I. Both ND and XRD data gave similar crystallographic parameters and agree well with the literature values [36,43].

The BaNiSn$_3$-type body-centered tetragonal structure (space group $I4mm$) of LaIrSi$_3$ is illustrated in Fig. 2 and is compared with the common ThCr$_2$Si$_2$-type body-centered tetragonal structure (space group $I4/mmm$). Like the ThCr$_2$Si$_2$-type structure, the BaNiSn$_3$-type structure is also a layered structure and a ternary derivative of BaAl$_4$-type structure [44]. The $R$ (La, Th) atoms occupy identical positions in both structures and form a body-centered tetragonal sublattice. However, they differ in the positions of $T$ (Ir, Cr) and Si atoms. The $T$ atoms form a square sublattice in the $ab$ plane in both structures, but they are rotated by 45° in the $ab$ plane with respect to each other. Further, in the ThCr$_2$Si$_2$-type structure all the Si atoms occupy a single crystallographic site, whereas in the BaNiSn$_3$-type structure the Si atoms occupy two different sites, and hence the stacking order of $T$ and Si layers along the $c$ axis is different in the two structures. The structural difference in the two structures is evident from Fig. 2. It is seen that the BaNiSn$_3$-type structure is not symmetric about the $R$ plane and there is a loss of the mirror plane along the $c$ axis in the BaNiSn$_3$-type structure which is present in the ThCr$_2$Si$_2$-type structure. The ThCr$_2$Si$_2$-type structure is centrosymmetric, whereas the BaNiSn$_3$-type structure is noncentrosymmetric.

### Table I. Crystallographic parameters obtained from the structural Rietveld refinement of room-temperature powder neutron diffraction data of LaIrSi$_3$.

| Structure            | BaNiSn$_3$-type tetragonal |
|----------------------|----------------------------|
| Space group          | $I4mm$ (No. 107)           |
| Lattice parameters   |                            |
| $a$ (Å)              | 4.2784(3)                  |
| $c$ (Å)              | 9.8308(7)                  |
| $V_{cell}$ (Å$^3$)   | 179.95(4)                  |

Atomic coordinates

| Atom | Wyckoff position | $x$  | $y$  | $z$     | $U_{iso}$ (Å$^2$) |
|------|------------------|------|------|---------|------------------|
| La   | 2a               | 0    | 0    | 0       | 0.0008(3)        |
| Ir   | 2a               | 0    | 0    | 0.6554(2)| 0.0021(3)        |
| Si1  | 2a               | 0    | 0    | 0.4140(3)| 0.0003(3)        |
| Si2  | 4b               | 0    | 1/2  | 0.2624(2)| 0.0033(3)        |

IV. ELECTRICAL RESISTIVITY

The electrical resistivity $\rho$ of LaIrSi$_3$ as a function of $T$ for 0.35 K $\leq T \leq 300$ K measured at applied magnetic field $H = 0$ is shown in Fig. 3(a). A metallic behavior is inferred from the $T$ dependence of $\rho$; $\rho$ decreases with decreasing $T$, becomes nearly constant in the low-$T$ limit below 25 K, and undergoes a sharp transition to a zero-resistance state due to the occurrence of superconductivity. It is seen that the onset of superconductivity takes place at $T_c(\text{onset}) \approx 1.6$ K, and the zero-resistivity state is reached at $T_c \approx 1.3$ K. Thus a $T_c = 1.45$ K (defined as the midpoint of the transition) is obtained from the resistivity data. The $\rho(T)$ data at various $H$ for $0 \leq H \leq$
represents the BG resistivity. \( \Theta_D \) is the Debye temperature from the resistivity data, and \( \mathcal{R} \) is a material-dependent prefactor.

Our fit of \( \rho(T) \) data in 1.6 K \( \leq T \leq 300 \) K by the BG model is shown by the solid red curve in Fig. 3(a), where we used an analytic Padé approximant fitting function for \( \rho_{BG} \) from Ref. [46]. From the fitting of \( \rho(T) \) data we obtain \( \rho_0 = 2.81(2) \mu \Omega \cdot \text{cm} \), \( \Theta_D = 331(2) \) K, and \( \mathcal{R} = 95.7 \mu \Omega \cdot \text{cm} \). Further details about the fitting of \( \rho(T) \) using the Bloch-Grüneisen model of resistivity can be found in Refs. [46,47].

V. HEAT CAPACITY

The heat capacity \( C_p \) of LaIrSi\(_3\) as a function of \( T \) for 0.35 K \( \leq T \leq 300 \) K measured at \( H = 0 \) is shown in Fig. 4(a). As shown in the inset, a sharp jump is observed in \( C_p \) due to the superconducting transition at \( T_c = 0.72(1) \) K. The observation of such a sharp jump in \( C_p(T) \) indicates the occurrence of bulk superconductivity in LaIrSi\(_3\). The \( C_p(T) \) data measured at different magnetic fields are shown in Fig. 4(b). It is seen that the jump \( \Delta C_p \) in \( C_p \) and \( T_c \) decrease with the increasing \( H \). \( T_c \) is found to decrease to 0.44(2) K at \( H = 5.0 \) mT from its value of \( T_c = 0.72(1) \) K at \( H = 0 \), and it is suppressed to a temperature below 0.35 K by a field of \( H = 7.0 \) mT. The suppression of \( T_c \) with \( H \) for \( C_p(T,H) \) is very different from that observed for the \( \rho(T,H) \) data above, where superconductivity survives even at an applied field of 0.4 T.

