Probing the vortex state of $PrRu_4Sb_{12}$ through muon spin rotation and relaxation

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We have investigated the magnetic penetration depth λ and the superconducting coherence length ξ in the vortex lattice of the filled skutterudite compound $PrRu_4Sb_{12}$ ($T_c \sim 0.97$ K) using transverse-field muon-spin rotation measurements. Zero-field and longitudinal-field studies were also carried out to investigate the time-reversal symmetry of the superconducting state. We found $\lambda = 3650(20)$ Å and $\xi = 345(5)$ Å at 0.05 K and using these values of λ and ξ, together with the Sommerfeld constant we have calculated an effective mass of the quasiparticles $m^* \approx 10 m_e$ and superfluid carrier density $n_s \approx 4 \times 10^{27}$ carriers/m^3. The temperature dependence of the vortex state muon-spin relaxation rate $\sigma_s(T)$ is consistent with the phenomenological two-fluid model. Further, the zero-field and longitudinal field measurements do not reveal any signature of a spontaneous internal magnetic field below the superconducting transition temperature, indicating the preservation of time-reversal symmetry in the superconducting state of $PrRu_4Sb_{12}$, unlike the broken time-reversal symmetry of the superconducting state of the heavy-fermion superconductor $PrOs_4Sb_{12}$.

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I. INTRODUCTION

The filled skutterudite compounds of the type RT_4M_{12} (R=rare earth, T=Fe, Ru, and Os, and M=P, Sb, and As), which exhibit the simple body-centered cubic structure (space group Im-3), have recently attracted considerable interest because of the various properties exhibited by many members of this class, such as unconventional superconductivity, metal-insulator transitions, heavy Fermion behavior, quadrupolar ordering and hybridization gap semiconductivity.[1–5] In addition to the interest in their basic condensed matter physics, these compounds are excellent candidates for future solid state thermoelectric device technology.

Among the rare-earth filled skutterudites, the Pr-based compounds PrT_4M_{12} are of particular interest as Pr^{3+} ions can have a magnetic or nonmagnetic ground state.[6–8] The latter can be either a nonmagnetic singlet (Γ_1) or a nonmagnetic doublet (Γ_3) with a finite quadrupole moment (we will use the more familiar group theory labels for O_h point group symmetry; see Ref. 11 to see how they map onto T_h groups). Interestingly, superconductivity has been observed in the La and Pr-based filled skutterudite compounds with the highest T_c of 10.3 K for $LaRu_4As_{12}$ and 2.4 K for $PrRu_4As_{12}$.[7] Superconductivity is also observed in the ruthenium antimonide skutterudite compounds with T_c=2.8 K for $LaRu_4Sb_{12}$ and 1 K of $PrRu_4Sb_{12}$.[3,7] It is interesting to note that $PrOs_4Sb_{12}$ becomes a superconductor at 1.85 K, which is higher than that of its nonmagnetic analog $LaOs_4Sb_{12}$ (T_c=0.74 K). This unusual feature has been attributed to the effects of quadrupolar fluctuations on the pairing mechanism in $PrOs_4Sb_{12}$.[9–11] Naturally the crystal field level scheme of the Pr^{3+} f electrons imposes important restrictions on possible theoretical models for the superconductivity.

Recently, we have investigated the crystal field potential in $PrOs_4Sb_{12}$ and $PrRu_4Sb_{12}$ using inelastic neutron scattering, showing that the ground state of the split $^3H_4$ multiplet is the Γ_1 singlet in both the compounds.[11,12] This eliminates any quadrupolar fluctuations within the ground state level, which is required by the quadrupolar Kondo effect.[13] On the other hand, the first excited triplet (Γ_5) state is located at 0.6 meV for $PrOs_4Sb_{12}$ and 5.6 meV for $PrRu_4Sb_{12}$ above the ground state. It was argued that inelastic quadrupolar fluctuations, or aspherical Coulomb scattering by transitions to the first excited triplet state, are responsible for the enhancement of T_c of $PrOs_4Sb_{12}$ compared with that of $LaOs_4Sb_{12}$.[11,12]

