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Weak interband-coupling superconductivity in the filled skutterudite $LaPt_4Ge_{12}$

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The superconducting pairing state of $LaPt_4Ge_{12}$ is studied by measuring the magnetic penetration depth $\lambda(T,B)$ and superfluid density $\rho_s(T)$ using a tunnel-diode-oscillator (TDO)-based method and transverse-field muon-spin rotation (TF-μSR) spectroscopy. The penetration depth follows an exponential-type temperature dependence at $T \ll T_c$, but increases linearly with magnetic field at $T = 1.5$ K. A detailed analysis demonstrates that both $\lambda_L(T)$ and $\rho_s^{\mathrm{TDO}}(T)$, measured in the Meissner state using the TDO method, are well described by a two-gap γ model with gap sizes of $\Delta_1(0) = 1.31k_BT_c$ and $\Delta_2(0) = 1.80k_BT_c$, and weak interband coupling. In contrast, $\rho_s^{\mu\mathrm{SR}}(T)$, derived from the μSR data, can be fitted by a single-gap BCS model with a gap close to $\Delta_2(0)$. We conclude that $LaPt_4Ge_{12}$ is a marginal two-gap superconductor and the small gap Δ_1 seems to be suppressed by a small magnetic field applied in the μSR experiments. In comparison, the $4f$ electrons in $PrPt_4Ge_{12}$ may enhance the interband coupling and, therefore, give rise to more robust multiband superconductivity.

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The discovery of superconductivity (SC) in a series of Pr-based skutterudite compounds, i.e., PrT_4X_{12} (T = Fe, Ru, Os, and X = pnictogen), has attracted considerable interest. Despite intensive investigations of the physical properties of these compounds, their superconducting order parameter and, therefore, the pairing mechanism remains highly controversial [1], with evidence for nodal superconductivity in some compounds and conventional BCS SC in others. Even for the same compound, most prominently, the heavy-fermion superconductor $PrOs_4Sb_{12}$, the gap symmetry is still under debate. Here, the early measurements provided evidence of point nodes with a possible triplet pairing state [2–4]. Upon improving the sample homogeneity and measuring to lower temperatures, recent measurements give evidence for multiband SC in $PrOs_4Sb_{12}$ [5,6]. Exploration of other filled skutterudite superconductors may help to elucidate the superconducting pairing state and the question of its universality.

A family of Ge-based skutterudites with T_c ranging from 5 to 8.3 K, i.e., MPt_4Ge_{12} (M = Sr, Ba, La, Pr), was discovered a few years ago [7–10]. Much as $PrOs_4Sb_{12}$, zero-field muon-spin rotation (μSR) experiments showed evidence for time-reversal symmetry (TRS) breaking in the superconducting state of $PrPt_4Ge_{12}$, but not for $LaPt_4Ge_{12}$ [11–13]. However, more recent experiments suggested multigap SC for $PrPt_4Ge_{12}$ [14–16]. Furthermore, a coherence peak was observed in the nuclear spin-lattice relaxation rate $1/T_1$ just below T_c for $PrPt_4Ge_{12}$, which is unexpectedly suppressed in $LaPt_4Ge_{12}$ [17]. However, the weak variation of T_c and of the specific heat jump at T_c with the Pr content x in $La_{1-x}Pr_xPt_4Ge_{12}$ suggests that the order parameters of the two end members are similar [12]. Thus, $LaPt_4Ge_{12}$ is not a simple reference compound of $PrPt_4Ge_{12}$, as seen in many heavy-fermion systems. In order to clarify the superconducting order parameter in $LaPt_4Ge_{12}$, further investigations are badly needed. Furthermore, a comparative study of $PrPt_4Ge_{12}$ and $LaPt_4Ge_{12}$ could shed new light on the role of Pr-$4f$ electrons in the formation of SC and, therefore, provide a more general picture of the pairing states in skutterudite superconductors.

