PHYSICAL REVIEW B
CONDENSED MATTER

THIRD SERIES, VOLUME 37, NUMBER 7 1 MARCH 1988

Muon spin rotation and magnetic order in the heavy-fermion compound URu_2Si_2

D. E. MacLaughlin
University of California, Riverside, California 92521

D. W. Cooke, R. H. Heffner, R. L. Hutson, M. W. McElfresh, M. E. Schillaci,
H. D. Rempp, J. L. Smith, J. O. Willis, and E. Zirngiebl
Los Alamos National Laboratory, Los Alamos, New Mexico 87545

C. Boekema
California State University, San Jose, California 95192

R. L. Lichti
Texas Tech University, Lubbock, Texas 79409

J. Oostens
University of Cincinnati, Cincinnati, Ohio 45221

(Received 8 June 1987)

Muon spin rotation and relaxation experiments have been carried out in the paramagnetic and magnetically ordered states of the magnetic heavy-fermion superconductor URu_2Si_2. The positive-muon (μ^+) linewidth σ_μ in zero and low applied fields ($\lesssim 100$ Oe) was consistent with nuclear dipolar broadening at temperatures above the ordering temperature T_N, but increased below T_N to a value of $\sim 0.1\ \mu s^{-1}$. This is definite evidence for magnetic ordering. However, the value of σ_μ corresponds to a rms local field of only ~ 1 Oe, which is more than an order of magnitude smaller than expected from an antiferromagnetic (AF) array of U moments $\mu \sim 0.03\mu_B$, as obtained from neutron diffraction. Either the μ^+ site is approximately symmetrically placed between the AF sublattices, or the muon suppresses AF ordering in its vicinity. The isotropic component of the observed μ^+ frequency shift is small, and the anisotropic shift is of the order of the calculated dipolar contribution for several candidate μ^+ sites (including the symmetric one).

I. INTRODUCTION

The ternary intermetallic compound URu_2Si_2 is unique among uranium-based materials in that it exhibits both magnetic ordering and a superconducting phase transition at low temperatures.[1-3] A form of antiferromagnetic (AF) order, possibly accompanied by a charge density wave (CDW), sets in at a Néel temperature $T_N \simeq 17.5$ K, and superconductivity is observed below a critical temperature $T_c \simeq 1.2$ K. The large linear specific-heat coefficient $\gamma = 70-180\ \mathrm{mJ\,mol^{-1}\,K^{-2}}$ classifies URu_2Si_2 as a "moderately" heavy-fermion system. The occurrence of two low-temperature phase transitions in URu_2Si_2 emphasizes the relative instability of ordered ground states in heavy-fermion systems, and has led to extensive study of this compound in an attempt to understand such instabilities.

The technique of muon spin rotation and relaxation[4] (μSR) is well suited to investigate magnetic properties of solids on the microscopic level. The distribution of positive-muon (μ^+) Larmor precession frequencies directly reflects the distribution of local internal magnetic fields H_L at interstitial μ^+ sites in a remarkably sensitive manner: quasistatic[4,5] magnetic oredering of host moments as small as $10^{-3}\mu_B$ can yield measurable shifts and broadening of μ^+ frequency spectra. In addition, thermal fluctuations of H_L lead to equilibrium between the μ^+ polarization and its surroundings, with a characteristic spin-lattice relaxation time T_1.

The relative shift of a spin-probe (nuclear or muon) Larmor frequency due to electronic paramagnetism in metals is commonly called the Knight shift.[6] The isotropic Knight shift K_i is the average shift over crystal orientation, and can be obtained from the centroid of the spin-probe resonance spectrum in a polycrystalline specimen.[6] The anisotropic shift K_a broadens the resonance line, but does not shift its centroid unless there is preferential orientation of the crystallites in the sample. The same information on K_i and K_a is present in the time dependence of the freely precessing transverse spin-probe magnetic moment. An additional contribution to the line shape arises from any inhomogeneity in H_L.