Despite having the same crystal field ground state in $PrOs_4Sb_{12}$ and $PrRu_4Sb_{12}$, various experimental investigations suggest that these two superconducting compounds have different order-parameter symmetry. The heat capacity measurements reveal heavy-fermion nature with $\gamma = 350$ mJ/mol K^2 for $PrOs_4Sb_{12}$, while a marginal heavy-fermion behavior with γ=59 mJ/mol K^2 is found for $PrRu_4Sb_{12}$.[1,3] The Sb-NQR spin-lattice measurements $1/T_1$ results reveal a distinct coherence peak just below T_c in $PrRu_4Sb_{12}$,[14] but no such peak has been observed in $PrOs_4Sb_{12}$.[15] A muon spin relaxation study on $PrOs_4Sb_{12}$ reveals the spontaneous appearance of static internal magnetic fields below T_c indicating that the superconducting state breaks time-reversal-symmetry.[16] The transverse field muon-spin rotation of $PrOs_4Sb_{12}$ reveals an isotropic or nearly isotropic energy gap with magnetic penetration depth $\lambda(0) = 3440(20)$ Å.[17]

It is therefore interesting to see whether or not the microscopic nature of the superconducting state in $PrRu_4Sb_{12}$ is similar to that of $PrOs_4Sb_{12}$. The muon-spin rotation (μ^+SR) technique provides sensitive measurements of the microscopic field distribution inside a type-II superconductor,[18,19] e.g., the magnetic penetration depth λ that is one of the fundamental length scales of superconductors. It can be further used to determine the symmetry of the superconducting order parameter. Although the penetration depth of $Pr(Os_{1-x}Ru_x)_4Sb_{12}$ $(x=0-1)$ was previously measured through tunnel diode oscillator measurements, this is a less accurate method to estimate reliable absolute values of the penetration depth because of the limitations of the technique.[20,21] In this paper, we have therefore carried out μSR measurements on $PrRu_4Sb_{12}$ in zero-field (ZF), longitudinal-field (LF), and transverse-field (TF) configurations to characterize the properties of the superconducting state of $PrRu_4Sb_{12}$.

II. EXPERIMENTS

Our polycrystalline sample of $PrRu_4Sb_{12}$ was prepared by the flux growth technique at the University of Tokyo as described in Ref. 3. The phase purity of the sample was checked by powder neutron diffraction measurements using the high resolution powder diffractometer (HRPD) at the ISIS Pulsed Neutron and Muon Facility, UK. The positive muon spin-relaxation (μ^+SR) measurements were performed using the MUSR spectrometer at ISIS. We have carried out ZF, LF, and TF μSR measurements at temperatures ranging from 30 mK to 1.5 K. The sample was fixed to a silver backing plate and mounted onto the cold finger of a dilution refrigerator. The silver sample holder was used because it gives a nonrelaxing muon signal and hence only contributes a temperature independent constant background. In the TF measurements, the field was applied at 1.5 K and then the sample was cooled down to 0.05 K at constant applied magnetic field to avoid any effect due to flux trapping. To analyze the TF data we have projected all the data from 32 detectors onto two histograms with orthogonal phases of 0 and $\pi/2$, using the method outlined in Ref. 22. We refer to these histograms as the "real" and "imaginary" components of the signal.

III. RESULTS AND DISCUSSION

A. Neutron diffraction

Rietveld analysis of our neutron-diffraction data using GSAS confirmed that $PrRu_4Sb_{12}$ crystallises in the *Im*-3 space group and that the sample was nearly single-phase with ~6.7 and ~0.6 % of impurity phases of $RuSb_2$ and pure Sb metal, respectively (see Fig. 1), both of which are nonmagnetic. The estimated value of the cubic lattice parameter $a=9.27660(1)$ Å and the atomic position parameters of Sb atoms are $x_{Sb}=0$, $y_{sb}=0.15754(7)$, and $z_{sb}=0.34140(6)$. Occupancy factors and isotropic thermal factors were also refined. The values of the occupancy factors are 1.030(10) for Pr and 0.981(2) for Sb (the occupancy 1.000 for Ru was kept fixed during the Rietveld refinement), which corresponds to essentially full occupancy of all the atomic sites, and the values of isotropic thermal factors (in Å^2) are 0.0420(16) for Pr, 0.0097(3) for Ru, and 0.0062(6) for Sb. The larger value of the isotropic temperature factor observed for the Pr atom is consistent with the rattling motion expected for the Pr atoms in this structure.