The low-temperature normal-state heat-capacity data are well described by \( C_p(T) = \gamma_n T + \beta T^3 \). The normal-state Sommerfeld electronic heat-capacity coefficient \( \gamma_n \) is estimated by fitting the normal-state \( C_p(T) \) data measured at \( H = 0 \) (at 0.75 K \( \leq T \leq 3.8 \) K) and at \( H = 10.0 \) mT (at 0.35 K \( \leq T \leq 0.9 \) K) simultaneously. The simultaneous linear fit of \( C_p(T)/T \) versus \( T^2 \) by \( C_p(T)/T = \gamma_n + \beta T^2 \) at 0.35 K \( \leq T \leq 3.8 \) K yields \( \gamma_n = 4.60(2) \) mJ/mole K\(^2\) and \( \beta = 0.17(1) \) mJ/mole K\(^2\). The coefficient \( \beta \) according to the relation \([48] \) \( \Theta_D = (12\pi^4 R_p^2/\beta) \)\(^{1/3}\), where \( R_p \) is the molar gas constant and \( \pi = 5 \) is the number of atoms per formula unit, gives the Debye temperature \( \Theta_D = 385(8) \) K. The experimental \( C_p(T = 300K) \approx 106 \) J/mole K does not reach the Dulong-Petit high-\( T \) limit of the lattice heat capacity \( C_V = 3pR = 15R = 124.7 J/mole K \).

The coefficient \( \gamma_n \) can be used to estimate the density of states at the Fermi level \( D(E_F) \), which, according to the relation \([48] \) \( \gamma_n = (\pi^2 k_B^2/3) D(E_F) \), gives \( D(E_F) = 1.95(1) \) states/eV f.u. for both spin directions. This \( D(E_F) \) contains the quasiparticle mass enhancement by many-body electron-phonon interaction and is related to the bare density of states \( D_{band}(E_F) \) by \([49] D(E_F) = (1 + \lambda_{e-ph}) D_{band}(E_F) \), where \( \lambda_{e-ph} \) is the electron-phonon coupling constant that can be estimated from \( \Theta_D \) and \( T_c \) using McMillan’s relation \([50] \),

\[
\lambda_{e-ph} = \frac{1.04 + \mu^* \ln(\Theta_D / \sqrt{1.45 T_c})}{(1 - 0.62 \mu^*) \ln(\Theta_D / 1.45 T_c) - 1.04}.
\]  

(3)

Here \( \mu^* \) is the repulsive screened Coulomb parameter usually assigned as \( \mu^* = 0.13 \). For LaIrSi\(_3\) we have \( T_c = 0.72 \) K and \( \Theta_D = 385 \) K, which together with \( \mu^* = 0.13 \), according to Eq. (3), give \( \lambda_{e-ph} = 0.41 \). The small value of \( \lambda_{e-ph} \) implies a weak-coupling superconductivity in LaIrSi\(_3\). This value of \( \lambda_{e-ph} \) combined with \( D(E_F) = 1.95(1) \) states/eV f.u. for both
The dotted red line marks the superconducting transition temperature $T_c$ in zero magnetic field. Inset: (b) $\alpha$-model superconductor state capacity as a function of temperature $T$. The solid red curve is the theoretical prediction of the $\alpha$-model for $\alpha = \Delta(0)/k_B T_c = 1.54$. The BCS prediction for $\alpha_{BCS} = 1.764$ is also shown for comparison.

VI. SUPERCONDUCTING-STATE PROPERTIES

The electronic contribution to the heat capacity $C_e(T)$ after subtracting the phonon contribution from the measured $C_p(T)$ data, i.e., $C_e(T) = C_p(T) - \beta T^3$, is shown in Fig. 4(c), which clearly shows the sharp jump in $C_e$ at $T_c$. The jump $\Delta C_e$ in $C_e$ at $T_c$ is found to be $\Delta C_e = 3.6(1) \text{ mJ/mol K}$, corresponding to the vertical dotted line at $T_c$ in Fig. 4(c). This gives $\Delta C_e/\gamma_n T_c = 1.09(3)$ for $\gamma_n = 4.60(2) \text{ mJ/mol K}^2$ and $T_c = 0.72(1) \text{ K}$, which is significantly smaller than the BCS value of $\Delta C_e/\gamma_n T_c = 1.43$ in the weak-coupling limit [51]. The presence of a residual heat capacity due to a small impurity/nonsuperconducting phase can lead to a reduced $\Delta C_e/\gamma_n T_c$. However, in view of the fact that the jump in $C_e(T)$ at the superconducting transition is very sharp, the entire sample seems to be superconducting without any residual $\gamma$, which in turn suggests that the reduction in $\Delta C_e/\gamma_n T_c$ is intrinsic. In a single-band model, such a reduction can be caused by the presence of an anisotropic superconducting energy gap (order parameter) in momentum space [38]. That the reduction in $\Delta C_e/\gamma_n T_c$ is intrinsic will be clear from our analysis of the superconducting-state data using the single-band $\alpha$ model of BCS superconductivity [38–40] below, which is applicable to the system with $\Delta C_e/\gamma_n T_c \neq 1.43$.