For typical distances between μ^+ sites and local moments in magnetically dense materials the dipolar contribution H_d to H_L is of the order of 1 kOeμ_B^{-1}, i.e., a local moment of $1\mu_B$ gives rise to a dipolar field of order 1 kOe at a distance of a few Å. This dipolar interaction contributes to the width of the local field distribution but not to the isotropic Knight shift, since its orientational average vanishes. Any observed isotropic shift must arise from a hyperfine contribution H_{hf} to H_L; i.e., to nonzero unpaired electronic spin density at the spin-probe site. The hyperfine field can also be anisotropic, and thus contribute to the linewidth of a polycrystalline sample.

This paper describes the results of a μSR investigation of the phase transition at $T_N \simeq 17.5$ K in URu_2Si_2. We have found that the transition is indeed magnetic, since a sharp increase in the zero- and low-field μ^+ linewidth σ_μ is observed below T_N. The magnitude of the increase, however, is much smaller than would be expected from a moment of order $1\mu_B$ on the uranium sites. A qualitatively similar result was recently reported by Broholm *et al.*,[7] who used neutron Bragg scattering to investigate the phase transition. They found a simple AF structure, with a (100) modulation wave vector and an ordered U moment of $0.03\mu_B$.[8] But the μSR linewidth yields a moment $\sim 10^{-3}\mu_B$, some 30 times smaller even than that of Broholm *et al.*, if the μ^+ site is not assumed to possess any particular symmetry with respect to the AF structure. If this assumption is relaxed our results may be more nearly in accord with those of the neutron-scattering study. On the other hand, it is possible that the presence of the charged muon alters its local environment in such a manner as to suppress the already small magnetic moment. There is some evidence for moment suppression from μSR studies of another heavy-fermion antiferromagnet[9] U_2Zn_{17}, but not in the similar systems[9,10] UCu_5 and UCd_{11}.

The μ^+ isotropic Knight shift K_i was found to be very small and to correspond to an isotropic hyperfine field of order 100 Oeμ_B^{-1}. The μ^+ linewidth increases with field at a rate which is consistent with the calculated dipolar contribution. This increase is quite rapid; in 5 kOe the "paramagnetic" contribution to σ_μ below T_N is considerably larger than that due to AF ordering.

As discussed below, measured values of the spin-lattice relaxation time T_1 in URu_2Si_2 are much longer than the muon decay lifetime of 2.2 μs and, hence, cannot be measured accurately. This paper is therefore concerned primarily with the quasistatic component of H_L.

II. EXPERIMENTAL RESULTS

A polycrystalline specimen of URu_2Si_2 was arc melted and spark cut to a disk of approximately 25 mm diam $\times 6$ mm thickness. Specific-heat and superconducting quantum interference device (SQUID) susceptibility measurements were carried out on pieces of the same sample used for the μSR study. A Néel temperature $T_N = 17.7 \pm 0.3$ K was determined from the discontinuity in the specific heat, and the onset of the superconducting transition was found at ~ 1.3 K from ac susceptibility measurements.

μSR experiments were carried out at the Stopped Muon Channel of the Clinton P. Anderson Meson Physics Facility (LAMPF), Los Alamos. A standard time-differential μSR spectrometer was used, together with a cold-finger cryostat capable of temperatures between 3 and 300 K. Magnetic fields of up to 5 kOe were provided by a water-cooled Helmholtz-pair magnet.

A. Zero- and low-field linewidths

Representative time-differential muon depolarization functions $G_z(t)$ in zero applied field are shown in Fig. 1 for temperatures above [Fig. 1(a)] and below [Fig. 1(b)] the Néel temperature T_N. Values of Gaussian linewidths σ_μ were obtained by fitting such data to a Gaussian functional form

$$G_z(t) = G_z(0)\exp[-(\sigma_\mu t)^2/2] \; ;$$

the results of these fits are shown in Fig. 1 as solid curves. The squared relaxation rate σ_μ^2 is then essentially the second moment of the μ^+ frequency distribution even if the relaxation is not Gaussian, because the relaxation is slow ($\sigma_\mu \tau_\mu \ll 1$, where τ_μ is the muon decay lifetime) and