In this Rapid Communication, we report a comprehensive study of the magnetic penetration depth $\lambda(T,B)$ and superfluid density $\rho_s(T)$ of $LaPt_4Ge_{12}$ using μSR and a tunnel-diode-oscillator (TDO)-based technique. The London penetration depth $\lambda_L(T)$, measured by the TDO method, follows exponential-type behavior in the low-temperature limit, suggesting nodeless SC for $LaPt_4Ge_{12}$. The penetration depth $\lambda_m(B)$, determined from the μSR experiments at T = 1.5 K, follows a linear field dependence, giving evidence for multigap SC. The superfluid density $\rho_s^{\mathrm{TDO}}(T)$, converted from $\lambda_L(T)$, is best described by a two-gap γ model, with gap sizes of $\Delta_1(0) = 1.31k_BT_c$ and $\Delta_2(0) = 1.80k_BT_c$ at $T = 0$. However, the superfluid density $\rho_s^{\mu\mathrm{SR}}(T)$ derived from μSR with a transverse field of 75 mT is fitted by a conventional BCS model with a gap of $\Delta(0) \approx \Delta_2(0)$. We argue that marginal two-gap SC exists in $LaPt_4Ge_{12}$ and also some other skutterudite superconductors. Due to very weak interband coupling in $LaPt_4Ge_{12}$, the small gap may be destroyed by a weak magnetic field (as used in our μSR measurements) or other perturbations.

Polycrystalline samples of $LaPt_4Ge_{12}$, as described in Ref. [7], were used for the μSR experiments, which were performed using the πM3 beamline at the Paul Scherrer Institute (Switzerland) down to temperatures of T = 1.5 K on the General Purpose Surface-Muon instrument (GPS) and $T \approx$ 0.02 K on the Low Temperature Facility (LTF) spectrometer.

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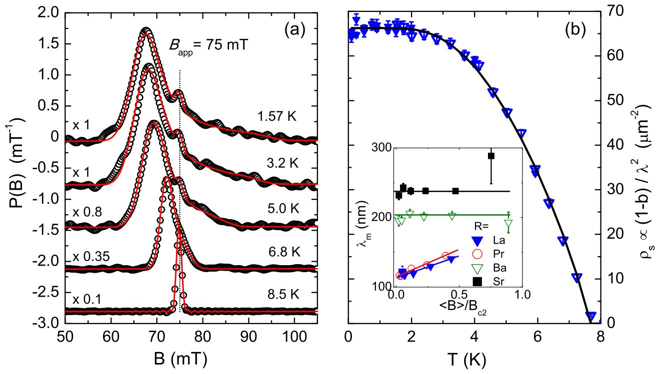

FIG. 1. (Color online) (a) Fourier transforms of the μSR time spectra $P(B)$ of $LaPt_4Ge_{12}$ at various temperatures, with an applied field of $B_{app} = 75$ mT. For reasons of clarity, the data at each temperature were scaled and shifted vertically. The solid lines are fits to the data. (b) Temperature dependence of the superfluid density $\rho_s^{\mu SR} \propto (1-b)/\lambda_m^2$. The solid line shows a BCS fit. Inset: Magnetic penetration depth λ_m as a function of normalized field $\langle B\rangle/B_{c2}$ in MPt_4Ge_{12}, M = La, Pr [11], Sr, and Ba [22].