In the so-called $\alpha$ model of BCS superconductivity in order to fit the superconducting-state thermodynamic data, $\alpha_{BCS} \equiv \Delta(0)/k_B T_c = 1.764$ is replaced by a variable $\alpha$ [38,39]. $\alpha$ is determined from the jump $\Delta C_e$ at $T_c$ according to the relation [38]

$$\frac{\Delta C_e(T_c)}{\gamma_n T_c} = 1.426 \left(\frac{\alpha}{\alpha_{BCS}}\right)^2,$$

which for $\Delta C_e/\gamma_n T_c = 1.09(3)$ gives $\alpha = 1.54(2)$. This value of $\alpha$ is significantly smaller than the BCS value of 1.764. The temperature dependence of $\alpha$-model superconducting-state heat capacity $C_e(T)$ calculated for $\alpha = 1.54$ is shown by the solid red curve in Fig. 4(c) together with that of the BCS prediction for $\alpha = \alpha_{BCS}$. A reasonable agreement is observed.
between the $\alpha$-model prediction and the superconducting-state $C_s(T)$ data, which supports the applicability of the $\alpha$ model and in turn indicates that the $s$-wave order parameter of LaIrSi$_3$ is anisotropic in momentum space. The details about the fitting of $C_s(T)$ data by the $\alpha$ model of BCS superconductivity can be found in Refs. [38,52]. The lack of perfect agreement between the $\alpha$-model prediction and the experimental data may indicate that the anisotropy of the gap is not well accounted for, as the simplified $\alpha$ model does not account for the energy dependence of the gap function or for a complex $\Delta$.

The thermodynamic critical field $H_c$ is estimated from the zero-field $C_s(T)$ data by integrating the entropy difference between the superconducting and normal states [51,53],

$$H_c^2(T) = 8\pi \int_0^T [S_n(T') - S_s(T')]dT',$$

where $S_n$ and $S_s$ are the electronic entropy of normal and superconducting states, respectively, with $S_s(T') = \int_0^{T'} [C_s(T'/T'')]dT''$. $H_c(T)$ obtained this way from the zero-field $C_s(T)$ is shown in Fig. 5. The $T$ dependence of $H_c$ can be approximated to the conventional relation $H_c(T) = H_c(0)[1 - (T/T_c)]^2$, which gives $H_c(0) = 6.10(3)$ mT. The solid red curve in Fig. 5 represents the fit of $H_c(T)$ data with this expression.

The experimental $H_c(0)$ so obtained is somewhat higher than the theoretical $H_c(0)$, which for the $\alpha$ model is given by [38]

$$\frac{H_c(0)}{(\gamma_N T_c^2)^{1/2}} = \sqrt{\frac{6}{\pi}} \alpha \approx 1.382 \alpha,$$

where the Sommerfeld coefficient per unit volume $\gamma_N$ is in units of erg/cm$^3$ K$^2$. From this relation for $\alpha = 1.54$ we obtain $H_c(0) = 4.5$ mT, which is a little lower than $H_c(0) = 6.10(3)$ mT obtained above. Even for $\alpha_{BCS} = 1.764$, Eq. (6) gives a lower $H_c(0) = 5.1$ mT. The reason for this discrepancy between the experimental and theoretical values of $H_c(0)$ is not clear. We suspect that this might be the result of a nonspherical/anisotropic Fermi surface in LaIrSi$_3$ which is not properly accounted by equation (6).

Further, as can be seen from Fig. 5, $H_c(T)$ obtained from the zero-field $C_s(T)$ data and the $H$-$T$ phase diagram determined from the $H$-dependent $T_c$ from $C_p(T, H)$ data in Fig. 4(b) both give very low critical fields. For a type-II superconductivity $T_c(H)$ obtained from $C_p(T, H)$ gives the upper critical field $H_{c2}$, which is usually much higher than the thermodynamic critical field, which is not the present case. The fact that $H_{c2}(T)$ is close to $H_c(T)$ may suggest a type-I superconductivity in LaIrSi$_3$ or a type-1.5 behavior that may arise from the presence of split spin-up and spin-down energy bands similar to what has been observed in two-band superconductor MgB$_2$ [54].