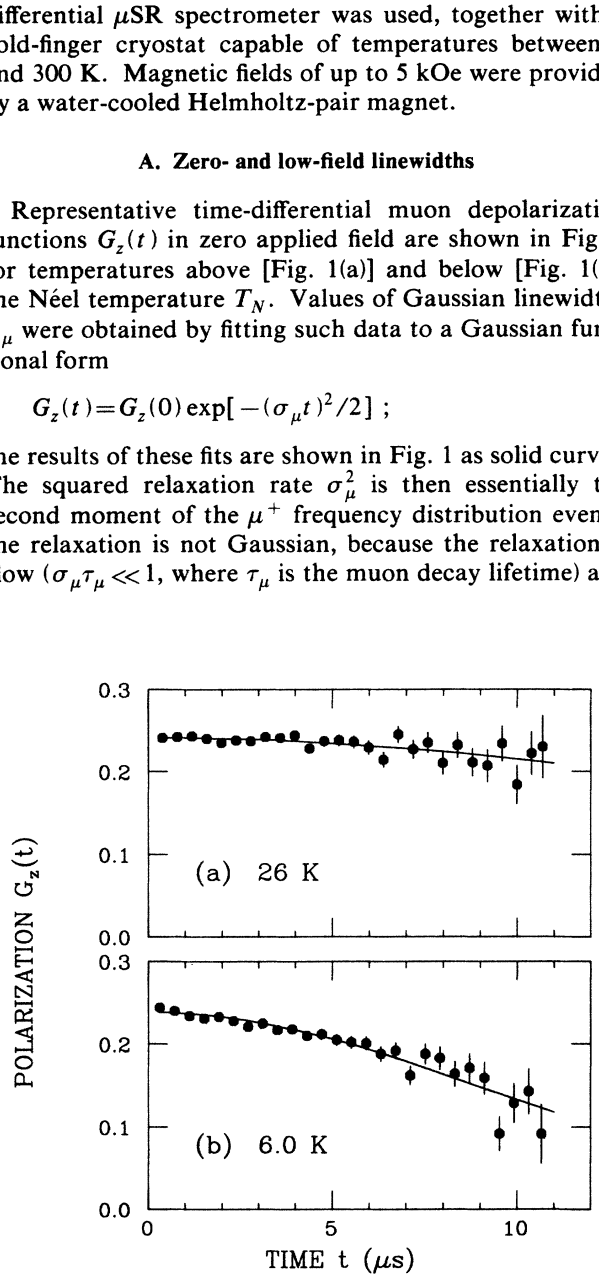

FIG. 1. Zero-field time-differential μSR spectra [polarization $G_z(t)$] above and below the Néel temperature $T_N = 17.7$ K in URU_2Si_2. (a) $T = 26$ K $> T_N$. (b) $T = 6.0$ K $< T_N$. Solid lines: fits of the Gaussian functional form $G_z(t) = G_z(0)\exp[-(\sigma_\mu t)^2/2]$ to the data. The increase of the linewidth σ_μ below T_N is evident.