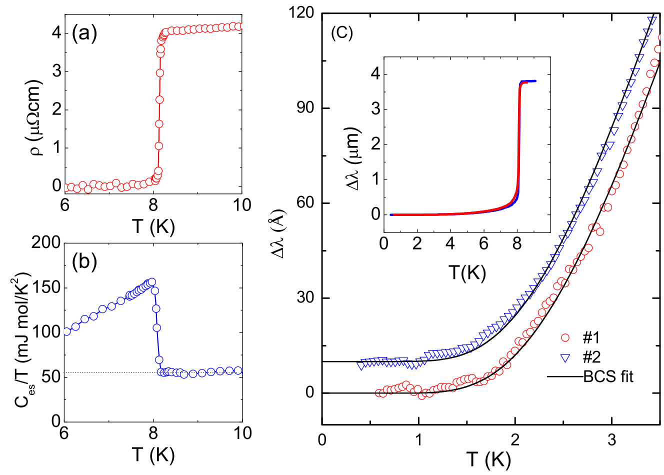

FIG. 2. (Color online) Temperature dependence of (a) the electrical resistivity $\rho(T)$, (b) the electronic specific heat, and (c) the London penetration depth $\Delta\lambda_L(T)$ for $LaPt_4Ge_{12}$ single crystals. The inset shows $\Delta\lambda_L(T)$ over a wide temperature range. For clarity, the $\Delta\lambda_L(T)$ data of sample No. 2 are shifted by 10 Å. The solid lines present a fit of the BCS model to the experimental $\Delta\lambda_L(T)$ at $T \ll T_c$.

The samples were cooled in $B_{app} = 75$ mT from above T_c down to 1.5 K, and then measured as a function of temperature. At $T = 1.5$ K, we also collected data in several applied magnetic fields. Typical counting statistics were 7–8 $\times 10^6$ positron events per data point. High quality single crystals of $LaPt_4Ge_{12}$, grown using multistep thermal treatments [15,18], were utilized for the London penetration depth study by a TDO-based method. The ac field induced by the coil of the experimental setup was less than 2 μT, which is much smaller than the lower critical field of $LaPt_4Ge_{12}$, guaranteeing that the sample was in the Meissner state. Typical sample dimensions were about $0.6 \times 0.6 \times 0.2$ mm^3. The change of the London penetration depth $\Delta\lambda_L(T)$ is proportional to the frequency shift, i.e., $\Delta\lambda_L(T) = G\,\Delta f(T)$, where G is solely determined by the sample and coil geometries [19].

Figure 1(a) shows the Fourier transforms of the μSR time spectra $P(B)$ of $LaPt_4Ge_{12}$ at various temperatures for $B_{app} =$ 75 mT. Above $T_c = 8.3$ K, a narrow and sharp peak is visible in $P(B)$, as is expected for weak nuclear depolarization. Upon lowering the temperature, the peak significantly broadens and becomes asymmetric, in accordance with the field distribution of a well arranged flux-line lattice (FLL). These features point to isotropic SC and weak pinning of the FLL in the polycrystalline samples. In this context, we adopt the exact solution of the Ginzburg-Landau equations following the method suggested by Brandt to analyze the data [20,21]. The complete analysis of the data follows the procedures described in Ref. [22]. Accordingly, we considered the spatial magnetic field distribution $B(\mathbf{r}) = B(\mathbf{r},\lambda_m,\xi,\langle B\rangle)$ within the unit cell of the FLL, where λ_m is the magnetic penetration depth and ξ is the coherence length. The data are well fitted using the theoretical polarization function $P(t)$ given in Ref. [22], as denoted by the solid lines in Fig. 1(a).

The mean value of the superfluid density can be expressed as $\rho_s^{\mu SR} \propto (1-b)/\lambda_m^2$, where $b = \langle B\rangle/B_{c2}(0)$ is the reduced field. In Fig. 1(b), we plot $\rho_s^{\mu SR}$ derived from the μSR data as a function of temperature, which can be fitted by the conventional s-wave BCS model with a gap amplitude of $\Delta(0) = 1.9 k_B T_c$. The field dependence of the penetration depth provides an alternative method for probing the low-energy excitations. For nodal or multigap superconductors, the penetration depth λ_m depends on the applied magnetic field [23], but there is hardly any variation for conventional s-wave superconductors [24]. In the inset of Fig. 1(b) we show the field dependence of λ_m obtained at 1.5 K for several Pt_4Ge_{12}-based superconducting skutterudites. In order to avoid correlation effects between λ and ξ, the values of the coherence length ξ were fixed to 15.0, 14.2, 19.7, and 27.0 nm for the La-, Pr-, Sr-, and $BaPt_4Ge_{12}$ skutterudites, respectively [25]. These values were obtained from the corresponding upper critical fields B_{c2} at 1.5 K using the Ginzburg-Landau relation $B_{c2} = \Phi_0/2\pi\xi^2$ [11,22]. For $LaPt_4Ge_{12}$ we used $B_{c2}(1.5\text{ K}) =$ 1.48 T [26]. λ_m of $SrPt_4Ge_{12}$ and $BaPt_4Ge_{12}$ are independent of the applied field, as expected for s-wave SC with an isotropic gap [see Fig. 1(b)]. In contrast, λ_m of both $LaPt_4Ge_{12}$ and $PrPt_4Ge_{12}$ displays a linear field dependence with a similar slope, which remarkably resembles that of MgB_2 [27], a prototype example of a two-band superconductor. Such behavior is compatible with multiband SC, as previously shown for $PrPt_4Ge_{12}$ [14–16], but is unexpected for a single-band BCS superconductor.

