

Muon spin rotation measurements of the superfluid density in fresh and aged superconducting PuCoGa₅

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(Received 27 March 2007; published 6 August 2007)

We have measured the temperature dependence and magnitude of the superfluid density $\rho_s(T)$ via the magnetic field penetration depth $\lambda(T)$ in PuCoGa₅ (nominal critical temperature $T_{c0}=18.5$ K) using the muon spin rotation technique in order to investigate the symmetry of the order parameter, and to study the effects of aging on the superconducting properties of a radioactive material. The same single crystals were measured after 25 days ($T_c=18.25$ K) and 400 days ($T_c=15.0$ K) of aging at room temperature. Penetration depths $\lambda(0)=265(5)$ and $\geq 498(10)$ nm are derived for the fresh and aged samples, respectively. The temperature dependence of the superfluid density $\rho_s(T)/\rho_s(0)=\lambda(0)^2/\lambda(T)^2$ is well described in both materials by a model using d -wave gap symmetry. Within the context of a strong-coupling dirty d -wave model a zero-temperature gap value $\Delta_0=3.0k_B T_{c0}$ is obtained in the fresh sample for a scattering rate $\Gamma=0.005\pi k_B T_{c0}$, which is consistent with Abrikosov-Gor'kov (AG) pair-breaking theory. This Δ_0 should be compared to the weak-coupling value for a clean d -wave gap $\Delta_0=2.14k_B T_{c0}$. In the aged sample the same model yields $\Delta_0=2.4k_B T_{c0}$ for $\Gamma=0.010\pi k_B T_{c0}$. This value of Γ is much less than required by the AG pair-breaking formalism. Furthermore, the aged $\rho_s(0)$ is reduced by at least 70% compared to the fresh sample, which is also incompatible with $\Delta T_c/T_{c0}\sim 20\%$, according to AG theory. We conclude that the data in aged PuCoGa₅ support the postulate that the scattering from radiation-induced defects is not in the limit of the AG theory of an order parameter which is spatially averaged over impurity sites, but rather in the limit of short-coherence-length superconductivity. We show that a model calculation consistent with this assumption fits $\rho_s(T)$ in the aged sample rather well.

DOI: 10.1103/PhysRevB.76.064504

PACS number(s): 71.27.+a, 74.70.Tx, 61.80.-x, 76.75.+i

I. INTRODUCTION

Interest in the magnetism and superconductivity of f -electron materials has remained strong over the last several decades, starting with the discovery¹ of heavy fermion superconductivity in Ce- and U-based materials, and continuing today with the investigation² of ground states in the tetragonal Ce-based CeTIn₅ ($T=\text{Co, Ir, Rh}$) materials. Pu-based compounds exhibit a particular richness and complexity, as is illustrated by the discovery³ of superconductivity in PuCoGa₅ at an order of magnitude higher transition temperature ($T_{c0}=18.5$ K) than the structurally similar heavy fermion superconductor² CeCoIn₅ ($T_c\sim 2.3$ K). One reason for this complexity in Pu materials is that their f electrons sit at the boundary between more fully localized (as in Am compounds) and more fully itinerant (as in U and Np compounds) behavior.⁴

There is evidence that the superconductivity in PuCoGa₅ may be unconventional in nature. Recent nuclear magnetic resonance (NMR) and nuclear quadrupole resonance (NQR) studies⁵ display an absence of a coherence peak, together with a decreasing Knight shift and a T^3 behavior in the spin-lattice relaxation rate $1/T_1$ just below T_c , suggesting that the superconducting order parameter has d -wave symmetry, as in the copper-oxide superconductors.⁶ Calculations⁷⁻⁹ of the electronic structure of PuCoGa₅ show a Fermi surface con-

sisting of several cylindrical sheets, which is favorable for d -wave, spin-fluctuation mediated superconductivity. It has been suggested⁵ that PuCoGa₅ belongs to a class of spin-fluctuation induced superconductors, lying between the high-temperature, copper-oxide superconductors and the low-temperature, heavy-fermion superconductors on a plot of transition temperature versus spin fluctuation temperature.^{5,10}

Additional measurements which test this supposition are, therefore, important. This is particularly true because the natural radioactivity of Pu (²³⁹Pu half-life=24 000 years) creates lattice defects which scatter electrons and can create impurity bands which partly obscure the signature of a pure d -wave superconductor. Such effects are evident in the low-temperature NQR spin-lattice relaxation rate $1/T_1$,⁵ where the T^3 temperature dependence gives way to a linear- T behavior below about $0.4T_c$. As we show, the temperature dependence of the penetration depth $\lambda(T)$ in PuCoGa₅ is less sensitive to these defects, thus providing a clean test of the gap symmetry and of the robustness of the order parameter to the presence of pair-breaking defects. The magnitude of $\lambda(0)$ is strongly sensitive to defects, however, a fact which we are able to explain in terms of the relatively short superconducting coherence length in PuCoGa₅. Finally, the study of radiation effects in superconductors is of practical importance as well, because increases in the critical current density can be achieved by the pinning of flux lines at defects.

In this paper we present transverse-field (TF) muon spin rotation (μ SR) measurements of the in-plane magnetic field penetration depth λ in the same single crystals of PuCoGa_5 after 25 days and 400 days of aging at room temperature. $\lambda(T)$ is determined by the spectrum of quasiparticle excitations exceeding the superconducting energy gap $\Delta(T)$, and is thus a sensitive measure of the temperature dependence of the superfluid density $\rho_s(T) \propto \lambda(T)^{-2}$, and the gap structure. Some of the data after 25 days of aging has been reported previously.¹¹

II. EXPERIMENTAL DETAILS

A. Sample preparation and experimental setup

Two crystals of PuCoGa_5 measuring $\sim 5 \times 6 \text{ mm}^2$ and about 1/2 mm thick were grown from excess Ga flux³ as flat plates with the c axis normal to the surface. The crystals were encapsulated in a polyimide coating about 70 μm thick to prevent contamination from particle and ejecta emission. The encapsulated crystals were then attached and sealed under a helium atmosphere inside a Ti cell for further protection. The cell was cooled using a continuous-flow He cold-finger cryostat.

