Limits for ordered magnetism in Pu from muon spin rotation spectroscopy

R. H. Heffner,[1,2] G. D. Morris,[3,2] M. J. Fluss,[4] B. Chung,[4] S. McCall,[4] D. E. MacLaughlin,[5] L. Shu,[5] K. Ohishi,[1] E. D. Bauer,[2] J. L. Sarrao,[2] W. Higemoto,[1] and T. U. Ito[6,1]

[1]*Japan Atomic Energy Agency, Tokai-mura, Ibaraki-ken 319-1195, Japan*

[2]*Los Alamos National Laboratory, K764, Los Alamos, New Mexico 87545, USA*

[3]*TRIUMF, 4004 Wesbrook Mall, Vancouver, British Columbia V6T 2A3, Canada*

[4]*Lawrence Livermore National Laboratory, P.O. Box 808, Livermore, California 94550, USA*

[5]*Department of Physics, University of California, Riverside, California 92521, USA*

[6]*Department of Physics, Tokyo Institute of Technology, Meguro-ku, Tokyo 152-8551, Japan*

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We present μSR measurements on Pu metal which set stringent upper limits on the magnitude of the ordered moments μ_{ord} in α-Pu and δ-stabilized Pu (alloyed with 4.3 at.% Ga). The magnitude of the low-temperature ($4\ \mathrm{K} < T < 100-150\ \mathrm{K}$) μSR rate in zero applied field is independent of temperature and consistent with nuclear dipolar broadening alone. The Knight shift in α-Pu in 2 T applied field is also independent of temperature below about 100 K, but increases abruptly at higher temperatures, an effect likely caused by muon diffusion to paramagnetic impurities. A rough estimate for the muon hopping rate $1/\tau_h$ at 150 K in α-Pu yields $1/\tau_h \approx 3\times 10^7\ \mathrm{s}^{-1}$; no estimate is yet obtained for δ-Pu. The principal results reported here are limits for μ_{ord} in both α- and δ-Pu $\leqslant 10^{-3}\mu_{\mathrm{B}}$ at $\mathrm{T} \cong 4$ K. These limits are discussed in terms of recent models of the electronic properties of δ-Pu, where magnetism has been previously predicted. Our results are consistent with, but cannot unambiguously distinguish between, cases where the moments in δ-Pu are essentially zero, possess very weak interatomic exchange, are reduced by hybridization, or are washed out by dynamical fluctuations.

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I. INTRODUCTION

There has been a resurgence of interest in the electronic properties of actinide materials recently, and in particular in the properties of Pu metal and Pu compounds. Pu exists in six different allotropic phases[1] as a function of temperature and volume. The low temperature α phase possesses a monoclinic structure. As temperature increases the volume expands significantly before melting at 912.5 K. Interest has centered on the high-temperature δ phase (near 700 K), which has an fcc structure and a unit cell volume which is 26% larger than α-Pu. Theoretical calculations attribute the large volume increase in δ-Pu to the partial localization of the f electrons,[2] which is confirmed by recent photoemission[3] experiments. Because of this partial localization many theories of the electronic structure in δ-Pu have predicted magnetic order.[4] There is, however, a clear lack of experimental evidence for such ordering.[5] For example, neutron scattering experiments set an upper limit of only $0.4-0.04\ \mu_B$ for the ordered moment in δ-Pu.[5]

Given this situation, we undertook μSR experiments designed to answer the question: Can magnetism, either ordered or *disordered* freezing of the spins, be completely eliminated in Pu metal? In Kondo lattice systems, for example, small magnetic moments can survive at temperatures much less than the effective Kondo temperature. μSR is an excellent probe to address this issue because the upper limits set by neutron scattering are far larger than what are achievable with μSR ($\leqslant 0.001\mu_B$). Furthermore, μSR is equally sensitive to the freezing of *disordered* spins, and, because μSR is a local probe, the measurement is equivalent to a sum over all points in momentum space. Thus, more stringent upper limits can be set than with most other probes. In this paper we summarize our initial investigations of the magnetic properties of Pu metal. Some of this work has been reported previously.[6]