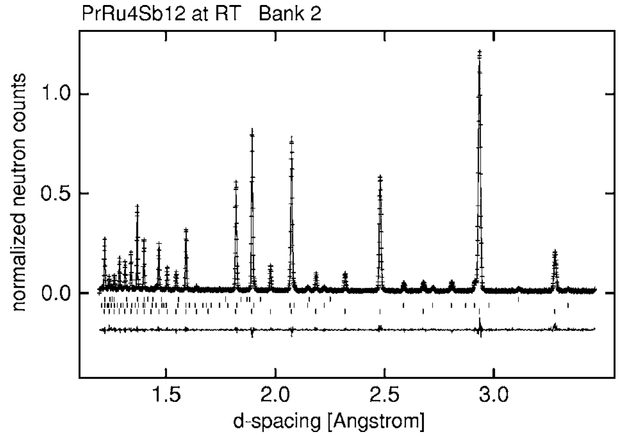

FIG. 1. Powder neutron diffraction pattern from $PrRu_4Sb_{12}$ taken at 300 K. Symbols are the data points while the line underneath them is the refined profile. Three rows of vertical bars correspond to the phases of $PrRu_4Sb_{12}$ (bottom), $RuSb_2$ (middle), and pure Sb-metal (top). The line at the bottom represents the difference curve.

B. Muon spin rotation and relaxation

Figure 2 shows the zero-field and longitudinal-field (50 G)μSR spectra at 0.03 and 1.5 K (i.e., below and above T_c) from $PrRu_4Sb_{12}$. The field of 50 G was applied after cooling the sample in ZF to 0.03 K. The most important result is that both the ZF and LF muon depolarization rates show no temperature dependence, indicating that there is no spontaneous internal magnetic field, and confirming that time-reversal symmetry is preserved in the superconducting state of $PrRu_4Sb_{12}$. This is in contrast to $PrOs_4Sb_{12}$, in which

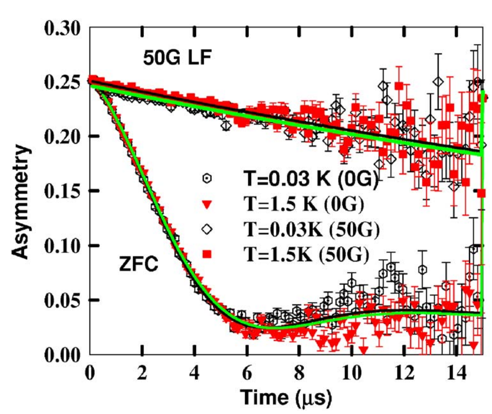

FIG. 2. (Color online) Zero-field and 50 G longitudinal field μSR spectra below and above the superconducting transition temperature (T_c=0.97 K). The solid lines represent the fit to the function described in the text.