The experiments were performed at the M20 surface muon channel at TRIUMF, Vancouver, Canada. Muons entered the cell through a 50 μm Ti-foil window, with their polarization rotated vertically 90°, perpendicular to the beam momentum. The applied field H_0 was along the incoming beam direction in the TF mode, and parallel to the crystalline c axis. A negligible fraction of the beam stopped in the Ti window and polyimide coating.

The initial set of measurements (after 25 days) were performed with the sample mounted directly on the Ti-cell backing with $H_0 = 60 \text{ mT}$. The second set of measurements (after 400 days, the exact time determined by TRIUMF's beam-time schedule) was carried out in the same geometrical and spin configurations as before, but with the sample surrounded by a Ag backing plate and in applied fields of 60 mT and 0.3 T. All data with $T \leq T_c$ were taken in a field-cooled mode.

B. Data analysis

In an ideal experiment the muons stop randomly on the scale of the flux line lattice (FLL) spacing. Therefore, the muon spin precession signal $\hat{P}(t)$ provides a random sampling of the internal field distribution $n(B)$,¹²

$$\hat{P}(t) \equiv P_x(t) + iP_y(t) = \int_{-\infty}^{\infty} n(B) \exp(i\gamma_\mu B t) dB, \quad (1)$$

where $n(B) = \langle \delta[B - B(r)] \rangle_r$ is the spatial average of a Dirac delta function, $\gamma_\mu (= 2\pi \times 135.53 \text{ MHz/T})$ is the muon gyromagnetic ratio and $B(r)$ is the internal field. This equation indicates that the real amplitude of the Fourier transformed muon precession signal corresponds to the spectral density $n(B)$. In general, an asymmetric $n(B)$, the so-called "Redfield pattern," is expected when the FLL consists of straight, rigid flux lines and the ratio $\kappa = \lambda/\xi$ is not too large.¹³ In such a

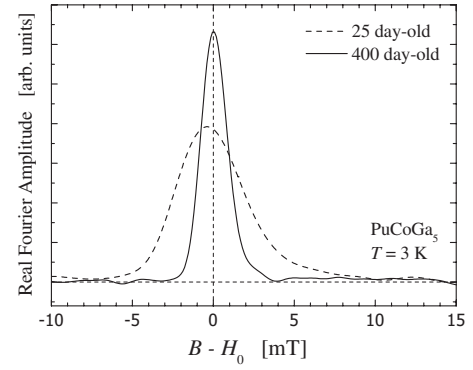


FIG. 1. The fast Fourier transform spectra of the μ SR signals in PuCoGa_5 at 3 K under 60 mT, after subtracting the sample holder signal, in both the 400 day-old sample and the 25 day-old sample. H_0 was determined from a fit to the normal-state data.

case, $n(B)$ possesses a high-field tail which characterizes the flux-line core radius (or coherence length ξ); the second moment $\langle (\Delta B)^2 \rangle$ of $n(B)$ determines the London penetration depth.

The fast Fourier transform (FFT) provides a reasonably good representation of $n(B)$ in the FLL state. Figure 1 shows FFT spectra for the μ SR signals from the sample after 25 and 400 days at $T = 3 \text{ K}$. Here the background signal from the sample cell has been subtracted from the time spectrum before carrying out the FFT. Some age-related differences in the spectra are apparent. First, while the fresh sample exhibits a slightly asymmetric line shape and a small negative field shift, the aged sample shows a nearly symmetric shape with almost no negative shift. Second, the linewidth has narrowed appreciably with aging.

Attempts to fit the fresh sample data using standard models^{14,15} which incorporate a broadened (due to instrumental and nuclear linewidth effects) Redfield pattern for a perfect FLL were not successful. This is due to the fact that the line shape is only moderately asymmetric, and thus unique determinations of λ and the core radius, which depend upon having a high-field tail in the field distribution, cannot be obtained. Most of the decreased asymmetry in the line shape is due to the fact that PuCoGa_5 is a high κ (> 100) superconductor with a relatively large penetration depth (see below), but part may also be due to distortions of the FLL from radiation induced pinning centers, as we discuss below.

As described earlier, reasonable fits to the fresh-sample data were obtained with simple Gaussian or Lorentzian line shapes, and these two functional forms yielded the same temperature dependence for the linewidths.¹¹ Nevertheless, in an attempt to account phenomenologically for any small asymmetry, we also performed fits using a sum of two Gaussians with different centroid frequencies and widths.¹⁶ When these widths were convoluted into a single width (as in Ref. 16) the same temperature dependence and overall linewidth was obtained as for the single Gaussian fits, but with somewhat larger ($\approx 10\text{--}15\%$) uncertainties.

Accordingly, the TF precession spectra for both samples were fit to the sum of two terms, corresponding to muons stopping in the sample and background materials (either Ti or Ag), respectively,

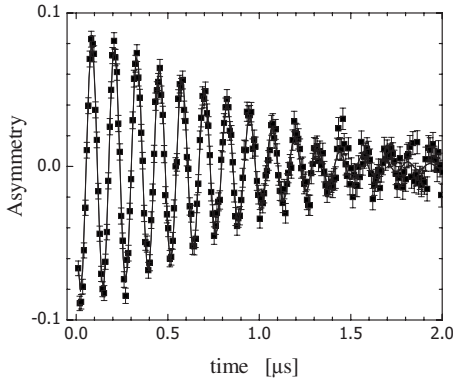


FIG. 2. Muon TF precession signal at $T=4$ K, corresponding to the first term in Eq. (2). The background component was characterized at times between $t=3-10$ μs , where the sample signal is fully relaxed, and subtracted to show the inhomogeneous relaxation produced by the flux lattice in the superconducting state. The solid line is a fit for a Gaussian $P(t)$ in Eq. (2).

$$A_0 G_z(t) = A \cos(\omega t + \phi) P(t) + A_b \cos(\omega_b t + \phi) \exp(-\sigma_b^2 t^2 / 2), \quad (2)$$

where A and A_b and ω and ω_b are the partial asymmetries and central frequencies for PuCoGa_5 and the sample holder, respectively, A_0 is the total positron decay asymmetry ($A_0 = A + A_b$), and ϕ is the initial phase.¹⁷ Here we used $P(t) = \exp(-\sigma^2 t^2 / 2)$. Fits to the data yielded $A/A_b \approx 1/2$. The background signal from Ti in $H_0=60$ mT applied field was well characterized by a Gaussian relaxation function $G_b(t) = \exp(-\sigma_b^2 t^2 / 2)$, with $\sigma_b \approx 0.014 \mu\text{s}^{-1}$. The relaxation rate from the Ag backing used in the measurements on the aged sample is negligible.