II. μSR EXPERIMENTS IN α- AND δ-Pu

The measurements[7] were performed on α-Pu and Ga-stabilized δ-Pu (4.3 at. % Ga) at the M20 surface muon channel at TRIUMF in Vancouver, Canada, using positive muons (μ^+). The data were taken with the muon spin rotated approximately 90 deg vertically from the incoming muon momentum. For experiments in zero applied field (ZF) any residual field was reduced to $\leqslant 10^{-6}$ T using trim coils. Special care had to be taken in order to perform μSR experiments on these radioactive materials. To prevent possible contamination, a Kapton coating approximately 70 μm thick was applied to all of the samples, which were then placed under He atmosphere inside a closed titanium cell having a 50 μm thick Ti-foil beam window. The entire cell was then attached to a continuous-flow, He cold-finger cryostat mounted along the incident beam direction. A negligible fraction of the muons stopped in the Ti window or the Kapton coating. The background signal from an empty Ti holder in either ZF or $H_0 = 0.06$ T transverse field (TF) was well characterized by a Gaussian relaxation function $G_{\mathrm{Ti}}(t) = \exp(-\sigma_{\mathrm{Ti}}^2 t^2/2)$ with $\sigma_{\mathrm{Ti}} \approx 0.014\ \mu\mathrm{s}^{-1}$.

The α-Pu sample (99.98% pure) was electromechanically refined two years before the first set of measurements, shaped to a disk approximately 12 mm diameter and 0.1 mm thick, and cleaned of oxide. The sample consisted of ^{239}Pu (93.7%), with smaller concentrations of ^{240}Pu (5.86%) and ^{238}Pu (0.17%), together with magnetic impurities Fe(235 at. ppm), Ni(24 at. ppm),

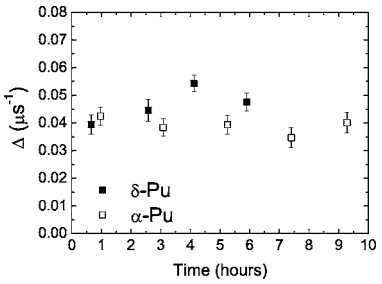

FIG. 1. The ZF static Gaussian Kubo-Toyabe widths in α-Pu and δ-Pu are independent of time, showing no significant effects of accumulated radiation damage. The times are the midpoint times for a particular run.

Cr(12.4 at. ppm), and Mn(10.4 at. ppm). The δ-Pu sample was synthesized from the same Pu source as the α-Pu sample, alloyed with Ga and prepared in the same manner, thus containing about the same magnetic impurity levels as the α-Pu.

A. Zero and low TF measurements

The μSR data were fit using the sum of two relaxation functions, one for Pu and one for Ti. A Gaussian Kubo-Toyabe (GKT) function[8] and a simple Gaussian $[\exp(-\sigma^2 t^2/2)]$ were used for ZF and TF fits, respectively. The rate for Ti was fixed at its measured value, and the relative amplitudes at low fields were fixed at a ratio determined in a separate experiment with a blank Cu pellet the same size as the Pu samples.

Radioactive decay of ^{239}Pu (half life 2.4×10^4 y) damages the lattice by producing interstitials and vacancies. For the α- and δ-Pu specimens the displacement damage cascade is immobile below about 30 K,[9] and for both materials nearly all of the accumulated damage is annealed at room temperature [>95% for α-Pu and 100% for δ-Pu (Ref. 9)]. However, U, Am, and He from the Pu decay remain and continue to accumulate.

To check that the muon sensed only undamaged regions, we allowed the samples to sit for up to nine hours at $T\sim4$ K, during which time μSR runs were performed about every 2 h. No significant change in the μSR rates were observed over this period, as seen in Fig. 1 which shows the GKT rate Δ for α- and δ-Pu as a function of time. Thus, no observed cumulative effects from radiation damage were observed. For subsequent data sets the samples were always reannealed at 300 K every 6–8 h.