In order to understand this contradictory behavior, we measured the London penetration depth $\Delta\lambda_L(T)$ using the TDO-based method for single-crystalline $LaPt_4Ge_{12}$. A sharp superconducting transition at $T_c = 8.2$ K is observed in both the electrical resistivity $\rho(T)$ and specific heat $C_{es}(T)/T$, as shown in Figs. 2(a) and 2(b). Along with the residual resistivity ratio RRR = 16.3, this confirms the high quality of the $LaPt_4Ge_{12}$ crystals. Figure 2(c) presents $\Delta\lambda_L(T)$ for two samples from the same batch. The $\Delta\lambda_L(T)$ data are well reproducible between the two samples, showing nearly temperature-independent behavior below 1.5 K. This provides further evidence for nodeless SC in $LaPt_4Ge_{12}$. According

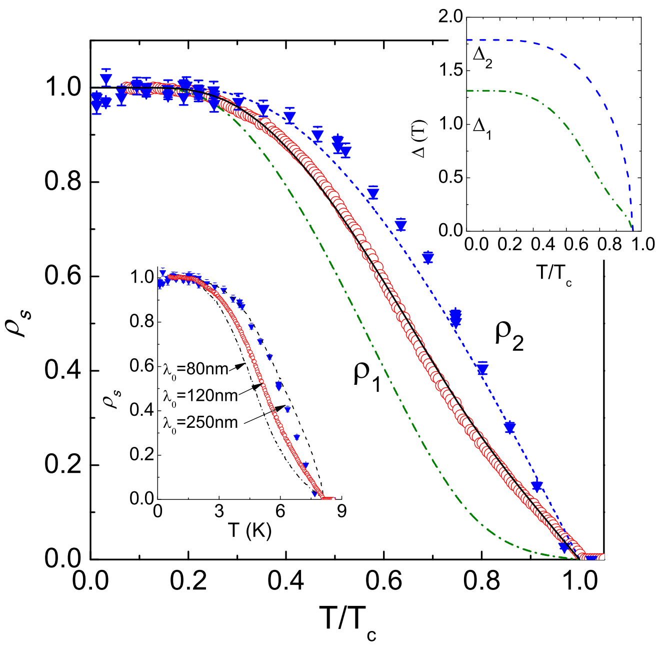

FIG. 3. (Color online) The normalized superfluid density $\rho_s(T)$ of $LaPt_4Ge_{12}$. The circles and triangles represent the experimental data obtained by using the TDO method (ρ_s^{TDO}) and the TF-μSR measurements ($\rho_s^{\mu\mathrm{SR}}$), respectively. The solid line is a fit based on the γ model; the derived partial superfluid densities $\rho_1(T)$ and $\rho_2(T)$ are shown by the dashed and dashed-dotted lines, respectively. Lower inset: The superfluid density $\rho_s^{\mathrm{TDO}}(T)$ calculated with $\lambda_0 = 80$, 120, and 250 nm. Upper inset: Temperature dependence of the gap amplitudes $\Delta_1(T)$ and $\Delta_2(T)$.