The quality of the Gaussian fits for the 25 day-old sample is illustrated in Fig. 2 for data taken at $T=4$ K. Here the $G_z(t)$ data [Eq. (2)] from $t=3-10$ μs were fit separately, and this long-time Ti signal was then subtracted from the total spectrum, leaving only the signal from the sample in the superconducting state. One sees that the Gaussian form for $P(t)$ gives a satisfactory fit. Thus, we conclude that single Gaussian fits give an acceptably accurate measure of the linewidths in the 25 day-old sample. Regarding the 400 day-old data, a single Gaussian fit is obviously appropriate, as can be seen in Fig. 1.

III. RESULTS

A. Flux pinning

We also performed a series of measurements in the aged sample to observe the effects of flux pinning. The sample was cooled to 4 K in $H_0=99$ mT field and a spectrum was obtained. The field was then reduced by 10 mT without changing the temperature and another spectrum was taken. The sample was then warmed in $H_0=89$ mT field, and several spectra were accumulated at increasing temperatures. The results are shown in Fig. 3. In the field-shifted spectrum at 4 K one observes an unshifted line with an amplitude of 90–100 % of the sample signal, together with a line at re-

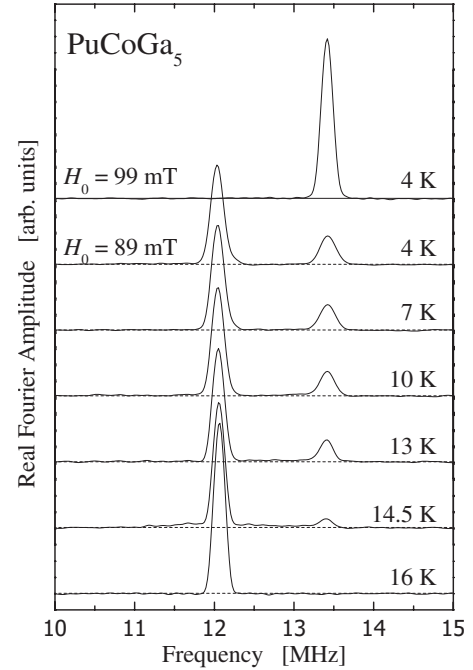


FIG. 3. Temperature dependence of the real Fourier amplitude [transform of $A_0 G_z(t)$] in the 400 day-old sample showing flux trapping. In the top figure the sample was field cooled in $H_0=99$ mT. In the lower figures the field has been reduced to 89 mT with trapped flux giving rise to a resonance at ≈ 13.4 MHz.

duced frequency from muons stopping in the sample backing material. This clearly demonstrates strong pinning, and is consistent with the temperature-independent, field-cooled susceptibilities found in fresh³ and aged¹⁸ samples of PuCoGa_5 . As seen in Fig. 3, the trapped flux gradually leaks out of the sample as the temperature is raised. Note, however, that for the data in Fig. 4, where the field is held constant at the field-cooled value, the sample asymmetry A is independent of temperature, as expected.

B. Temperature dependence of σ

The temperature dependences of σ for the fresh and aged samples for $H_0=60$ mT are shown in Fig. 4(a). In both samples σ increases with decreasing temperature below T_c due to the formation of the FLL. This increase occurs below about 18 K in the fresh sample, in good agreement with the measured T_c . In the 400 day-old sample the data are consistent with a decrease in T_c of about 3 K, in reasonable agreement with the radiation-induced reduction of T_c (≈ 0.24 K/month) reported for PuCoGa_5 samples of slightly different isotopic concentrations.¹⁹

As seen in Fig. 4(a), there is no temperature dependence to the normal state values of σ (denoted σ_n below). Assuming the muons occupy the same sites²⁰ as in CeRhIn_5 we estimate a nuclear dipolar linewidth of about $0.21 \mu\text{s}^{-1}$ for both of the suggested $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ and $(0, \frac{1}{2}, 0)$ sites, which is close to the average of the measured values $\sigma_n \approx 0.2 \mu\text{s}^{-1}$. Furthermore, there is little change in σ_n with aging [Fig. 4(a)], indicating that additional aging has little effect on the muon site(s). None of our conclusions regarding the super-

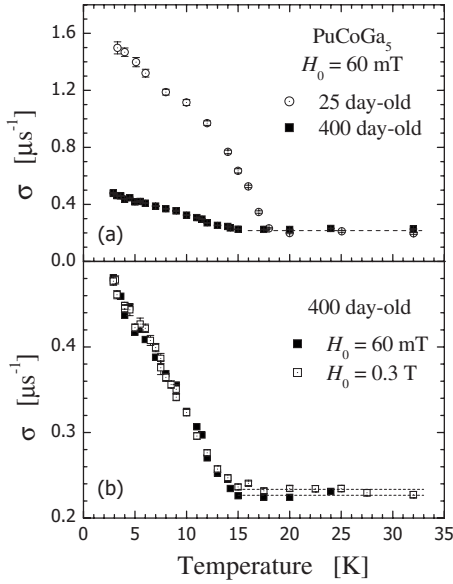


FIG. 4. (a) Temperature dependence of Gaussian muon spin relaxation rate σ for $H_0=60$ mT in the 25 day-old and 400 day-old samples. The dotted line is a guide to the eye. (b) Temperature dependence of σ in the 400 day-old sample for $H_0=60$ mT and 0.30 T.

conducting properties of PuCoGa_5 depend on knowing these exact sites, however.

We also performed measurements with $H_0=0.3$ T in the 400 day-old sample to investigate the field dependence of σ . In some superconductors σ displays a nonmonotonic field dependence which has been attributed²¹ to motion of the FLL in low fields where the interactions between the flux lines is weak due to their large separation. Figure 4(b) shows that both the magnitude and temperature dependence of σ remain essentially unchanged for $60 \text{ mT} \leq H_0 \leq 0.3 \text{ T}$ in the 400 day-old sample. Note that $H_0=60$ mT is at least twice the lower critical field $H_{c1} = \Phi_0 \ln(\lambda/\xi)/4\pi\lambda^2 \approx 0.035 \text{ T}$ reported previously³ and 4–5 times that implied by our measurements of $\lambda(0)$ in the fresh sample, as discussed below. Here $\Phi_0 = hc/2e = 2.07 \times 10^{-15} \text{ Tm}^2$ is the magnetic flux quantum.