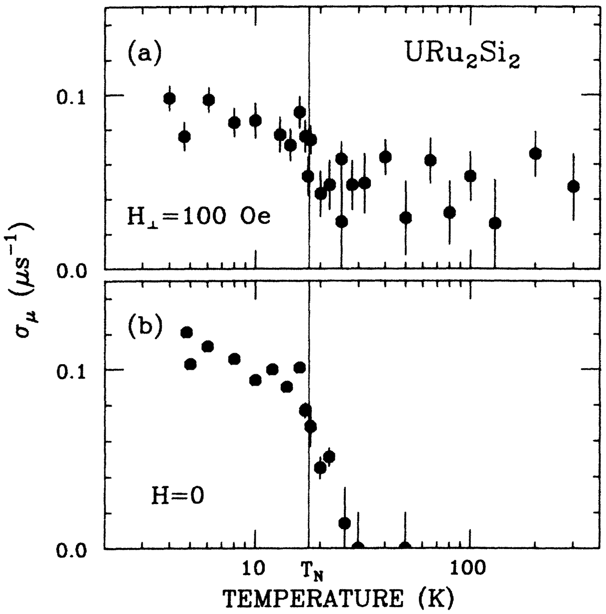

FIG. 2. Temperature dependence of the μ^+ Gaussian linewidth σ_μ above and below the AF ordering temperature T_N in Uru$_2$Si$_2$. (a) Transverse field $H_\perp = 100$ Oe. (b) Zero applied field. An increase of σ_μ below T_N is evident in both fields. The apparent vanishing of σ_μ for $H=0$, $T>T_N$, is an artifact of the fitting program.

the relaxation function is well approximated by the parabola

$$G_z(0)[1-(\sigma_\mu t)^2/2]$$

over observable times $t < 10$ μs. It is clear from the data of Fig. 1 that σ_μ increases markedly below T_N.

Figure 2 shows the observed temperature dependence of σ_μ in transverse applied fields $H_\perp$ of 0 and 100 Oe transverse to the initial μ^+ polarization. As can be seen, the linewidths above T_N are small and relatively uncertain: the accuracy of time-differential μSR deteriorates for small linewidths, because of the time limitation described above. In addition the fitting program for $H=0$, which must also fit for an unknown background term, biases the best-fit value of the linewidth toward zero as can be seen in Fig. 2(b). The average value

$$\sigma_\mu(100 \text{ Oe}) = 0.04 \pm 0.02 \ \mu\text{s}^{-1}$$

above T_N is compatible with the linewidth σ_{nuc} due to dipolar fields from ^{29}Si, ^{99}Ru, ^{101}Ru, and ^{238}U nuclear moments; the calculated values, shown in Table I, fall in this range for a number of candidate μ^+ sites. The linewidth data therefore do not determine the μ^+ site, in large part because the broadening is too weak.

In zero and low field σ_μ increases rapidly with decreasing temperature below T_N (Fig. 2), and in zero field attains a value of 0.12 ± 0.01 μs^{-1} as T approaches zero. In zero field the linewidth increase appears to begin at ~25

TABLE I. Calculated and measured nuclear and electronic (U ion) dipolar linewidths at candidate muon stopping sites in URu$_2$Si$_2$.

Site[a]	$\sigma_{\text{nuc}}(\mu\text{s}^{-1})$[b]	$\sigma_{\text{U}}(\mu\text{s}^{-1}\mu_B^{-1})$ AF ordering	Anisotropic shift
$(\frac{1}{4},\frac{1}{4},\frac{1}{4})$[c]	0.059	0	50
$(\frac{1}{2},\frac{1}{2},\frac{5}{16})$	0.028	157	136
$(0,\frac{1}{2},\frac{1}{8})$	0.072	70	57
$(\frac{1}{2},\frac{1}{2},0)$	0.048	134	35
$(\frac{1}{2},0,0)$	0.015	186	148
$(0,0,\frac{1}{4})$	0.044	69	62
$(\frac{1}{2},\frac{1}{4},0)$	0.023	156	72
$(\frac{1}{2},0,\frac{1}{16})$	0.023	142	113
$(\frac{1}{8},\frac{1}{2},\frac{3}{32})$	0.035	166	113
$(\frac{1}{4},\frac{1}{4},\frac{5}{32})$	0.040	76	33
$(\frac{1}{8},\frac{1}{8},\frac{7}{32})$	0.041	67	73
Experimental	0.04±0.02[d]	0.09±0.01[e]	56±3[f]

[a] Referred to the body-centered-tetragonal unit cell.

[b] From the Van Vleck second moment, due predominantly to ^{99}Ru and ^{101}Ru. Calculated assuming strong quadrupolar splitting (see Ref. 4).

[c] Symmetric site for AF structure of Broholm *et al.* (Ref. 7).