The $H$-dependent $T_c$ from $\rho(T, H)$ data in Fig. 3(b) is shown in the inset of Fig. 5, which shows a very different behavior than $T_c(H)$ from $C_p(T, H)$ data. This is consistent with the observation of different $T_c$ in resistivity and heat-capacity measurements in zero field mentioned above, apparently due to the filamentary/surface superconductivity that sets in at a temperature higher than the bulk superconductivity. Our estimate of the Ginzburg-Landau parameter $\kappa$ below gives $\kappa = 0.55$, which suggests a type-I behavior. Usually, for a type-I superconductor we do not expect a filamentary or surface superconductivity. However, surface superconductivity is predicted for a type-I superconductor with $\kappa$ values between $1/\sqrt{2}$ and $1/2.39$ [55,56]. Indeed, our estimated $\kappa$ lies between these limits, and we can expect a surface superconductivity in LaIrSi$_3$. The critical field associated with the surface superconductivity is given by [55,56] $H_{c3} = 2.39\kappa H_c$; accordingly, for LaIrSi$_3$ we estimate $H_{c3} = 8.0$ mT, which is much smaller than the observed upper critical field from the resistivity measurement (inset of Fig. 5). Thus the observed $T_c(H)$ from $\rho(T, H)$ could not be understood to arise from surface superconductivity. The reason for such a high critical field in the resistivity measurement of LaIrSi$_3$ is not clear. The type-I superconductor LaPdSi$_3$ was also found to exhibit a higher critical field in the resistivity measurement compared to that in the heat capacity [31]. In a recent study Kimura et al. [57] found a similar high critical field from the resistivity measurement on the type-I superconductor LaRhSi$_3$, and they argue that this behavior may be a common feature in noncentrosymmetric superconductors. However, we are not aware of any theoretical discussion of this aspect of noncentrosymmetric superconductors, and this hypothesis needs to be tested theoretically.

Our estimate of the superconducting London penetration depth in the clean limit at $T = 0$, $\lambda_L(0)$ from $\nu_F$ using the relation [51]

$$\frac{\lambda_L(0)}{\nu_F} = \frac{m^* c^2}{4\pi n e^2} = \frac{3\pi^2 c^2 h^3}{4m^* e^2 \nu_F},$$

where $c$ is the speed of light in vacuum, gives $\lambda_L(0) = 25.9$ nm for $\nu_F = 9.49 \times 10^7$ cm/s. The BCS coherence length $\xi_0$ can be obtained from $\nu_F$ and the energy gap $\Delta(0)$, which for the $\alpha$ model is given by [38]

$$\xi_0 = \frac{\hbar \nu_F}{\pi \alpha} \frac{\hbar \nu_F}{\Delta(0)} \frac{1}{\kappa T_c}.$$
This gives $\xi_0 = 2081$ nm for $\alpha = 1.54$ and $v_F = 9.49 \times 10^3$ cm/s. We see that $\xi_0$ is much larger than the above-estimated mean free path $\ell = 33.7$ nm, $\ell/\xi_0 \approx 0.016 < 1$, suggesting that the superconductivity in LaIrSi$_3$ is in the dirty limit. In the dirty limit, the Ginzburg-Landau parameter $\kappa_{GL} = 0.715 \lambda_1(0)/\ell$ [51], which gives $\kappa_{GL} = 0.55 < 1/\sqrt{2}$, as expected for a type-I superconductivity. This value of $\kappa_{GL}$ is close to the value obtained using the dirty-limit relation for a fully gapped (isotropic) superconductor [58], $\kappa_{GL} = 7.49 \times 10^3 \rho_0 \sqrt{\gamma_{SV}}$, with $\rho_0$ in $\Omega$ cm, which gives $\kappa_{GL} = 0.59$. Both estimates of $\kappa_{GL}$ consistently indicate a type-I superconductivity in LaIrSi$_3$.

The effective magnetic penetration depth $\lambda_{eff}$ can be estimated using the relation [51]

$$\lambda_{eff}(0) = \lambda_1(0) \sqrt{1 + \frac{\xi_0}{\ell}} \quad (\text{dirty limit}),$$

which gives $\lambda_{eff}(0) = 205$ nm. Then, using the relation $\kappa_{GL} = \lambda_{eff}(0)/\xi(0)$, we estimate the Ginzburg-Landau coherence length $\xi(0)$, which for $\kappa_{GL} = 0.55$ yields $\xi(0) = 373$ nm. The measured and derived superconducting parameters of LaIrSi$_3$ are listed in Table II.

### VII. MUON SPIN RELAXATION AND ROTATION

The time evolution of muon spin relaxation in zero field (ZF) is shown in Fig. 6 for $T$ both above and below the bulk $T_c$. It is evident from the ZF $\mu$SR spectra that there is no noticeable change in the relaxation rates at 1.0 K ($\geq T_c$) and 0.05 K ($< T_c$). This indicates that the time-reversal symmetry remains preserved upon entering the superconducting state. The ZF $\mu$SR spectra are best described by the Gaussian Kubo-Toyabe function,

$$G_z(t) = A_0 \left[ \frac{1}{2} + \frac{2}{3}(1 - \sigma^2 t^2) e^{-\sigma^2 t^2/2} \right] e^{-\lambda t} + A_{BG},$$