C. Temperature dependence of ρ_s and λ

The μSR linewidth from the vortex lattice σ_v is obtained by subtracting the temperature-averaged normal-state linewidth $\bar{\sigma}_n$ from the total linewidth σ in quadrature $\sigma_v^2 = \sigma^2 - \bar{\sigma}_n^2$. The temperature-averaged $\bar{\sigma}_n = 0.204(3) \mu\text{s}^{-1}$ in the fresh sample at $H_0=60$ mT, and $0.227(3) \mu\text{s}^{-1}$ and $0.204(3) \mu\text{s}^{-1}$ for $H_0=60$ mT and 0.3 T, respectively, in the aged sample.

The penetration depth $\lambda(T)$ can be deduced from $\sigma_v(T)$ as follows:^{13,15}

$$\sigma_v(\mu\text{s}^{-1}) = 48300(1-h)[1+3.9(1-h)^2]^{1/2}\lambda^{-2} \text{ (nm)}, \quad (3)$$

where $h=H_0/H_{c2}(0)$ and $H_{c2}(0) \approx 74 \text{ T}$ is the upper critical field in the fresh³ sample, so $h \leq 1$. (H_{c2} is larger in the aged

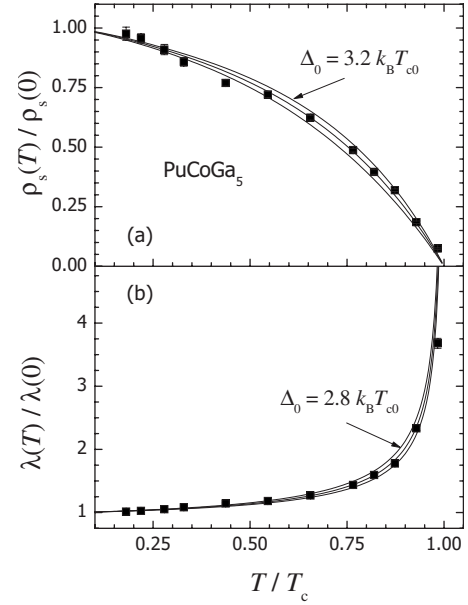


FIG. 5. Temperature dependence of (a) the normalized μSR linewidth $\sigma_v(T) \propto \rho_s(T)$ and (b) penetration depth $\lambda(T)$ for $H_0=60$ mT in PuCoGa_5 after 25 days aging. The solid lines are fits using the d -wave model in the strong scattering limit described in the text for $\Delta_0=2.8, 3.0$, and $3.2 k_B T_{c0}$, where $T_{c0}=18.5 \text{ K}$, $T_c=18.25 \text{ K}$, and scattering rate $\Gamma=0.005\pi k_B T_{c0}$. $\sigma_v(0)=1.52(5) \mu\text{s}^{-1}$ and $\lambda(0)=265(5) \text{ nm}$.

sample.¹⁹) Equation (3) is valid for an isotropic extreme type-II superconductor ($\lambda \gg \xi$) with a hexagonal FLL. The μSR rate σ_v measures the rms width $\langle \Delta B^2 \rangle$ of the field distribution from the FLL: $\sigma_v = \gamma_\mu \langle \Delta B^2 \rangle^{1/2} \propto \rho_s/m^*$, where m^* is the in-plane effective mass. In the following we assume that the temperature dependence of σ_v reflects the temperature dependence of ρ_s , i.e., m^* is temperature independent. Figures 5 and 6 display the temperature dependence of $\rho_s(T)$ and $\lambda(T)$ for the fresh and aged samples, respectively, normalized to their values at $T=0$. The choice of normalization is described below.

D. Possible effects of flux-line lattice distortion

As stated above, the μSR linewidth is directly related to the penetration depth assuming a perfect FLL. In real materials, however, some distortion of the lattice always occurs due to slight misalignments and pinning of the flux lines at defect centers. Therefore, before discussing our data in terms of possible theoretical models, it is important to address these issues in PuCoGa_5 , particularly because radiation damage produces pinning centers.

It is generally agreed that if the FLL is stable, that is, does not melt or move with temperature, then the temperature dependence of $\sigma_v(T)$ accurately reflects the temperature dependence of $\rho_s(T)$. The small scatter in the data observed in Fig. 4 is fully compatible with statistical fluctuations (where Gaussian statistics allow a $\sim 37\%$ probability for fluctuations greater than one standard deviation from the mean), and with nonstatistical errors associated with data fitting, and thus is

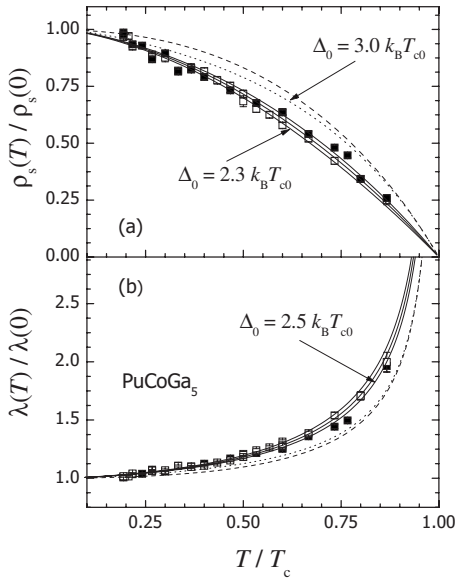


FIG. 6. Temperature dependence of (a) the normalized μ SR linewidth $\sigma_v(T) \propto \rho_s(T)$ and (b) penetration depth $\lambda(T)$ in PuCoGa₅ after 400 days aging. The solid squares are for $H_0=60$ mT and the open squares for $H_0=0.30$ T. The solid lines are fits using the d -wave model in the strong scattering limit described in the text for $\Delta_0=2.3, 2.4$, and $2.5 k_B T_{c0}$, where $T_{c0}=18.5$ K, $T_c=15.0$ K and fitted scattering rate $\Gamma=0.01 \pi k_B T_{c0}$. $\sigma_v(0)=0.43(1) \mu\text{s}^{-1}$ and $\lambda(0)=498(10)$ nm. The dotted curve is the best fit to the fresh sample data in Fig. 5. The dashed curve is for $\Delta_0=2.4 k_B T_{c0}$ and $\Gamma=0.074 \pi k_B T_{c0}$.

not evidence for FLL motion. Furthermore, as discussed above, both susceptibility and μ SR measurements are consistent with strong pinning in PuCoGa₅. These facts, and the absence of a field dependence for $\sigma(T)$ in the aged sample, lead to the conclusion that the temperature dependence of $\sigma_v(T)$ provides a good measurement of $\rho_s(T)/\rho_s(0)$.