The temperature dependencies of the ZF rates (Δ) and TF ($H_0=0.25$ T) rates (σ) in α-Pu and δ-Pu are shown in Figs. 2 and 3. (The TF data for α-Pu shown in Fig. 3 result from an improved analysis compared to the provisional analysis presented previously.[6]) A small longitudinal field ($\sim$0.005 T) reduced the ZF μSR rate to zero, indicating a static distribution of local fields at μ^+ sites.[8] In both samples the data are T independent within statistical errors below about 100 K. A small magnetic field drift ($\ll\sigma$ over a 1 h run) prevented an accurate analysis of the temperature dependence of the muon frequency at $H_0=0.25$ T.

We are unable to determine the muon sites from these data; however, none of our conclusions depend upon where the muon resides. We have calculated ZF nuclear dipolar linewidths of 0.018 and 0.023 μs^{-1} for the octahedral and tetrahedral positions in δ-Pu, respectively. The observed values are higher, indicating that the muon is perturbed from these high symmetry sites due to the Ga doping or to strain fields.

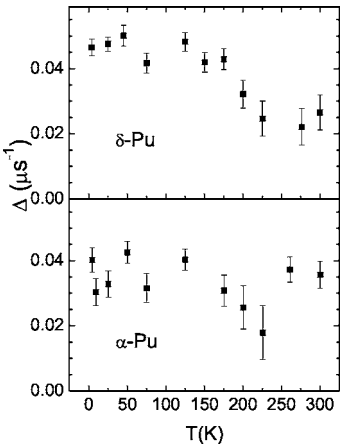

FIG. 2. ZF static Gaussian Kubo-Toyabe widths in α- and δ-Pu between $T=3.8$ and 300 K. Muon diffusion likely causes the reduction of the linewidths above 150 K (see discussion of Knight shift in α-Pu).

B. High-field Knight shifts in α-Pu

To investigate the local susceptibility in Pu metal we performed a second set of measurements at applied fields $H_0=0.25-2.0$ T on the same sample of α-Pu, but approximately one year later. (Unfortunately, a sample of δ-Pu was not available for these measurements.) The applied field was stabilized to within 10 ppm, and the sample was surrounded by a Ag backing foil which covered the Ti base plate. The Ag signal was used as a measure of the applied field, using the known Ag Knight shift of +94 ppm.[10] The μSR rates obtained at lower fields agree quite well with rates obtained one year earlier on this sample in $H_0=0.25$ T, indicating no dis-

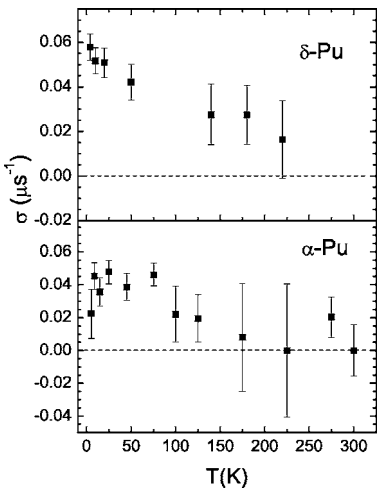

FIG. 3. Temperature dependence of the TF Gaussian widths σ in α- and δ-Pu.

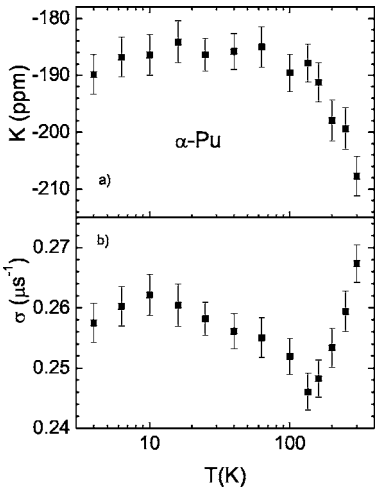

FIG. 4. Temperature dependence of the (a) Knight shift K and (b) Gaussian linewidth σ for $H_0=2.0$ T in α-Pu.

cernible effects from radiation damage over the previous year.