[d] Average for $T > T_N$.

[e] Linewidth (in μs^{-1}) for $H_\perp = 100$ Oe, $T \to 0$.

[f] From high-field linewidths at 6 and 25 K (Fig. 6).

K, which is considerably higher than T_N, but, as mentioned above, low values of σ_μ are not accurately determined by the fitting program and there is no reason to believe this higher temperature is significant. At 5 K $\sigma_\mu(H=0)=(1.22\pm0.17)\sigma_\mu(100\ \mathrm{Oe})$, which is consistent with the factor $\sqrt{2}$ expected from an ensemble of randomly oriented local fields.[11]

The structure obtained from neutron Bragg scattering[7] consists of an AF lattice of U moments, with sublattice points at (0,0,0) and $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ positions in the tetragonal unit cell and spins parallel to the *c* axis. The dipolar local field H_d due to U moments in this structure has been calculated for 11 distinct candidate μ^+ sites.[12] The rms powder-pattern μ^+ linewidths σ_{U} due to H_d are also shown in Table I. With the exception of the symmetric $(\frac{1}{4},\frac{1}{4},\frac{1}{4})$ site the values of H_d are $\gtrsim 10^3\ \mathrm{Oe}\,\mu_B^{-1}$, and the second moments of the corresponding powder-pattern spectra are $\gtrsim 70\ \mu\mathrm{s}^{-1}\mu_B^{-1}$. The observed low values of σ_μ below T_N are therefore compatible with very small ($\lesssim 10^{-3}\mu_B$) ordered moments. It should be noted that at the $(\frac{1}{4},\frac{1}{4},\frac{1}{4})$ site the μ^+ position is symmetric between the two AF sublattices, and the dipolar fields from the two sublattices exactly cancel. This is true for any U-μ^+ spin-spin interaction bilinear in the spin operators, which is a very general property.

Since the μ^+ site or sites are unknown, we are faced with two possibilities. Either (1) the quasistatic magnetic moment per U atom in the AF state is of the order of $10^{-3}\mu_B$, which is more than an order of magnitude smaller than the value $0.03\mu_B$ obtained from neutron scattering, or (2) the μ^+ sites are near (but not precisely at) the symmetric $(\frac{1}{4},\frac{1}{4},\frac{1}{4})$ points. In the former case the existence of a large nondipolar local field would reduce even further the upper bound on the U moment. In the latter case random crystal defects could distribute the equilibrium μ^+ sites over a range of positions somewhat away from the symmetric point. Unfortunately the present data do not permit a choice between these alternatives.

The amplitude $G_z(0)$ of the μSR signal, often referred to as the "asymmetry," decreased by only a few percent below T_N. This rules out the existence of any sizable rapidly relaxing μSR signal, which in turn means that few μ^+ sites experience a wide distribution of local fields. It was also determined that for $H_\perp = 100$ Oe the average μ^+ precession frequency did not change by more than $\sim 0.2\%$ below T_N; the broadening is very symmetric.

Spin-lattice relaxation rates $1/T_1$ were measured by applying a large (0.5–5 kOe) longitudinal decoupling field.[11] Small, but nonzero, values $1/T_1 \sim 0.03\ \mu\mathrm{s}^{-1}$ were measured between 5 and 16 K, but above T_N the rates were too small ($<0.01\ \mu\mathrm{s}^{-1}$) to be observed. This relaxation is much weaker than that observed in transverse applied field, which implies that the latter is due predominantly to quasistatic line broadening.

B. Knight shift

Figure 3 gives the observed temperature dependence of the isotropic μ^+ Knight shift K_i between 3 and 300 K. The μ^+ signal from a copper sample was used as a reference. The data have been corrected for demagnetizing and Lorentz fields, using the measured shape and bulk susceptibility χ of our sample, and for the μ^+ Knight shift of 60 ppm in pure Cu.