The second issue concerns the absolute magnitude of $\sigma_v(T)$. It is evident from Fig. 4 that there is a strong depression of σ_v in the aged sample ($\approx 70\%$) despite only a modest 15–20 % reduction of T_c . Conventional pair-breaking theory, originally developed by Abrikosov and Gor'kov (AG),²² would predict about a 40% reduction in ρ_s for a reduction in T_c of 20%.^{23,24} It is, therefore, important to investigate other possible causes for the reduced linewidth in addition to impurity scattering. These include (1) significant normal-state inclusions in the aged sample which might give rise to linewidth narrowing and (2) the effects of distortions to the FLL caused by pinning at defects.

We first discuss the small probability of large scale normal-state inclusions, noting that a definitive answer to this question requires a detailed knowledge of the number and morphology of the sample defects following room-temperature aging, which is not known with certainty. A ²³⁹Pu nucleus decays into a 5 MeV α particle and an 86 keV recoiling ²³⁵U atom. Most of the damage cascade is caused by the recoiling heavy U atom which has a range of ≈ 17 nm.¹⁸ Lattice vibrations at room temperature usually cause some of this damage to be self-annealed.²⁵ Note, however, that vacancy clusters or dislocation loops with radii

$\leq \xi \sim 2$ nm retain their superconducting character via the proximity effect.²⁶ Unfortunately, reliable TEM studies of aged PuCoGa₅, which could visualize the damage, are presently lacking. We note, however, that the field-shifting experiments shown in Fig. 3 do not show any evidence for flux trapped inside normal-state inclusions, because the asymmetry of the unshifted line is about equal to that measured in a field-cooled state. Given the sensitivity of our measurements, we thus estimate an upper limit for the normal-state fraction in our aged sample of $\leq 10\%$, which is slightly less than, but of the same order as, estimated from recent EXAFS experiments.²⁷ Hence, we find little compelling evidence for large-scale, normal-state inclusions in the aged sample, and dismiss this effect as a significant cause of the observed linewidth narrowing.

We now discuss possible effects of FLL distortion by pinning of vortices. Normally one expects that disorder in the FLL will lead to linewidth broadening. This is, in fact, predicted for the random pinning of stiff, relatively straight vortex lines.¹³ Experimental evidence for such broadening has been reported for many superconductors, e.g., Y(Ni_{0.8}Pt_{0.2})₂B₂C,²⁸ CeRu₂,²⁹ and YBa₂Cu₃O_{6.95}, (Ref. 30). On the other hand, to explain the observed narrowing of the linewidth in some layered high κ superconductors, a model in which point vortices randomly displaced within layers with no alignment or correlation between the layers was developed.³¹ In some cases, this could be topologically similar to the random pinning of segments of flexible vortex lines. If so, then this effect would be most dominant at relatively low applied fields, where the interaction between vortices is weakest. The fact that we observe the same linewidth in the aged sample at both $H_0=60$ mT and 0.3 T seems to argue against this scenario. Furthermore, this model for extreme linewidth narrowing was developed for a highly anisotropic (layered) superconductor. The effective mass anisotropy in PuCoGa₅ can be estimated from the values³² of the slopes of the upper critical fields for $H\parallel c$ and a axes.³³ These data yield only a small mass anisotropy of about 60–70 %, and thus PuCoGa₅ is not a candidate for such a model. We, therefore, conclude that the most likely cause of the sharply reduced linewidth in aged PuCoGa₅ is impurity scattering. Note that since any FLL distortion is, therefore, assumed to broaden the linewidth, the reduction in superfluid density deduced for the aged sample is therefore a lower limit; it could be greater.

IV. MODELING THE DATA

A. Dirty d -wave model

In a superconductor whose electrons are paired in an $L=0$ (s wave) orbital angular momentum state, ρ_s and $\Delta\lambda \equiv \lambda(T) - \lambda(0)$ are relatively temperature independent below about $T/T_c=0.3$, reflecting exponentially activated quasiparticle excitations over a superconducting gap which is non-zero over the entire Fermi surface.²⁶ This is clearly not observed in PuCoGa₅, as seen in Figs. 5 and 6, where the behavior of ρ_s and $\Delta\lambda$ in both the fresh and aged samples is approximately linear at low temperatures.

TABLE I. Parameters derived from comparing μ SR rates $\sigma_v(T)$ in 25 day-old (fresh) and 400 day-old (aged) PuCoGa₅ to a model of dirty d -wave superconductivity with gap Δ_0 , penetration depth $\lambda(0)$ (at $T=0$), impurity scattering rate Γ , and the transition temperature of the nominally pure sample $T_{c0}=18.5$ K. Note that within AG pair-breaking theory the suppressed $T_c=15.0$ K corresponds to $\Gamma=0.074\pi k_B T_{c0}$, which is more than seven times bigger than the Γ needed to fit the T dependence of ρ_s .