where $A_0$ is the initial asymmetry, $\sigma$ and $\lambda$ are the depolarization rates, and $A_{BG}$ is the time-independent background contribution. $\sigma$ accounts for the Gaussian distribution of static fields from nuclear moments (the local field distribution width $H_{zz} = \sigma/\gamma_{\mu}$, with muon gyromagnetic ratio $\gamma_{\mu} = 135.53$ MHz/T), and $\lambda$ accounts for the electronic moments. The fits of $\mu$SR spectra in Fig. 6 by the decay function in Eq. (10) give $\sigma = 0.074(1) \mu s^{-1}$ and $\lambda = 0.009(3) \mu s^{-1}$ at 1.0 K and $\sigma = 0.074(1) \mu s^{-1}$ and $\lambda = 0.011(2) \mu s^{-1}$ at 0.05 K. The fits are shown by the solid red curve in Fig. 6. Since within the error bars both $\sigma$ and $\lambda$ at $T < T_c$ and $T > T_c$ are similar, there is no evidence of time-reversal symmetry breaking in LaIrSi$_3$.

The time evolution of muon spin rotation in transverse field (TF) is shown in Fig. 7. The TF muon spin precession signals were collected on a field-cooled sample at different applied fields both above (1.0 K) and below (0.05 K) $T_c$. The TF $\mu$SR spectra are best described by an oscillatory function damped with a Gaussian and an oscillatory background, i.e., by

$$G_z(t) = A_0 \cos(\omega t + \varphi)e^{-\sigma^2 t^2/2} + A_{BG} \cos(\omega t + \varphi),$$

where $\omega = \gamma_{\mu} H_{int}$ is the precession frequency ($H_{int}$ is the internal field at the muon site). Solid curves in Fig. 7 are the fits of the TF $\mu$SR spectra by the decay function in Eq. (11). In the superconducting state at $T = 0.05$ K the depolarization rate is found to increase significantly; for example, for the TF $\mu$SR spectra at 2.5 mT, $\sigma$ increases from its value $\sigma = 0.010(2) \mu s^{-1}$ at 1.0 K to $\sigma = 1.45(4) \mu s^{-1}$ at 0.05 K. Such an increase of the depolarization rate reveals bulk superconductivity in LaIrSi$_3$.

The maximum entropy spectra for TF $\mu$SR precession at 1.0 and 0.05 K are shown in Fig. 8. The maximum entropy spectra depict the magnetic field probability distribution $P(H)$. It is clear from Fig. 8 that at 1.0 K (in the normal state) sharp peaks are observed at $H_{int}$ exactly equal to the applied $H$, whereas at 0.05 K (in the superconducting state) one can see additional peaks. At $H = 4.0$ mT, $P(H)$ at 0.05 K shows an additional peak at $H_{int} > H$ (inset of Fig. 8). The appearance of an additional peak (near 5.5 mT, which gives an estimation of $H_{c1}$) at an internal field greater than the applied $H$ is a characteristic

| Parameter                  | Value       |
|----------------------------|-------------|
| $T_c$ (K)                  | 0.72(1)     |
| $\gamma_0$ (mJ/mol K$^2$) | 4.60(2)     |
| $\Theta_D$ (K)            | 385(8)      |
| $\lambda_{iso}$           | 0.41        |
| $\Delta C_e$ (mJ/mol K)   | 3.6(1)      |
| $\Delta C_e/\gamma_0 T_c$| 1.09(3)     |
| $\alpha = \Delta(0)/k_B T_c$ (from $\Delta C_e/\gamma_0 T_c$) | 1.54(2)     |
| $\Delta(0)/k_B$ (K)       | 1.11        |
| $H_c(T = 0)$ (mT)          | 6.10(3)     |
| $\kappa_{GL}$             | 0.55        |
| $\xi(0)$ (nm)             | 373         |
| $\xi_0$ (nm)              | 2081        |
| $\ell$ (nm)               | 33.7        |
| $\lambda_1(0)$ (clean limit) (nm) | 25.9 |
| $\lambda_{eff}(0)$ (dirty limit) (nm) | 205 |
also observe an increase in $P(H)$ is observed from the Meissner volume in the intermediate state (i.e., at $H = 5.0$ mT). At $H = 6.0$ mT no additional peak is observed in $P(H)$ at $H_{\text{int}} > H$ even on an expanded scale. This could be understood to be due to the fact that the applied $H$ is close to $H_c = 6.1$ mT (see Table II) and the sample is on the verge of a transition from a superconducting to a normal state at $H = 6.0$ mT. Thus the $\mu$SR data also reflect a low thermodynamic critical field $H_c \approx 5.5$ mT, in line with the estimate of $H_c$ from the heat-capacity data.

VIII. CONCLUSIONS

The superconducting and normal-state properties of the noncentrosymmetric superconductor LaIrSi$_3$, which crystallizes in a BaNiSn$_3$-type tetragonal crystal structure (space group $I4_{1}mm$), were investigated using $C_p(T, H)$, $\rho(T, H)$, and $\mu$SR measurements which demonstrate bulk BCS superconductivity below $T_c = 0.72(1)$ K. A nonbulk superconductivity sets in at a higher $T_c$ in $\rho(T)$. In the normal state $\rho$ exhibits metallic behavior, and $\rho(T \geq 1.6$ K) is well described by the Bloch-Grüneisen model of resistivity. Our analysis of low-temperature normal-state $C_p(T)$ data yields a Sommerfeld coefficient $\gamma_e = 4.60(2) \text{ mJ/mol K}^2$, corresponding to the density of states at Fermi energy $D(E_F) = 1.95(1)$ states/eV f.u. for both spin directions.