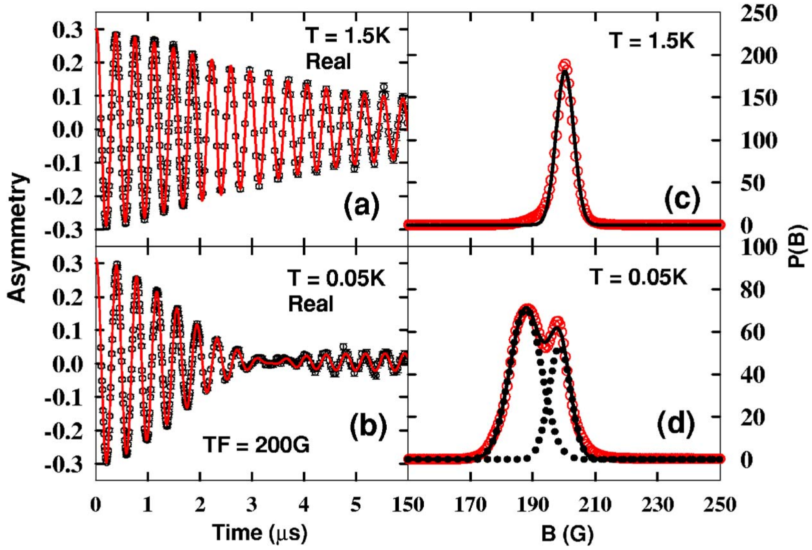

FIG. 3. (Color online) (a),(b) Transverse-field (TF) μSR spin precession signals from $PrRu_4Sb_{12}$ obtained under an applied field of 200 G. Here we have shown only the real component: (a) in the normal state at 1.5 K and (b) in the superconducting state at 0.05 K. The weakly nonrelaxing signal at longer time seen in (b) is due to muons stopping on the sample holder and nonsuperconducting impurities. The solid line represents the fit (see the text). (c),(d) Probability field distribution $p(B)$ of the internal magnetic field obtained using the maximum entropy method in $PrRu_4Sb_{12}$ at 1.5 and 0.05 K. The sharp peak on the right-hand side at 0.05 K is due to the background signal. The solid line represents a Gaussian fit to the data and the dotted line shows the components of the fit.

the ZF μSR study clearly indicates a spontaneous static internal magnetic field below T_c, which has been interpreted as broken time-reversal symmetry of the superconducting state.[16] When the time-reversal symmetry is broken, the nonzero magnetic moments of Cooper pairs that align locally are responsible for the spontaneous, but extremely small, internal magnetic field below T_c.[16] Further, we also measured μSR spectra of $PrRu_4Sb_{12}$ in a 50 G field cooled (FC) state. These results are indistinguishable from the zero-field cooled data, indicating that the paramagnetic ground state persists down to 0.03 K.

Our ZF field μSR spectra were fitted using a product of Gaussian Kubo-Toyabe and Lorentzian components. The former is attributed to muon-spin relaxation due to the randomly oriented local fields caused by nuclear moments and the latter to muon-spin relaxation due to electronic fluctuations. For the 50 G LF data we used only the Lorentzian component. In addition to this, a constant background component A_{bg} was also added to account for muons stopping on the sample holder and for possible impurity contributions. The form of the relaxation function used in our analysis is

$$A_{ZF}(t) = A_0\left\{\frac{1}{3} + \frac{2}{3}\left[1 - (\sigma_{KT}t)^2 \exp\left(-\frac{1}{2}(\sigma_{KT}t)^2\right)\right]\right\} \times \exp(-\Lambda t) + A_{bg}. \tag{1}$$

The values of the parameters obtained by simultaneous fitting of the four spectra at 1.5 K {0.03 K} shown in Fig. 2 are Gaussian Kubo-Toyabe relaxation rate σ_{KT}=0.245(3) μs^{-1} {0.256(2)}, electronic damping Λ=0.106(4) μs^{-1} {0.100(4)} in zero-field, and Λ=0.022(1) μs^{-1} {0.022(1)} in 50 G longitudinal field. The estimated initial asymmetry is $A_0 \sim 0.232(1)$ and the constant background A_{bg}=0.020(1). It is interesting to note that the ZF values of σ_{KT} and Λ estimated for $PrRu_4Sb_{12}$ are very similar to those estimated for $PrOs_4Sb_{12}$ (above T_c), σ_{KT}=0.19 μs^{-1} and Λ=0.13 μs^{-1}.[17] The value of A_{bg} gives a background contribution of 8.62%, which is close to the impurity percentage of 6.7% estimated from our neutron diffraction analysis. It is to be noted that there will be a contribution to the relaxation rate (Λ) coming from the dynamic behavior of the Pr 4f electrons. However, this contribution is very small and temperature independent in the temperature range of the investigation, 0.03 and 1.5 K, as the crystal field ground state of the Pr^{3+} ions in $PrRu_4Sb_{12}$ as well as in $PrOs_4Sb_{12}$ is a singlet.[11,12]