Samples	T_c (K)	$\sigma_v(0)$ (μs^{-1})	$\lambda(0)$ (nm)	$n=\Delta_0/k_B T_{c0}$	$\Gamma/\pi k_B T_{c0}$
Fresh	18.25(10)	1.52(5)	265(5)	3.0(1)	0.005
Aged	15.0(1)	0.43(1)	498(10)	2.4(1)	0.010

A pairing state with relative angular momentum $L=2$ (d wave) has been found to produce $\rho_s \propto T$ at low temperatures in the clean high-temperature copper-oxide superconductors.¹⁴ We have, therefore, compared the data in Figs. 5 and 6 to model calculations for the superfluid density ρ_s in a d -wave superconductor. The calculations follow from the standard response formula^{23,34} for quasi-two-dimensional quasiparticles with a cylindrical Fermi surface that relates the superfluid density with the penetration depth $\rho_s(T) \propto 1/\lambda(T)^2$,

$$\frac{1}{\lambda(T)^2} = \frac{\omega_p^2}{c^2 k_B T} \sum_{m=-\infty}^{+\infty} \int_0^{2\pi} d\phi \frac{\cos^2 \phi \Delta(\phi; T)^2}{[\tilde{\varepsilon}_m^2 + \Delta(\phi; T)^2]^{3/2}}, \quad (4)$$

where ω_p is the Drude plasma frequency, c is the speed of light, and $\tilde{\varepsilon}_m$ is the impurity renormalized Matsubara frequency $\varepsilon_m = (2m+1)\pi k_B T$. The impurity self-energy is calculated within the framework of the T -matrix approximation.³⁵ For the clean case this expression simplifies to $1/\lambda(0) = \omega_p/c$. Here $\Delta(\phi; T)$ is the phenomenological gap function given by $\Delta(\phi; T) = \Delta_0 \tanh(b\sqrt{T_c/T-1}) \cos 2\phi$, and Δ_0 and b are phenomenological parameters.^{36,37} We used $b=1.65$, as for the case of weak-coupling d -wave superconductivity, though our results are not very sensitive to this parameter. The relevant scales are set by the magnitude Δ_0 at $T=0$ and the T dependence near T_c : $\Delta(T) \sim b\Delta_0 \sqrt{1-T/T_c}$. The model calculations include the effects of strong impurity scattering to account for the radioactive decay of the Pu atom. Although PuCoGa₅ is not strictly a two-dimensional (2D) superconductor, electronic structure calculations predict several cylindrical Fermi surfaces.⁷⁻⁹ Thus, 2D model calculations for the temperature dependence of the in-plane $\lambda(T)$ should yield qualitatively reasonable comparisons with the data. In what follows, we refer to this model as the “dirty d -wave” model.

The data for the fresh sample are compared to the model calculations in Fig. 5, where $\sigma_v(0)$ and T_c were used as adjustable parameters. Curves for a range of gap parameters Δ_0 are drawn as solid lines to show the sensitivity of the data to the model parameters. Good overall agreement with the data is obtained with this model using a scattering rate $\Gamma = 0.005\pi k_B T_{c0}$, a value consistent with the reduction in T_c according to conventional AG pair-breaking theory (see below). The measured penetration depths and derived parameters are shown in Table I. It is worth noting that the penetration depth of our fresh sample [$\lambda(0)=265(5)$ nm] is in good agreement with NMR measurements [$\lambda(0)$

$=250(5)$ nm].³⁸ The parameter n in the relation $\Delta_0 = n k_B T_{c0}$ is an indirect measure of the coupling strength of the pairing. For a clean weak-coupling d -wave state $n=2.14$. Our best fit to the fresh sample data yields $n=3.0(1)$, indicating an enhanced coupling strength.

The temperature dependence of $\rho_s(T)$ or $\Delta\lambda(T)$ exhibits an essentially linear behavior at low temperatures. There are at least two phenomena which might be expected to destroy this linearity in a d -wave superconductor. The first is nonlocal dynamics in clean superconductors,^{39,40} which causes a T^3 temperature dependence in ρ_s below a temperature $k_B T^* \approx [\xi/\lambda(0)]\Delta_0$. We estimate $T^* \approx (2\text{nm}/265\text{nm})3 \times 18.5$ K ≈ 0.4 K. Thus, we do not anticipate nonlocal effects to be significant in our measurements.

Strong impurity scattering can induce a T^2 dependence in $\Delta\lambda(T)$ at low temperatures in a d -wave superconductor.⁴¹⁻⁴⁴ Conventional AG pair-breaking theory, originally developed for magnetic impurities in an s-wave superconductor,²² has also been found to be applicable to nonmagnetic impurities in a d -wave superconductor.²⁶ The reduction in T_c for $\Delta T_c/T_c \ll 1$ in the presence of impurity scattering is given by $\Delta T_c = \pi\Gamma/4$. For our fresh sample $\Delta T_c \approx 0.25$ K, yielding $\Gamma \approx 0.005\pi k_B T_{c0}$, the scattering rate used to model the data in Fig. 5. The crossover temperature to T^2 behavior for unitary (strong) scattering has been estimated⁴⁴ (independently from the model^{36,37} used in Figs. 5 and 6) to be $T_{cr} \approx 0.83\sqrt{\Gamma\Delta_0}$, where Δ_0 is the maximum gap amplitude. Taking $\Delta_0 = 3k_B T_{c0}$ for the fresh sample yields $T_{cr} \approx 3.3$ K. The crossover from T -linear to T^2 behavior is expected to be smooth, so that $\Delta\lambda(T) \approx bT^2/(T+T_{cr})$.⁴⁴ Therefore, a T^2 behavior is only anticipated for temperatures significantly below T_{cr} ; practically, this works out to be $T \leq T_{cr}/3$, as observed by μ SR (Ref. 45) for Zn doping in YBa₂Cu₃O_{7- δ} . Thus, using either our model or this simpler analytic estimate, we do not expect to see evidence for strong T^2 behavior as long as $T \gtrsim 3$ K in fresh PuCoGa₅, and we do not.

The same model has been used to fit the aged sample data, shown in Fig. 6, where again a range of gap parameters is displayed. The best fits are obtained for $n=2.4$ and $\Gamma = 0.01\pi k_B T_{c0}$. For comparison, the dotted curve in Fig. 6 shows the best fit to the fresh sample data ($\Delta_0 = 3k_B T_{c0}$, $\Gamma = 0.005\pi k_B T_{c0}$), indicating either that the coupling strength has been reduced with aging, or that pair breaking has noticeably reduced the gap value by about 20% after 400 days of aging. The dashed curve in Fig. 6 is a calculation for a dirty d -wave superconductor with $\Delta_0 = 2.4k_B T_{c0}$ and $\Gamma = 0.074\pi k_B T_{c0}$ in the strong scattering limit. This value of Γ is required by conventional AG pair-breaking theory to re-

produce the reduction of T_c in the aged sample, but it clearly leads to poor agreement with the temperature dependence of the superfluid density. Furthermore, as we now discuss, this model does not account for the strong reduction in ρ_s in the aged sample.