The superconducting transition is revealed by a very sharp jump in $C_p$ at $T_c = 0.72(1)$ K, however, with a value of $\Delta C_e/\gamma_e T_c = 1.09(3)$ lower than the BCS expected value of 1.43. The reduced value of $\Delta C_e/\gamma_e T_c$ seems to indicate an anisotropic energy gap in LaIrSi$_3$. The superconducting-state electronic heat-capacity data are analyzed by the single-band $\alpha$ model of BCS superconductivity that describes the experimental data reasonably. $\alpha = \Delta(0)/k_B T_c = 1.54(2)$ obtained from the jump in $C_p$ is smaller than $\alpha_{\text{BCS}} = 1.764$, indicating the $s$-wave order parameter of LaIrSi$_3$ is anisotropic in momentum space. Even though the single-band $\alpha$ model describes the superconducting-state data reasonably, considering the split spin-up and spin-down bands in LaIrSi$_3$, the possibility of two-band superconductivity cannot be ruled out. The presence of two bands is also known to result in a reduced $\Delta C_e/\gamma_e T_c$ [38, 59]. Since the two-band effect and an anisotropy modification to a single band have similar manifestations, it is very difficult to distinguish between them by examining the thermodynamic quantities. Recently, a two-band superconductivity with equal energy gaps was reported in SrPt$_3$P [60]. Further investigations are desired to check for the possibility of a similar two-band single-gap superconductivity in LaIrSi$_3$.

Various normal- and superconducting-state parameters have been estimated which indicate a dirty-limit weak-coupling type-I $s$-wave BCS superconductivity in LaIrSi$_3$. Type-I superconductivity is further confirmed by $\mu$SR. The $\mu$SR measurement also revealed that the time-reversal symmetry is preserved in the superconducting state, thus confirming a conventional $s$-wave singlet pairing superconductivity in LaIrSi$_3$. Thus, despite a large splitting of Fermi surfaces due to antisymmetric coupling on account of the noncentrosymmetric structure inferred from the de Haas–van Alphen effect study [33], no clear signature of parity mixing or a
The effect of the lack of inversion symmetry on the superconducting properties of LaIrSi\(_3\) does not seem to be desired. This aspect of superconductivity in these compounds are what one would expect the electron-phonon-mediated type-I superconductivity with pre-ecd model to be. However, we see that the \(T_c\) of these two superconductors is higher than those with 5d. Further investigations to understand this aspect of superconductivity in these compounds are desired.

The effect of the lack of inversion symmetry on the superconducting properties of LaIrSi\(_3\) does not seem to be pronounced despite a large splitting of spin-up and spin-down energy bands due to ASOC. Similar behavior is reported for NCS BaPdSi\(_3\) [21], for which electronic-structure calculations revealed the splitting of bands because of spin-orbit interactions; however, the superconducting transition turned to be conventional BCS-like with a singlet parring. Such observations raise an important question: what else besides ASOC controls the appearance of anomalous superconducting state in a noncentrosymmetric system? The anomalous superconducting properties of the Ce-based strongly correlated NCSs, such as CePt\(_3\)Si [9], CeRhSi\(_3\) [13], and CeIrSi\(_3\) [14] cannot be related to the magnetic pairing due to the presence of 4f moments. However, the unusual superconducting properties of weakly correlated NCSs Li\(_2\)Pt\(_3\)B [11], LaNiC\(_2\) [19,20], and Re\(_2\)Zr [30] are not then obvious as they do not show any evidence of magnetic order. An important difference between the two groups of nonmagnetic NCSs, i.e., those exhibiting unusual superconducting properties and those exhibiting conventional superconductivity, is the difference in their crystal structures/space groups. This may suggest that these two groups of nonmagnetic NCSs may have different Fermi surface topologies that may have some role in realizing the effect of ASOC. A comparative study of the extent of ASOC in nonmagnetic NCSs, preferably by a technique that can directly probe Fermi surface topology, such as angle-resolved photoemission spectroscopy, complemented with band-structure calculations can shed light on this issue and would be of help in understanding the relationship between ASOC, Fermi surface topology, and anomalous superconductivity in NCSs.

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[1] Non-centrosymmetric Superconductors: Introduction and Overview, edited by E. Bauer and M. Sigrist, Lecture Notes in Physics (Springer, Berlin, 2012), Vol. 847.

[2] V. M. Edel’shtein, Zh. Eksp. Teor. Fiz. 95, 2151 (1989) [Sov. Phys. JETP 68, 1244 (1989)].

[3] L. P. Gor’kov and E. I. Rashba, Phys. Rev. Lett. 87, 037004 (2001).