In order to determine the magnetic penetration depth λ and the superconducting coherence length ξ we have carried out TF-μSR measurements at various applied magnetic fields between 0 and 700 G, at 0.05 and 1.5 K. Figures 3(a) and 3(b) show the TF-μSR muon-spin precession signals, the real component (the imaginary component has not been shown here), at an applied field of 200 G above and below the superconducting transition temperature of $PrRu_4Sb_{12}$. A clear difference in the relaxation rate above and below T_c can be seen from the data in Figs. 3(a) and 3(b). This is due to the well-known fact that type-II superconductors exhibit a flux-line lattice (FLL) state at an applied field $B_{c1}<B<B_{c2}$ (below T_c) leading to spatial inhomogeneity of the magnetic induction. Since the implanted muons experience random precessions over the length scale of the FLL, the observed precession signal is a spatial sum of the internal fields. In a polycrystalline sample the Gaussian field distribution of local fields is to a good approximation[18]

$$\sigma = \gamma_\mu \sqrt{\langle(\Delta B^2)\rangle}, \tag{2}$$

where $\langle(\Delta B^2)\rangle$ is the average of the second moment of the field distribution ($\langle \Delta B^2\rangle = \langle [B(r) - B_0]^2\rangle$), with B_0 the applied field, and $\gamma_\mu/(2\pi)=135.53(\mathrm{MHz}/T)$ is the gyromagnetic ratio of the muon. To estimate $\langle(\Delta B^2)\rangle$ it is a common practice to assume a Gaussian distribution of local fields, where the time dependence of the muon spin polarization is proportional to $\exp[-(\sigma_s t)^2/2]$, and σ_s is the muon depolarization rate.[23] In order to check that the internal field distribution in our sample has a Gaussian line shape, we have used the maximum-entropy method to estimate the probability distribution of the internal magnetic field at 1.5 and 0.05 K [Figs. 3(c) and 3(d)].[24] Above T_c the line shape is Gaussian (the solid line shows the fitted results) and centered at the applied field, due to the uniform field penetration in the sample. Below T_c, two Gaussian components are observed due to the inhomogeneous field distribution in the superconducting FLL state, which has a large second moment $\langle(\Delta B^2)\rangle$, and in the nonsuperconducting background part. This increase in $\langle(\Delta B^2)\rangle$, in turn, gives rise to the large value of the muon-spin depolarization rate below T_c and is related to the magnetic penetration depth. For a triangular lattice[18,20]

$$\langle(\Delta B^2)\rangle = \left(0.00371\frac{\phi_0^2}{\lambda^4}\right), \tag{3}$$

where $\phi_0=2.07\times10^{-15}$ T m^2 is the flux quantum. As with other phenomenological parameters characterizing a superconducting state, the penetration depth can also be related to quantities at the atomic level. Using London theory[18]

$$\lambda^2 = \frac{m^* c^2}{4\pi n_s e^2}, \tag{4}$$

where m^* is the effective mass and n_s is the density of superconducting carriers. Within this simple picture, λ is independent of magnetic field.

The analysis of our TF-μSR data was carried out in the time domain using the following functional form:

$$A_{\mathrm{TF}}(t) = A_0 \exp\left(-\frac{1}{2}(\sigma_s t)^2\right)\exp\left(-\frac{1}{2}(\sigma_n t)^2\right) \times \exp(-\Lambda_{\mathrm{TF}} t)\cos(\omega t+\Phi_1) + A_{\mathrm{bg}}\cos(\omega_{\mathrm{bg}} t+\Phi_2), \tag{5}$$

where the first term is the contribution from the sample and the second term the background contribution. A_0 and A_{bg} are the initial asymmetries arising from the sample and background, respectively, while σ_n and σ_s are the muon-spin relaxation rates in the normal state and superconducting state, respectively. Λ_{TF} is the relaxation rate due to electronic moments. ω and ω_{bg} are the muon precession frequencies in the sample and background component with phase angle Φ_1 and Φ_2, respectively. It is to be noted that the TF-μSR data of $PrOs_4Sb_{12}$ were fitted using only a Gaussian component (i.e., with $\Lambda_{\mathrm{TF}}=0$).[17]