to the BCS model, the penetration depth at $T \ll T_c$ can be expressed by [28] $\Delta\lambda(T) \approx \lambda(0)\sqrt{\frac{\pi\Delta(0)}{2k_BT}}\exp\left(-\frac{\Delta(0)}{k_BT}\right)$, where $\lambda(0)$ is the penetration depth at $T = 0$. Here, we take the value of $\lambda(0) \approx 120$ nm from the μSR results. Usually, the parameter $\Delta(0)$ varies upon changing the fitted temperature range, but becomes saturated at sufficiently low temperatures. The solid lines in Fig. 2(c) represent the best fits to the BCS model in the low-temperature limit, giving $\Delta(0) = 1.39k_BT_c$ for sample No. 1 and $\Delta(0) = 1.34k_BT_c$ for sample No. 2. The fitted $\Delta(0)$ is much smaller than the BCS value of $\Delta(0) = 1.76k_BT_c$, indicating the appearance of a secondary gap at low temperatures or the anisotropic effect, which is unlikely in a cubic system.

In Fig. 3, we plot the normalized superfluid density $\rho_s^{\mathrm{TDO}}(T) = \lambda^2(0)/\lambda_L^2(T)$ for $LaPt_4Ge_{12}$, which shows an upward curvature near T_c. Such behavior may result from the underestimation of $\lambda(0)$. As a comparison, the lower inset of Fig. 3 shows $\rho_s^{\mathrm{TDO}}(T)$ calculated using different values of $\lambda(0)$. Indeed, the upturn of $\rho_s^{\mathrm{TDO}}(T)$ weakens with increasing $\lambda(0)$; $\rho_s^{\mathrm{TDO}}(T)$ resembles the μSR data for $\lambda(0) = 250$ nm, but still shows a clear deviation near T_c. It is noted that our samples have a regular geometry and the total uncertainties from the $\lambda(0)$ value and the G factor in the TDO measurements should be limited to about 20%. Thus, such a large enhancement of $\lambda(0)$ is also far beyond the uncertainties from our experiments. On the other hand, similar concave behavior was previously observed in some multiband superconductors with weak interband coupling [29]. Considering the fact that $LaPt_4Ge_{12}$ possesses multiple Fermi surface sheets [7], it is reasonable to analyze $\rho_s^{\mathrm{TDO}}(T)$ in terms of multiband SC.

According to the γ model [29], which applies generally to clean two-band superconductors, the total superfluid density can be written as $\rho_s(T) = \gamma\rho_1[\Delta_1(0),T] + (1-\gamma)\rho_2[\Delta_2(0),T]$, where γ is the relative weight of the contribution from the gap Δ_1. For a two-band system with known density of states n_μ and Fermi velocities υ_μ ($\mu = 1,2$), γ is determined by $\gamma = n_1\langle\upsilon_1^2\rangle/(n_1\langle\upsilon_1^2\rangle + n_2\langle\upsilon_2^2\rangle)$. Here, $\langle\upsilon_\mu^2\rangle$ represents the average of the squared Fermi velocities over the corresponding band. According to the electronic structure calculations [7], several energy bands cross the Fermi energy in $LaPt_4Ge_{12}$. As an approximation, we group the major energy bands into two effective bands according to the values of Fermi velocity in each band, which gives $n_1 = 0.12$, $n_2 = 0.88$, and $\upsilon_1/\upsilon_2 \sim 2.3$. Thus, a value of $\gamma = 0.42$ is estimated. The partial superfluid density ρ_μ and the energy gap Δ_μ are calculated self-consistently using the quasiclassic Eilenberger equations. Following the method described in Ref. [29], we fitted the experimental $\rho_s^{\mathrm{TDO}}(T)$ with fitting parameters of γ and $\lambda_{\mu\nu}$, where $\lambda_{\mu\nu}$ are the interband and intraband pairing couplings. The best-fit results, with $\lambda_{11} = 0.63$, $\lambda_{12} = 0.0025$, and $\lambda_{22} = 0.20$, are shown in Fig. 3, which overlaps nicely with the experimental data. The derived values of $n_1 = 0.23$ and $\gamma = 0.4$ are compatible with those estimated directly from the band structure calculations. Such a fit gives $\Delta_1(0) = 1.31k_BT_c$ and $\Delta_2(0) = 1.80k_BT_c$; the small gap $\Delta_1(0)$ agrees well with that obtained from the exponential fits of $\Delta\lambda_L(T)$ at $T \ll T_c$, where $\Delta(0) = 1.39k_BT_c$. As shown in the upper inset of Fig. 3, $\Delta_1(T)$ exhibits non-BCS behavior; the rapid vanishing of $\Delta_1(T)$ leads to the upturn in $\rho_s^{\mathrm{TDO}}(T)$ near T_c.