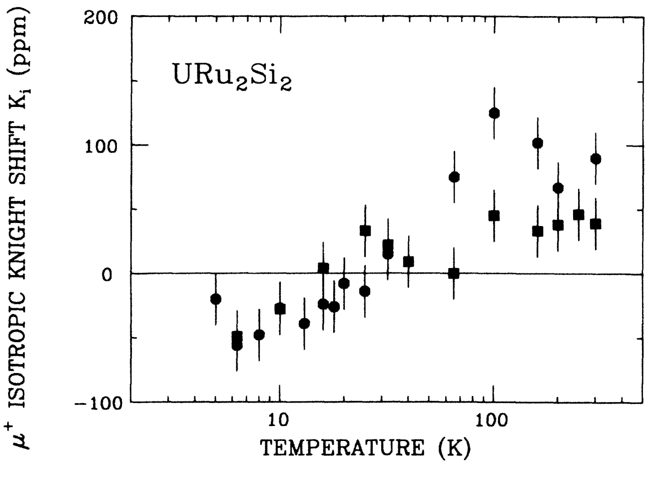

FIG. 3. Temperature dependence of the μ^+ isotropic Knight shift K_i in Uru$_2$Si$_2$. Applied field $H_\perp = 5.0$ kOe. Closed circles: original sample orientation. Closed squares: sample rotated 90° about an axis perpendicular to the applied field.

It can be seen from Fig. 3 that the shift is small ($\lesssim 100$ ppm). The temperature dependence of K_i does not track that of χ very well (see Figs. 4 and 5), and there is no noticeable feature at T_N. In addition, a small variation of K_i was observed above 35 K when the sample was rotated 90° about the disk axis. This indicates the presence of some preferred orientation of the crystallites in the sample which, in turn, implies that the measured K_i is not ex-

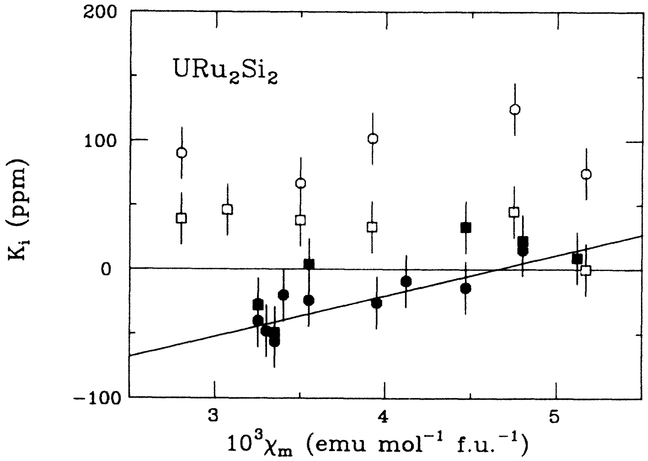

FIG. 4. Dependence of the μ^+ isotropic Knight shift K_i on bulk molar susceptibility χ_m, with temperature an implicit parameter, in URu$_2$Si$_2$ (see caption of Fig. 3 for symbol designations). Closed symbols: data below 35 K. The straight line is a fit to the latter data, and yields a hyperfine field of 160±20 Oeμ_B^{-1}. The unit f.u. in the abscissa label represents "formula unit."

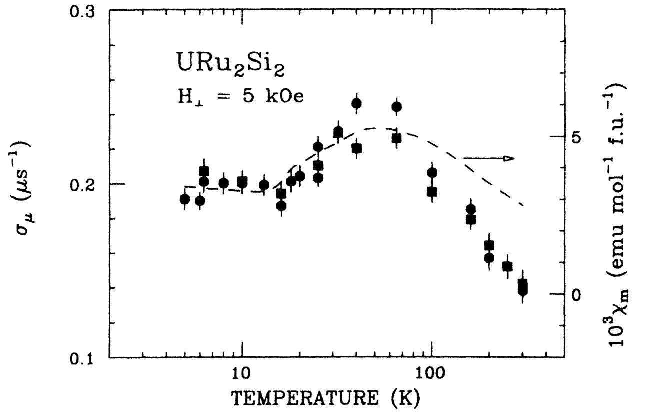

FIG. 5. Temperature dependence of the μ^+ Gaussian linewidth σ_μ in an applied transverse field $H_\perp = 5$ kOe (see caption of Fig. 3 for symbol designations). Dashed line: temperature dependence of bulk molar susceptibility χ_m, with scale and offset chosen to fit the linewidth data below ~50 K.