B. Short coherence length model

In our discussion above, we concluded that the most likely cause of the sharply reduced linewidth in aged PuCoGa₅ is impurity scattering. This situation is not unprecedented. In comparing doped and radiation-damaged YBa₂Cu₃O_{7- δ} (YBCO) superconductors to the conventional AG pair-breaking theory, it was found in many cases that T_c was remarkably robust, even though ρ_s was easily suppressed.^{46–50} In Ni-doped and He-irradiated YBCO materials, for example, a suppression of T_c by about 20% was accompanied by a suppression of ρ_s by about 70%. This large T_c/ρ_s ratio is in contradiction to conventional “dirty d -wave” theory, and has been addressed theoretically by accounting for the suppression of the order parameter around the vicinity of the defects.^{24,51,52} The conventional treatment assumes that the spatial variation of the superconducting order parameter (of order ξ) is large compared to the average distance \bar{l} between the scattering centers, but large compared to the lattice parameter a_0 , so that the effective order parameter is a spatial average, instead of having a large suppression only near the impurities. As expressed by Franz *et al.*,²⁴ the effect of a spatially inhomogeneous gap is enhanced by a short coherence length relative to the lattice parameter $\xi/a_0 \approx 2–5$.²⁴ As in YBCO, the coherence length in PuCoGa₅ is relatively small (~ 2 nm in both materials) compared to many superconductors where conventional AG pair-breaking theory applies. Although we do not know the actual spacing \bar{l} between defects after self-annealing, we can estimate the mean distance d between Pu atoms which have decayed randomly after 400 days as a crude measure of the spacing between damage cascades. We find $d \approx 20$ nm, which is $\approx 10\xi$. Thus, a spatially inhomogeneous gap model with a short coherence length, as considered by Franz and co-workers,²⁴ may explain our observations.

In this regard, we note that both the suppression of T_c with aging and the value of ξ are about twice as large in PuRhGa₅ compared to PuCoGa₅.^{53–55} Very recently, an enhanced suppression of the superfluid density with chemical doping has also been reported for the related compound CeCoIn_{5- x} Sn _{x} .⁵⁶

In order to test our hypothesis of short-coherence-length superconductivity for PuCoGa₅, we calculated the superfluid density by solving the Bogoliubov–de Gennes (BdG) equations of a 2D d -wave superconductor

$$\sum_j \begin{pmatrix} \mathcal{H}_{ij} & \Delta_{ij} \\ \Delta_{ij}^* & -\mathcal{H}_{ij}^* \end{pmatrix} \begin{pmatrix} u_j^n \\ v_j^n \end{pmatrix} = E_n \begin{pmatrix} u_i^n \\ v_i^n \end{pmatrix}. \quad (5)$$

Here $(u_i^n, v_i^n)^T$ are the eigenfunctions at site i corresponding to the quasiparticle excitation energy E_n . The normal-state lattice Hamiltonian is

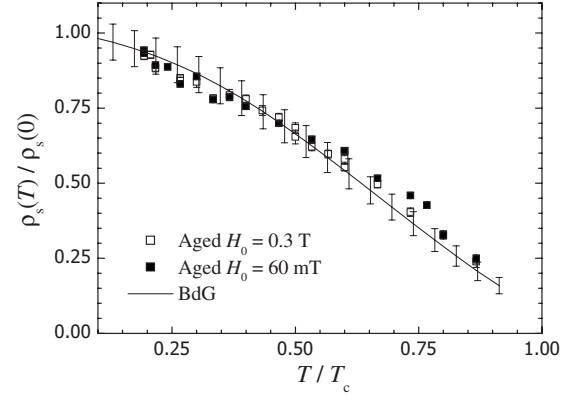


FIG. 7. Temperature dependence of the normalized superfluid density $\rho_s(T)/\rho_s(0) = \lambda(0)^2/\lambda(T)^2$ in PuCoGa₅. The solid squares are for $H_0 = 60$ mT and the open squares for $H_0 = 0.30$ T after 400 days, using $\sigma_v(0) = 0.45(1) \mu\text{s}^{-1}$ corresponding to $\lambda(0) = 487(10)$ nm. The solid line is the short-coherence-length BdG superfluid density [for $\rho_s(0; n_{\text{imp}} = 6\%) / \rho_s(0; n_{\text{imp}} = 0\%) \approx 0.35$] in the strong scattering limit described by $\Delta_0 = 0.187t$, $T_c = 0.115t$, and impurity concentration $n_{\text{imp}} = 6\%$. For the pure case, $n_{\text{imp}} = 0\%$, one finds $\Delta_0 = 0.314t$ with $T_{c0} = 0.143t$. The statistical error bars are due to averaging over 20 random impurity configurations at fixed concentration.

$$\mathcal{H}_{ij} = -t\delta_{i+\delta,j} + (U_{\text{imp}} - \mu)\delta_{ij}, \quad (6)$$

where t is the hopping integral between a specific lattice site and its four nearest neighbors as denoted by $\delta = (\pm 1, 0)$ and $(0, \pm 1)$, μ is the chemical potential, and U_{imp} is the impurity potential modeling the on-site disorder. The self-consistency equation for the gap function of a d -wave superconductor on a lattice is given by

$$\Delta_{i,j=i+\delta} = \frac{V}{2} \sum_n [u_i^n v_j^{n*} + u_j^n v_i^{n*}] \tanh(E_n/2k_B T), \quad (7)$$

with V being the pairing strength. Note that the quasiparticle energy is measured with respect to the chemical potential. We follow an iterative procedure to solve self-consistently the BdG equations by exact diagonalization on a 20×20 lattice, and, for comparison, on a 24×24 lattice. Using a suitable guess for an initial Δ_{ij} , a new one is then calculated, and the process is iterated until the desired convergence is achieved.