Remarkably, the calculated partial superfluid density $\rho_2(T)$ is nearly identical to $\rho_s^{\mu\mathrm{SR}}(T)$ (see Fig. 3). Such an agreement suggests that the small energy gap is likely destroyed by the small magnetic field applied in the μSR experiments. In a two-band superconductor with a vanishing λ_{12}, it was theoretically shown that the small gap may be suppressed at a largely reduced effective upper critical field [30]. The so-called "virtual" upper critical field B_{c2}^s, above which the vortex cores overlap and thus drive the majority of the electrons in the associated band normal, can be estimated by $B_{c2}^s(0) \sim B_{c2}(0)[\Delta_1(0)\upsilon_2/\Delta_2(0)\upsilon_1]^2$. For $LaPt_4Ge_{12}$, a value of $B_{c2}^s(0) \sim 150$ mT is estimated by using the parameters derived above and $B_{c2}(0) = 1.6$ T [26]. The small $B_{c2}^s(0)$ may account for the discrepancy between $\rho_s(T)$ determined from the TDO and μSR experiments. In the TDO measurements, the sample is in the Meissner state, which allows us to detect both the large and the small gaps. On the other hand, the transverse-field muon-spin rotation (TF-μSR) measurements were carried out in a field of 75 mT. In this case, the small gap Δ_1 is largely suppressed and $\rho_s^{\mu\mathrm{SR}}(T)$ is governed by the large gap, leading to a single-band BCS behavior with an energy gap Δ_2. Our analysis suggests that $LaPt_4Ge_{12}$ is a marginal two-band superconductor, the signatures of the small gap being easily extinguished by magnetic fields due to its weak interband coupling. It should be noted that the band structure of $LaPt_4Ge_{12}$ is more

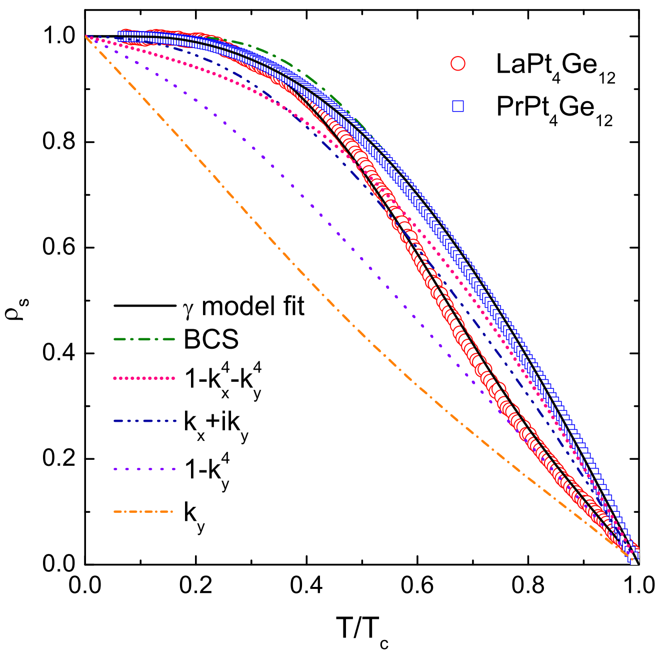

FIG. 4. (Color online) The normalized superfluid density $\rho_s^{\mathrm{TDO}}(T)$ for $LaPt_4Ge_{12}$ and $PrPt_4Ge_{12}$. The symbols and solid lines represent the experimental data and their corresponding fits using the two-band γ model. For comparison, we also fit $\rho_s^{\mathrm{TDO}}(T)$ of $LaPt_4Ge_{12}$, using various gap functions allowed by the crystal symmetry [15].

complex than the simplified scenario discussed above, which may explain the difference in the fields required to suppress the small gap in the measurements of field and temperature dependence of the penetration depth. On the other hand, the interband impurity scattering may mix the two bands, which may also lead to single-gap superconducting behavior [31].