The muon Knight shift K is defined as $K=(\nu-\nu_0)/\nu_0$, where ν is the measured frequency and $2\pi\nu_0=\gamma_\mu H_0$, where γ_μ is the muon's gyromagnetic ratio (8.51×10^8 Hz/T). Generally, $K=K_0+K_{\rm dem}+A\chi_f(T)/N_A\mu_B$, where $K_{\rm dem}$ is the shift caused by the demagnetization fields, χ_f is the temperature-dependent f-electron susceptibility, K_0 is the shift from temperature-independent sources, and A is the total hyperfine coupling field between the muon and the Pu moments. The constants N_A and μ_B are Avogadro's number and the Bohr magneton, respectively. $K_{\rm dem}=4\pi(\frac{1}{3}-N)\rho_{\rm mol}\chi$, where $\rho_{\rm mol}$ is the molar density and N is the geometrical demagnetization factor.

Figure 4(a) shows the raw Knight shift K as a function of temperature for $H_0=2.0$ T; the associated Gaussian linewidth σ is given in Fig. 4(b). Note that the magnitude of σ is much larger at $H_0=2.0$ T than at 0.25 T (see Fig. 3). The rough magnitude of σ at $H_0=2.0$ T can be understood by assuming a distribution of anisotropic Knight shifts in the polycrystalline sample of width $\delta K\approx|K|$. Thus, one expects $\sigma\approx\gamma_\mu H_0|K|$, which yields $\sigma\approx0.32$ μs^{-1} at $T=4$ K, close to the observed value.

One sees in Fig. 4(a) that the Knight shift data are essentially independent of temperature below 100 K. In particular, there is no indication of the rise in susceptibility seen below about 80 K in Fig. 5, indicating that this rise is probably an impurity effect, as discussed below. We calculate $K_{\rm dem}\approx-3.4\times10^{-4}$ using the high-temperature $\chi\approx5.1\times10^{-4}$ emu/mol (see Fig. 5) and $N\cong0.98$ for our sample. Thus, the intrinsic shift $K-K_{\rm dem}\approx156$ ppm at low temperatures is extremely small, near the limits of our detection capabilities. Because there is no measurable temperature dependence for $K<100-150$ K, we cannot separate the f-electron contribution to the shift from K_0 and, hence, cannot determine the μ^+-Pu coupling constant A from these data. The change in linewidth [Fig. 4(b)] and shift above 100–150 K is similar to the trends in the ZF and TF linewidths (Figs. 2 and 3, lower panels) and is attributed to rapid muon diffusion, as discussed below.

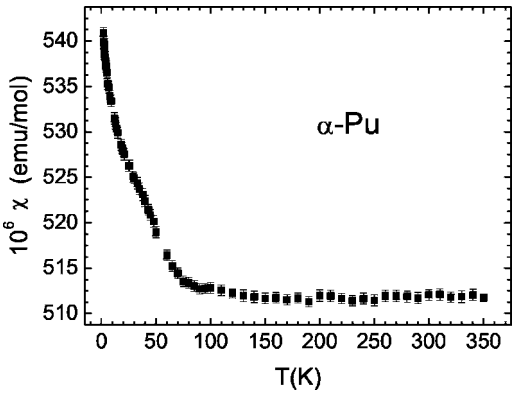

FIG. 5. Temperature dependence of susceptibility in α-Pu.

III. DISCUSSION

A. Possible effects of magnetic impurities

Before drawing conclusions from our measurements about the magnetism in Pu, it is important to determine whether the magnetic (Fe) impurities in our sample have any significant influence on our measurements. We conclude that they do not, as we now discuss. There are two possible scenarios. First, the Fe impurities may freeze into a spin-glass state at low temperatures. For this possibility one may refer to μSR experiments in the spin-glasslike state of AuFe(1%).[11] A μSR rate λ of ≈5 μs^{-1} is found in that material at $T=4$ K ($T_g=9.1$ K),[11] which scales to $\lambda\approx0.12$ μs^{-1} for 235 at. ppm Fe, but only below the estimated[12] glass temperature $T_g=235$ mK. Thus, we expect the Fe spins to exhibit paramagnetic behavior in Pu metal above $T=4$ K.