The linear response calculation of the superfluid density ρ_s for the lattice model follows the approach described by Scalapino *et al.*⁵⁷ and was applied to 2D d -wave^{24,58} and s -wave^{59,60} superconductors. A detailed technical description of the response calculation will be given elsewhere.⁶¹ Our calculations of the transition temperature T_c , the lattice-averaged zero-temperature order parameter $\Delta_0 = \langle \Delta_{ij}(0) \rangle$ and superfluid density $\rho_s(0)$ are in good agreement with those of Franz *et al.*²⁴ for the same set of parameters as used by them ($V = 1.13t$, $U_{\text{imp}} = 100t$, $\mu = -0.36t$). Because of finite size effects of our lattices, we averaged our calculations over 20 random impurity configurations at fixed concentration, as indicated by the statistical error bars in Fig. 7.

In Fig. 7 we compare the temperature dependence of $\rho_s(T)$ for the aged sample and the BdG response calculation in the limit of strong impurity scattering. The impurity concentration $n_{\text{imp}}=6\%$ was chosen to reproduce the observed suppression of T_c and $\rho_s(0)$ of the aged vs fresh sample. The relevant parameters are given in the caption to Fig. 7. The BCS coherence length is given by $\xi_0 \sim \hbar v_F / (\pi \Delta_0)$, where the Fermi velocity is given approximately by $v_F \sim a_0 t / \hbar$ with $\Delta_0 = 0.187t$, so that $\xi_0 \sim 2a_0$. The excellent agreement between the short-coherence-length BdG calculation and the measured superconducting properties of the aged sample, and, at the same time, the failure of the dirty d -wave calculation within AG pair-breaking theory to describe the data, indicates that the uniform, dilute-impurity theory by Abrikosov and Gor'kov is not applicable to PuCoGa₅, where superconductivity is seemingly not uniformly suppressed. Specifically, both the BdG and AG dirty d -wave models require $k_f \bar{\ell} \gg 1$, where k_f is Fermi wave vector. However, the former case requires strong impurity scattering with on-site suppression of Δ_0 and $k_f \bar{\ell} \sim 1$, while the AG pair-breaking theory requires a uniformly suppressed Δ_0 with $\xi / \bar{\ell} \lesssim 1$.

V. DISCUSSION AND SUMMARY

We now summarize our principal results. The radioactive decay of ²³⁹Pu allows one to study the effects of pair-breaking defects on the properties of the superconductivity in PuCoGa₅. The low-temperature, quasilinear temperature dependences of the superfluid density and penetration depth in both fresh (25 day-old) and aged (400 day-old) PuCoGa₅ are consistent with a line of nodes in a d -wave order parameter.

The fresh sample is almost defect free, as evidenced by the small reduction in T_c , e.g., $\Delta T_c / T_{c0} \approx 1.4\%$. We have, therefore, compared our data in the fresh sample to a dirty d -wave model for a strong-coupling superconductor in the presence of strong impurity scattering. We find that the zero-temperature penetration depth and superconducting gap are $\lambda(0) = 265(5)$ nm and $\Delta_0 = n k_B T_{c0}$ with $n = 3.0(1)$, respectively. The latter is enhanced compared to the weak-coupling d -wave case, where $n = 2.14$. The model scattering rate used was consistent with the slight reduction in T_c for the fresh sample, according to conventional AG pair-breaking theory. Our results on the fresh sample are consistent with recent NQR and NMR experiments in fresh PuCoGa₅.^{5,38}

In the case of the aged sample, we argued that the presence of strong pinning in PuCoGa₅ allows an accurate measure of the temperature dependence of $\lambda(T)$, but that distortion of the FLL (which broadens the linewidth) means that the measured magnitude of $\lambda(0)$ is only a lower limit. The dirty d -wave model can also fit the temperature dependence of the superfluid density $\rho_s(T)$ [or $\lambda(T)$] with Δ_0 reduced by about 20%, but with a scattering rate nearly an order of mag-

nitude smaller than predicted by conventional AG pair-breaking theory. Furthermore, the dirty d -wave model is unable to account for the reduction of at least 70% in $\rho_s(0)$. This is attributed to the fact that PuCoGa₅ possesses a relatively short coherence length, and, therefore, the conventional AG pair-breaking theory, in which the order parameter is spatially averaged over impurity sites, is inappropriate. This was pointed out by Franz *et al.*²⁴ for damaged or doped YBCO superconductors, but in that work a comparison with the data was made only for zero temperature.

Accordingly, we have modeled the full temperature dependence of $\rho_s(T)$ in aged PuCoGa₅ by solving the BdG equations for a short-coherence-length, weak-coupling d -wave superconductor in the strong scattering limit. (For small scattering rates the short-coherence-length model agrees fairly well with the dirty d -wave model.²⁴) The BdG model is able to reproduce quite well the temperature dependence and magnitude of ρ_s for a nominal 6% impurity concentration, chosen to reproduce the magnitude of $\rho_s(0)$. This impurity concentration is reasonable, though we attach no particular importance to its exact value. As in the dirty d -wave model, the BdG model is consistent with a reduction of Δ_0 in the aged material, compared to the fresh sample. Both the dirty d -wave and BdG models are consistent with $\lambda(0) = 498(10)$ nm in aged PuCoGa₅, where we remind the reader that because of FLL distortion this is a lower limit.

Computational feasibility dictates that the BdG lattice model calculations are for a two-dimensional system. Therefore, the derived gap parameters are only semiquantitative. [The values of $\lambda(0)$ are accurately determined from Eq. (3), however, and our quoted values are derived from this expression.] Nevertheless, we do not expect strong deviations between 2D and 3D models for the in-plane penetration depth, and, therefore, believe that a 2D model should provide a good qualitative description of the essential physics in this interesting superconductor.⁶² Our data and analysis in aged PuCoGa₅ suggest that although small parts of the sample are significantly disordered, and consequently the superfluid density is strongly suppressed in these regions, superconductivity remains remarkably resilient.

ACKNOWLEDGMENTS

This work was partially supported by a Grant-in-Aid for Scientific Research (Grant No. 18027014), the Ministry of Education, Culture, Sports, Science and Technology, Japan. Work at LANL (Contract No. DE-AC52-06NA25396) and LLNL (Contract No. W-7405-Eng-48) was performed under the U.S. Department of Energy. Work at Riverside was supported by the U.S. NSF, Grant No. DMR-0422674. We thank the staff of TRIUMF and acknowledge helpful discussions with I. Affleck, M. Franz, G. Lander, P. M. Oppeneer, J. E. Sonier, J. D. Thompson, and F. Wastin.

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