[4] K. V. Samokhin, E. S. Zijlstra, and S. K. Bose, Phys. Rev. B 69, 094514 (2004).

[5] P. A. Frigeri, D. F. Agterberg, A. Koga, and M. Sigrist, Phys. Rev. Lett. 92, 097001 (2004).

[6] S. Fujimoto, J. Phys. Soc. Jpn. 76, 051008 (2007).

[7] E. Bauer, G. Hilscher, H. Michor, C. Paul, E. W. Scheidt, A. Gribanov, Y. Seropegin, H. Noel, M. Sigrist, and P. Rogl, Phys. Rev. Lett. 92, 027003 (2004).

[8] E. Bauer, I. Bonalde, and M. Sigrist, Low Temp. Phys. 31, 748 (2005).

[9] E. Bauer, H. Kaldarar, A. Prokofiev, E. Royanian, A. Amato, J. Sereni, W. Brammer-Escamilla, and I. Bonalde, J. Phys. Soc. Jpn. 76, 051009 (2007).

[10] K. Togano, P. Badica, Y. Nakamori, S. Orimo, H. Takeya, and K. Hirata, Phys. Rev. Lett. 93, 247004 (2004).

[11] H. Q. Yuan, D. F. Agterberg, N. Hayashi, P. Badica, D. Vanderveld, K. Togano, M. Sigrist, and M. B. Salamon, Phys. Rev. Lett. 97, 017006 (2006).

[12] N. Kimura, K. Itou, K. Saitoh, Y. Umeda, H. Aoki, and T. Terashima, Phys. Rev. Lett. 95, 247004 (2005).

[13] N. Kimura, Y. Muro, and H. Aoki, J. Phys. Soc. Jpn. 76, 051010 (2007).

[14] I. Sugitani, Y. Okuda, H. Shishido, T. Yamada, A. Thamizhavel, E. Yamamoto, T. D. Matsuda, Y. Haga, T. Takeuchi, R. Settai, and Y. Önuki, J. Phys. Soc. Jpn. 75, 043703 (2006).

[15] R. Settai, I. Sugitani, Y. Okuda, A. Thamizhavel, M. Nakashima, Y. Önuki, and H. Harima, J. Magn. Magn. Mater. 310, 844 (2007).

[16] G. Knebel, D. Aoki, G. Lapertot, B. Salce, J. Fluquet, T. Kawai, H. Muranaka, R. Settai, and Y. Önuki, J. Phys. Soc. Jpn. 78, 074714 (2009).

[17] F. Honda, I. Bonalde, K. Shimizu, S. Yoshiuchi, Y. Hirose, T. Nakamura, R. Settai, and Y. Önuki, Phys. Rev. B 81, 140507 (2010).

[18] V. K. Pecharsky, L. L. Miller, K. A. Gschneider, Phys. Rev. B 85, 497 (1998).

[19] A. D. Hillier, J. Quintanilla, and R. Cywinski, Phys. Rev. Lett. 102, 117007 (2009).

[20] I. Bonalde, R. L. Ribeiro, K. J. Syu, H. H. Sung, and W. H. Lee, New J. Phys. 13, 123022 (2011).

[21] E. Bauer, R. T. Khan, H. Michor, E. Royanian, A. Grytsiv, N. Melnychenko-Koblyuk, P. Rogl, D. Reith, R. Podloucky, E.-W. Scheidt, W. Wolf, and M. Marsman, Phys. Rev. B 80, 064504 (2009).

[22] T. Shibayama, M. Nohara, H. A. Kotari, Y. Okamoto, Z. Hiroi, and H. Takagi, J. Phys. Soc. Jpn. 76, 073708 (2007).

[23] K. Wakui, S. Akutagawa, K. Kawashima, T. Morita, K. Nishida, H. Iwahori, J. Abe, and J. Akimitsu, J. Phys. Soc. Jpn. 78, 034710 (2009).

[24] T. Klimczuk, F. Ronning, V. Sidorov, R. J. Cava, and J. D. Thompson, Phys. Rev. Lett. 99, 257004 (2007).
[25] A. B. Karki, Y. M. Xiong, I. Vekhter, D. Browne, P. W. Adams, D. P. Young, K. R. Thomas, J. Y. Chan, H. Kim, and R. Prozorov, Phys. Rev. B 82, 064512 (2010).

[26] V. K. Anand, A. D. Hillier, D. T. Adroja, A. M. Strydom, H. Michor, K. A. McEwen, and B. D. Rainford, Phys. Rev. B 83, 064522 (2011).

[27] G. Eguchi, D. C. Peets, M. Kriener, and Y. Maeno, E. Nishibori, Y. Kumazawa, K. Banno, S. Maki, and H. Sawa, Phys. Rev. B 83, 024512 (2011).

[28] P. K. Biswas, M. R. Lees, A. D. Hillier, R. I. Smith, W. G. Marshall, and D. McK. Paul, Phys. Rev. B 84, 184529 (2011).

[29] A. B. Karki, Y. M. Xiong, N. Haldolaarachchige, S. Stadler, I. Vekhter, P. W. Adams, D. P. Young, W. A. Phelan, and J. Y. Chan, Phys. Rev. B 83, 144525 (2011).