A second impurity-driven effect may occur if the Fe impurities remain paramagnetic but influence either the measured linewidths or Knight shifts. The inhomogeneous TF linewidth Λ from a dilute concentration of magnetic impurities has been calculated for the case of dipole coupling applicable here.[13] One finds a Lorentzian $\Lambda=5.065\rho cB_{\rm dip}\langle S_z\rangle$, where c is the impurity concentration in a lattice of ρ sites per cubic centimeters, $B_{\rm dip}$ is the dipole coupling constant, and $\langle S_z\rangle$ is the magnitude of the induced spin in field H_0, averaged over the available S_z substates using a Boltzmann distribution. The quantity $\langle S_z\rangle$ is nearly proportional to H_0/T. For $H_0=2.0$ T we calculate an equivalent Gaussian rate ($\approx\Lambda\sqrt{2}$) of 0.13 μs^{-1} at $T=4$ K, which falls to 0.006 μs^{-1} by 100 K. At $H_0=0.25$ T the calculated rates are 0.019 and 0.0008 μs^{-1} at $T=4$ and 100 K, respectively. Not only are the experimentally measured rates approximately independent of temperature below about 100 K, but the calculated impurity-induced rates are dominated by the anisotropic Knight shift at 2.0 T and contribute only marginally to the measured TF rates at 0.25 T, as we now show. Consider the case of α-Pu. The average ZF linewidth Δ in α-Pu is about 0.035 μs^{-1} below 100 K, which should[8] yield a TF rate in $H_0=0.25$ T of $\sigma=0.035/\sqrt{\{5/2\}}=0.022$ μs^{-1} if the rates arise from nuclear dipoles alone. Instead the measured TF rate at $H_0=0.25$ T and $T=4$ K is about 0.04 μs^{-1}. The larger measured TF rate can be accounted for by additional

broadening from the anisotropic Knight shift shown in Fig. 4(a), which yields a contribution after extrapolating to H_0=0.25 T of 0.033 μs^{-1}. Thus, we would estimate $\sigma=\sqrt{\{0.022^2+0.033^2\}}=0.040$ μs^{-1} from these two sources. The calculated static width from the Fe impurities would add in quadrature to this value, and is thus relatively less important. This estimate of the importance of anisotropic broadening in the TF linewidths is valid for α-Pu, where we have high-field Knight shift data, but we believe the argument is also valid for δ-Pu because the magnitudes of χ (and, hence, of the Knight shift) in the two materials are similar.[9]

The contribution to the Knight shift K from the Fe is also negligible, for two reasons. First, the Curie susceptibility from the dilute Fe impurities is small, estimated to be $\approx 10^{-4}$ emu/mol. Second, the hyperfine field A falls off as the inverse cube of the μ^+-Fe distance, and thus should be $\approx 10^{-3}$ times a typical $A \approx 0.1 T/\mu_B$ for near neighbor coupling. Thus, the presence of magnetic impurities in our samples does not affect our results below about 150 K.

We now address the abrupt change in the Knight shift and increase in linewidth in α-Pu above 150 K. We postulate that this temperature dependence is caused by the onset of rapid muon diffusion, so that the muon's spin is relaxed and its frequency shifted as it diffuses to the vicinity of a Fe atom. This situation was studied previously for dilute paramagnetic impurities in Au, where the muon diffusion rate was measured.[14] We crudely estimate the muon hopping rate $1/\tau_h$ in α-Pu near 150 K (where σ begins to increase), as follows. We assume the muon must make n hops in its lifetime (2.2 μs) to travel an average distance $d/2$ to reach a Fe impurity, where d is the mean distance between Fe atoms. Assuming no clustering $d \approx a/c^{1/3}$, where a is the lattice constant and c=235 ppm. This yields $1/\tau_h \approx 3 \times 10^7$ s^{-1}, to be compared with 4×10^9 and 7×10^5 s^{-1} for muons in Au and Cu at 150 K, respectively.[14] High-temperature muon diffusion in δ-Pu is also likely, though an order-of-magnitude estimate for the hopping rate is not yet determined.