The analysis of our TF-μSR data of $PrRu_4Sb_{12}$ was carried out as follows. The ratio between $A_{\mathrm{bg}}/A_0=0.087$, estimated from the ZF μSR data, was kept fixed to reduce the number of variable parameters. First the real and imaginary components of the data at 1.5 K (i.e., above T_c), were fitted simultaneously to estimate A_0, σ_n, Λ_{TF}, ω, and ω_{bg} while keeping $\sigma_s=0$, and then these values of A_0, σ_n, and Λ_{TF} were kept fixed to estimate σ_s below T_c. The field dependence of the muon-spin relaxation rate (σ_s) obtained at 0.05 K in the FLL state, which is shown in Fig. 4, is nonlinear. A similar field dependence has also been observed in $PrOs_4Sb_{12}$,[17] YB_6,[25] and KOs_2O_6.[26] It was found that at 1.5 K $\sigma_n(B)$ exhibits an almost field independent behavior (these values were kept fixed at 0.05 K), but Λ_{TF} increases linearly with the applied magnetic field at a rate $\sim 2\times10^{-4}$ μs^{-1}/G. At 1.5 K the estimated value of $\Lambda_{\mathrm{TF}}=0.032(6)$ at 100 G and 0.121(9) at 600 G. A similar linear increase of σ_n (with Λ_{TF} assumed to be zero) with applied field was also observed for $PrOs_4Sb_{12}$.[17]

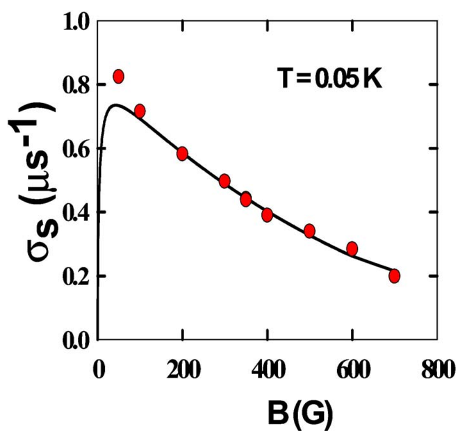

FIG. 4. (Color online) Field dependence of the muon spin relaxation rate associated with the flux line lattice σ_s at 0.05 K from $PrRu_4Sb_{12}$. The solid line represents the fit using the modified London model of Eq. (8).

We recall that the critical fields are directly related to the fundamental superconducting parameters, the penetration depth λ and the coherence length ξ, by the following relations:

$$B_{c2}(T) = (\phi_0/2\pi)1/\xi^2(T) \tag{6}$$

and

$$B_{c1}(T) = (\phi_0/4\pi)1/\lambda^2(T)\ln(\kappa), \tag{7}$$

where $\kappa=\lambda/\xi$. For an applied field $B_{\mathrm{app}}<B_{c2}/4$ where the simple London picture is valid, one can estimate λ from Eq. (3). This method has been used for $PrOs_4Sb_{12}$ ($B_{c2}\sim 23$ kOe), KOs_2O_6 ($B_{c2}\sim 400$ kOe), and high-temperature cuprate superconductors ($B_{c2}\sim 600$ kOe).[17,18,26] Since the upper critical field $B_{c2}=2$ kOe of $PrRu_4Sb_{12}$ is significantly smaller, we cannot apply the simple London picture and relate the muon depolarization rate to λ as given by Eq. (3). It is therefore necessary to use the field dependence of the muon-depolarization rate to determine λ using the modified London model[27]

$$\sigma_s(B) = \left(\frac{\gamma_\mu}{\sqrt{2}}\right)B_0\sum_{h,k}\left[\frac{\exp(-\xi^2 q_{h,k}^2)}{[1+q_{h,k}^2\lambda^2/(1-B_0/B_{c2})]^2}\right]^{1/2}$$

with

$$q_{h,k} \neq 0, \tag{8}$$

where B_0 is an applied magnetic field and $q_{h,k}$ the lattice sum over the FLL. We have fitted the estimated values of $\sigma_s(B)$ at 0.05 K to Eq. (8) with λ and ξ as free parameters. In our analysis, we have taken the lattice sum $q_{h,k}$ over the hexagonal flux line lattice, which is a reasonable assumption as the measured flux line lattice, using small angle neutron scattering (SANS), in $PrOs_4Sb_{14}$ exhibits a distorted hexagonal shape at low temperature.[28] The best fit to the data gave $\lambda = 3650(20)$ Å and $\xi = 345(5)$ Å for $PrRu_4Sb_{12}$ and the quality of the fit can be seen in Fig. 4. For comparison, the simpler picture expressed by Eq. (3), yields a value of $\lambda = 3610(16)$ Å at 0.05 K.