[30] R. P. Singh, A. D. Hillier, B. Mazidian, J. Quintanilla, J. F. Annett, D. McK. Paul, G. Balakrishnan, and M. R. Lees, Phys. Rev. Lett. 112, 107002 (2014).

[31] M. Smidman, A. D. Hillier, D. T. Adroja, M. R. Lees, V. K. Anand, R. P. Singh, R. I. Smith, D. M. Paul, and G. Balakrishnan, Phys. Rev. B 89, 094509 (2014).

[32] F. von Rohr, H. Luo, N. Ni, M. Wörle, and R. J. Cava, Phys. Rev. B 89, 224504 (2014).

[33] Y. Okuda, Y. Miyauchi, Y. Ida, Y. Takeda, C. Tonohiro, Y. Ouduchi, T. Yamada, N. D. Dung, T. D. Matsuda, Y. Haga, T. Takeuchi, M. Hagiwara, K. Kindo, H. Harima, K. Sugiyama, R. Settai, and Y. Ōnuki, J. Phys. Soc. Jpn. 76, 044708 (2007).

[34] Y. Tada, N. Kawakami, and S. Fujimoto, Phys. Rev. B 81, 104506 (2010).

[35] A. Thamizhavel, T. Takeuchi, T. D. Matsuda, Y. Haga, K. Sugiyama, R. Settai, and Y. Ōnuki, J. Phys. Soc. Jpn. 74, 1858 (2005).

[36] P. Lejay, I. Higashi, B. Chevalier, J. Etourneau, and P. Hagenmuller, Mater. Res. Bull. 19, 115 (1984).

[37] P. Haen, P. Lejay, B. Chevalier, B. Lloret, J. Etourneau, and M. Sera, J. Less Common Met. 110, 321 (1985).

[38] D. C. Johnston, Supercond. Sci. Technol. 26, 115011 (2013).

[39] H. Padamsee, J. E. Neighbor, and C. A. Shiffman, J. Low Temp. Phys. 12, 387 (1973).

[40] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. 108, 1175 (1957).

[41] J. Rodríguez-Carvajal, Physica B 192, 55 (1993); FULLPROF: LLB-JRC, Laboratoire Léon Brillouin, CEA-Saclay, Gif-sur-Yvette, France, 1996.

[42] A. C. Larson and R. B. Von Dreele, Los Alamos National Laboratory, Technical Report No. LAUR 86-748, 2004; B. H. Toby, J. Appl. Cryst. 34, 210 (2001).

[43] N. Engel, H. F. Braun, and E. Parthé, J. Less Common Met. 95, 309 (1983).

[44] E. Parthé, B. Chabot, H. F. Braun, and N. Engel, Acta Crystallogr. B 39, 588 (1983).

[45] F. J. Blatt, Physics of Electronic Conduction in Solids (McGraw-Hill, New York, 1968).

[46] R. J. Goetsch, V. K. Anand, A. Pandey, and D. C. Johnston, Phys. Rev. B 85, 054517 (2012).

[47] V. K. Anand, P. K. Perera, A. Pandey, R. J. Goetsch, A. Kreyssig, and D. C. Johnston, Phys. Rev B 85, 214523 (2012).

[48] C. Kittel, Introduction to Solid State Physics, 8th ed. (Wiley, New York, 2005).

[49] G. Grimvall, Phys. Scr. 14, 63 (1976).

[50] W. McMillan, Phys. Rev. 12, 331 (1968).

[51] M. Tinkham, Introduction to Superconductivity, 2nd ed. (Dover, Mineola, NY, 1996).

[52] V. K. Anand, H. Kim, M. A. Tanatar, R. Prozorov, and D. C. Johnston, Phys. Rev. B 87, 224510 (2013).

[53] P. G. de Gennes, Superconductivity of Metals and Alloys (Benjamin, New York, 1966).

[54] V. Moshchalkov, M. Menghini, T. Nishio, Q. H. Chen, A. V. Silhanek, V. H. Dao, L. F. Chibotaru, N. D. Zhigadlo, and J. Karpinski, Phys. Rev. Lett. 102, 117001 (2009).

[55] D. Saint-James and P. G. de Gennes, Phys. Lett. 7, 306 (1963).

[56] M. Strongin, A. Paskin, D. G. Schweitzer, O. F. Kammerer, and P. P. Craig, Phys. Rev. Lett. 12, 442 (1964).

[57] N. Kimura, H. Ogi, K. Satoh, G. Ohsaki, K. Saitoh, H. Iida, and H. Aoki, JPS Conf. Proc. 3, 015011 (2014).

[58] T. P. Orlando, E. J. McNiff, Jr., S. Foner, and M. R. Beasley, Phys. Rev. B 39, 442 (1964).

[59] M. Zehetmayer, Supercond. Sci. Technol. 26, 043001 (2013).

[60] R. Khasanov, A. Amato, P. K. Biswas, H. Luetskens, N. D. Zhigadlo, and B. Batlogg, arXiv:1404.5473. 014513-10