It was previously shown that the sister compound, $PrPt_4Ge_{12}$, is also a two-gap superconductor with a relatively small admixture of a second gap with reduced Δ_0/k_BT_c [15]. As a comparison, we plotted $\rho_s^{\mathrm{TDO}}(T)$ of $LaPt_4Ge_{12}$ and $PrPt_4Ge_{12}$ in Fig. 4, which show different curvatures near T_c. In Fig. 4, we reanalyzed the ρ_s^{TDO} of $PrPt_4Ge_{12}$ in terms of the γ model. The best fit gives intraband coupling parameters of $\lambda_{11} = 0.59$ and $\lambda_{22} = 0.18$, which are compatible with those of $LaPt_4Ge_{12}$. On the other hand, the fitted interband coupling $\lambda_{12} = 0.07$ is over one order of magnitude larger than that of $LaPt_4Ge_4$. The enhanced interband coupling leads to a more robust small energy gap, showing the same type of temperature dependence for the two gaps [15]. For $PrPt_4Ge_{12}$, the gap parameters derived from the γ model [$\Delta_1(0) = 0.88k_BT_c$, $\Delta_2(0) = 2.17k_BT_c$, and $\gamma = 0.15$] are consistent with those fitted by the α model [15]. These results suggest that the Pr-4f electrons may not play a crucial role in the formation of SC, but enhance the interband coupling in the MPt_4Ge_{12} compounds. The latter factor might arise from the local crystal field excitations at the Pr sites in $PrPt_4Ge_{12}$. This finding is consistent with the local-density approximation calculations, which show similar electronic structures for the two compounds, where the main contribution of the density of states at the Fermi level is from the Pt and Ge states [7]. Following the same methods used in Ref. [15] for $PrPt_4Ge_{12}$, we also fit $\rho_s^{\mathrm{TDO}}(T)$ of $LaPt_4Ge_{12}$ with various gap functions (see Fig. 4). It can be seen that all the other models give a poor fit to the experimental data, further supporting the two-gap SC of $LaPt_4Ge_{12}$.

Our results may have broader implications for reconciling the diverse behaviors of several multiband superconductors, in particular, those with a weak interband coupling. A number of filled skutterudite superconductors have been considered to be two-band superconductors on the basis of various experiments [5,6,32], while in others the data are not convincing [33,34]. Other similar examples also include V_3Si, La_2C_3, and $MgCNi_3$, where different experimental probes are not yet conclusive on the order parameter symmetry [35]. Focusing on our results, we find that, in $LaPt_4Ge_{12}$, the smaller gap is subtle and could be easily suppressed by magnetic fields due to the weak interband coupling. To further examine this scenario, it is desirable to study the evolution of the gap structure by tuning the magnetic field or the impurity scattering in $LaPt_4Ge_{12}$ and other superconductors with weak interband coupling.

In summary, we have investigated the superconducting order parameter of $LaPt_4Ge_{12}$ by measuring the penetration depth and the superfluid density using two different methods. We found that $\Delta\lambda_L(T)$ and $\rho_s^{\mathrm{TDO}}(T)$ from the TDO measurements in the Meissner state can be consistently described by the two-band γ model with $\Delta_1(0) = 1.31k_BT_c$ and $\Delta_2(0) = 1.80k_BT_c$. The small gap is likely suppressed by the small magnetic field applied in the μSR experiments. Our results suggest that $LaPt_2Ge_{12}$ is a two-band superconductor where the critical field for suppressing the small gap is significantly reduced due to the extraordinarily weak interband coupling. The presence of 4f electrons in the Pr-based skutterudite superconductors may enhance the interband coupling and, therefore, give rise to more robust multiband behavior.

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