It is interesting to compare the present value of λ and ξ with the values estimated using other methods and also with those of $PrOs_4Sb_{12}$. For $PrRu_4Sb_{12}$, the reported value of $\lambda = 3200$ Å using the relation $\lambda = [\phi_0 B_{c2}(0)]^{1/2} / [\sqrt{24}(\Delta(0)/k_B T_C)\gamma^{1/2}]$ and $\xi = 400$ Å using the relation $\xi^2 = [\phi_0 / 2\pi B_{c2}(0)]$.[20] The tunnel diode oscillator study gives $\lambda = 2900$ Å,[20] which is smaller than the above values. As mentioned previously, this may be due to the fact that the tunnel diode oscillator value depends on a calibration constant. The value of λ and ξ obtained from μSR measurements is more reliable than that obtained from the bulk measurements, as μSR is a microscopic probe, which directly probes the flux-line lattice in the superconducting state, whose signal represents a simple average over different regions of the sample. Therefore, the effect of inhomogeneity is less serious in μSR than in bulk measurements. We therefore believe that the observed differences in the value of λ are not due to the sample quality, but most likely due to the accuracy of the methods used for the estimation. Furthermore, one obtains $\lambda(0) = 3440$ Å from μSR measurements and $\xi = 116$ Å from B_{c2} for $PrOs_4Sb_{12}$.[1,17]

Using our estimated value of $\lambda \propto (m^*/n_s)^{1/2}$ and the heat capacity coefficient $\lambda \propto m^* n_s^{1/3}$ from Ref. 3, along with the assumption that roughly all the normal state carriers (n_e) contribute to the superconductivity (i.e., $n_s \sim n_e$), we have estimated the values of $m^* \approx 10 m_e$ and $n_s \approx 4 \times 10^{27}$ carriers/m^3. This indicates that $PrRu_4Sb_{12}$ is a good metal, in agreement with the observed metallic resistivity behavior.[3] For comparison, the values for $PrOs_4Sb_{12}$ are $m^* \approx 50 m_e$ and $n_s \approx 10^{28}$ carriers/m^3.[17] The mean free path l estimated from the dHvA study is 1300 Å for $PrRu_4Sb_{12}$ (Ref. 29) which implies that the sample is in the clean limit, i.e., $(l/\xi \approx 3.8) > 1$, whereas $l/\xi \approx 3$ for $PrOs_4Sb_{12}$, which validates the thermodynamic relation used above to estimate the atomic parameters.

We also measured the temperature dependence of the FLL relaxation rate $\sigma_s(T)$ of $PrRu_4Sb_{12}$. Figure 5 shows the temperature dependence of $\sigma_s(T)$ in an applied magnetic field of 350 G. We have fitted the $\sigma_s(T)$ data to the phenomenological two-fluid model,[30] $\sigma_s(T) = \sigma_s(0)[1 - (T/T_c)^N]$, with $\sigma_s(0)$, T_c and N as variable parameters. The best fit was obtained with $T_c = 0.973(2)$ K, $\sigma_s(0) = 0.452(1)$ μs^{-1}, and $N = 1.44(1)$ (solid line in Fig. 5). The estimated value of T_c is in agreement with $T_c = 1.04$ K (in zero field) reported in Ref. 3. It is worth pointing out that there is no discrepancy between T_c estimated in the present work and in Ref. 3. We determined T_c in our sample from the temperature dependence of the penetration depth, which was measured in a field of 350 G. According to Fig. 2 in Ref. 3, this field is sufficient to suppress T_c by the required amount. The value of N obtained for $PrRu_4Sb_{12}$ is much smaller than $N = 4$, which is expected for s-wave isotropic gap superconductors (the dotted line in Fig. 5 shows the plot with $N = 4$), and $N = 3.6$ observed for $PrOs_4Sb_{12}$. A smaller value of $N = 2.39$ was also observed in KOs_2O_6, which has $T_c = 8.91$ K.[26] It is to be noted that the muon depolarization rate σ_s does not depend only on λ but also depends on ξ. A low value of $\kappa(\lambda/\xi)$ results in the overlapping of the flux lines in the FLL. This, clearly, leads to a reduced measured value of σ_s. Therefore the temperature dependence of σ_s has been modelled using the modified London model [Eq. (8)] with the temperature dependence form of λ and ξ predicted on the basis of the two-fluid model. This provides a correction for the overlapping flux lines in the FLL. The data are extremely well described for $T < 0.6$ K, and the fit is shown by a dashed-dotted line in Fig. 5. However, for $T > 0.7$ K the modified London model is no longer an adequate description of the flux line lattice, due to a strongly increasing overlap of vortices as we approach T_c. We note that in the clean limit the size of the vortex core region increases more rapidly with increasing temperature than in the dirty limit.[18] As $PrRu_4Sb_{12}$ is in the clean limit, one would expect strong overlap of the vortices as we approach T_c and hence the modified London model is not adequate near T_c.