B. Nonobservation of ordered magnetism

The fundamental conclusion from our experiments in metallic Pu is, therefore, that the observed small, static, random field distribution and lack of temperature dependence below ~100 K in zero applied field is consistent with nuclear dipolar broadening *alone*; that is, *there is no evidence for any ordering whatsoever of electronic moments down to T* $\cong$4 K. The uniform ordering of Pu moments of any significant size ($\sim 0.01 \mu_B$) (e.g., ferro- or anti-ferromagnetism), or the disordered spin freezing of such moments, would produce either a precessing μSR signal below the ordering temperature or a T-dependent, exponential rate ten times that which is observed.

Our data thus allow us to set a limit on the ordered moment μ_{ord} in both Pu materials. Using a typical muon hyperfine field found in f-electron systems of $A \sim 0.1$ T/μ_B and taking $\Delta \leqslant 0.05$ μs^{-1} as an upper limit for the relaxation rate from the hypothesized ordering, one has $\mu_{ord} \leqslant \Delta/(A\gamma_\mu) \simeq 10^{-3}$ μ_B at $T \cong 4$ K.

This result is expected for α-Pu, where no ordered magnetism is predicted. A possible reason for our failure to observe magnetic ordering of the f-electron spins in δ-Pu is that they may not order until $T \ll 4$ K, so that their mean fluctuation rate is still too rapid to produce a significant μSR rate in the time scale of our measurement. To address this issue we, therefore, calculate the expected exchange frequency ω_e and hypothetical Néel temperature T_N from our μSR data using mean field theory. As a starting point, we assume that Pu spins with moment $1\mu_B$ are fluctuating at ω_e, giving rise to a temperature-independent dynamical μSR rate $1/T_1 \leqslant \Delta = 0.05$ μs^{-1}. (The value of $1\mu_B$ is typical of many calculations predicting magnetism in δ-Pu.) Moriya has shown that[15]

$$1/T_1 = \sqrt{2\pi} S(S+1)(\gamma_\mu g_{Pu} A \mu_B)^2/(3\omega_e), \tag{1}$$

where S and g_{Pu} are the Pu spin and g factor, respectively. The exchange frequency is related to the exchange integral J via the expression $\hbar\omega_e = J\sqrt{2zS(S+1)/3}$, where z is the number of nearest neighbor Pu spins to a given Pu spin (z=12 for δ-Pu). Finally, $T_N = zJS(S+1)/3k_B$.[15] Using a typical value for A=0.1 T/μ_B as discussed above, one obtains $\omega_e = 1.5 \times 10^{12}$ s^{-1} and $T_N \approx 2.2$ K. Note that because we measured essentially zero μSR rate in the longitudinal field, the actual lower detectable limit for ω_e is at least several times larger than this, raising the estimated T_N as well. Thus, within the context of this simplified estimate, it is very unlikely that we would see no increase in relaxation below 10 K if there were magnetic ordering. Consequently, this analysis of the spin dynamics leads one to conclude that either (1) the actual moment must be $\ll 1\mu_B$ or (2) the exchange interaction is much smaller than the mean-field estimate.

To summarize, our results are incompatible with theories predicting any magnetism for $T \geqslant 4$ K in δ-Pu. In addition to predictions of relatively large moments ($\geqslant 1\mu_B$), the approximate cancellation of the orbital and spin moments,[16] as well as other more exotic theoretical possibilities such as noncollinear intra-atomic magnetism,[17] are extremely unlikely in view of the limits set by μSR. Recent calculations[18] using the local density approximation with on-site Coulomb repulsion and spin-orbit coupling do predict essentially zero moment in δ-Pu; however, they also predict a negligible f-electron spectral density at the Fermi surface, which is incompatible with photoemission[3] experiments. Thus, the electronic structure in δ-Pu remains unexplained, although progress is being made. Whether the moments in δ-Pu are essentially zero, are reduced by hybridization,[19] or washed out by dynamical fluctuations[20] is currently unknown.

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