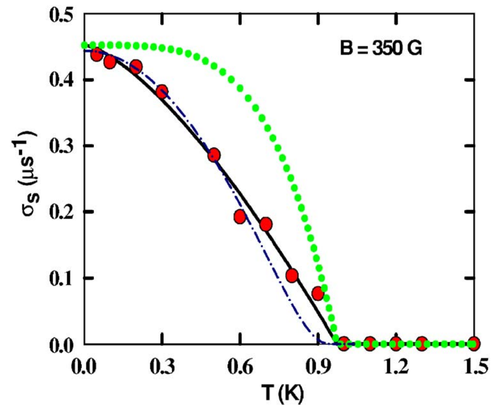

FIG. 5. (Color online) Temperature dependence of the muon spin relaxation rate associated with the flux line lattice σ_s in an applied field of 350 G from $PrRu_4Sb_{12}$. The solid line represents the fit based on the prediction of the phenomenological two-fluid model with $N = 1.44$ and the dotted line is for $N = 4$ as expected for an isotropic gap s-wave BCS superconductor. The dashed-dotted line represents the fit based on the modified London model, Eq. (8) (see the text).

IV. CONCLUSIONS

We have carried out μSR measurements to characterize the microscopic nature of the superconducting state of $PrRu_4Sb_{12}$. Our zero-field and longitudinal-field μSR studies reveal that the superconducting state preserves time-reversal symmetry in $PrRu_4Sb_{12}$, in contrast to the broken time-reversal symmetry reported for the heavy-fermion superconductor PrO_4Sb_{12}. However, it needs to be pointed out that the appearance of a static magnetic field below 2 K in $PrOs_4Sb_{12}$ (Ref. 16) could also be due to a rearrangement of the low lying CF levels induced by the implanted positive muon (μ^+) as observed in the singlet ground state compound $PrNi_5$.[31] In rare-earth systems with a non-magnetic ground state, the exchange interaction (J_{ex}) between f electrons competes with the CF splitting (Δ_{CF}). If $J_{ex}/\Delta_{CF}<1$ the system will remain paramagnetic (Van Vleck paramagnet) but if it is larger than one, the system orders magnetically. $PrOs_4Sb_{12}$ is a Van Vleck paramagnet with small value of $\Delta_{CF}=0.6$ meV and therefore it is quite possible that the implanted μ^+ will lift the degeneracy of the excited triplet and rearrange the low lying CF levels in such way that the ratio J_{ex}/Δ_{CF} becomes greater than one, creating local magnetic order. In the case of $PrRu_4Sb_{12}$, Δ_{CF} is an order of magnitude larger so the effect of the implanted μ^+ on the CF level scheme will be much weaker.

From the transverse-field μSR measurements, we estimated the magnetic penetration depth $\lambda=3650(20)$ Å and coherence length $\xi=345(5)$ Å at 0.05 K and derived an effective quasiparticle mass $m^*\approx 10m_e$ and superfluid carrier density $n_s\approx 4\times 10^{27}$ carriers/m^3. The temperature dependence of the vortex state muon-spin relaxation rate, $\sigma_s(T)$ is consistent with the phenomenological two-fluid model, although the observed value of the exponent 1.44(1) is markedly smaller than the value of 4 expected for an isotropic gap s-wave superconductor. Furthermore, the observed temperature dependent behavior of $\sigma_s(T)$ agrees very well (below 0.6 K) with the modified London model. We have compared the values of λ and ξ estimated from the μSR measurements with that estimated using bulk measurements and also with the value for the heavy-fermion superconductor $PrOs_4Sb_{12}$.

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