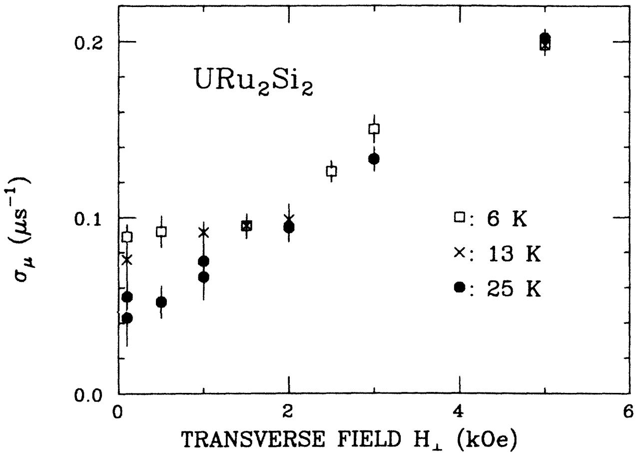

FIG. 6. Dependence of μ^+ Gaussian linewidth σ_μ on applied transverse field $H_\perp$ in URu_2Si_2 at representative temperatures above and below T_N.

actly the isotropic shift. The variation with angle is small, however, compared to the linewidth (Sec. II C below), and we assume without further proof that the anisotropic contribution to the measured K_i is a small fraction of the total anisotropy.

When plotted in the Clogston-Jaccarino form $K_i(\chi)$ (Fig. 4), with temperature an implicit variable, the data do not lie along a single-valued curve. For a given value of χ the shift is larger above ~35 K than below. For $T < 35$ K $K_i(\chi)$ is linear, and a rough value of the isotropic hyperfine field of 100–200 Oeμ_B^{-1} can be extracted. Typically μ^+ isotropic shifts in heavy-fermion compounds are considerably larger than 100 ppm at low temperatures.[13]

C. Linewidth

The temperature dependence of the μ^+ linewidth σ_μ at an applied transverse field $\mu_\perp = 5$ kOe is given in Fig. 5. In contrast to the isotropic Knight shift (Sec. II B above), $\sigma_\mu(T)$ follows the bulk susceptibility $\chi_m(T)$ rather well below ~50 K, and is therefore presumably a reflection of inhomogeneity or anisotropy in the μ^+ Knight shift. Again, there is no obvious feature in the temperature dependence other than a point of inflection near T_N. The decrease of σ_μ below χ_m for $T \gtrsim 50$ K may be due to the onset of muon diffusion.

The field dependence $\sigma_\mu(H_\perp)$ is given at several temperatures above and below T_N in Fig. 6. Above ~100 Oe σ_μ varies linearly with $H_\perp$ for $T > T_N$, whereas for $T < T_N$ a plateau is observed below 1.5 kOe. The linear slope of $\sigma_\mu(H_\perp)$ can be compared with the anisotropic shift expected from dipolar coupling to U moments, which are polarized by the applied field. The magnetization at U sites is given by $M = \chi H$ as usual, and the resulting rms dipolar field $H_d(\mathrm{U})$ after averaging over orientations has been calculated numerically at several candidate μ^+ sites. In the final column of Table I values of the rms linewidth σ_U due to $H_d(\mathrm{U})$ are given and compared with experimental values derived from the data of Fig. 6. It is clear that the observed linewidth is consistent with anisotropic dipolar broadening from U moments for several candidate μ^+ sites. Like the nuclear broadening above T_N, however, this electronic dipolar linewidth does not allow a unique μ^+ site determination.

III. DISCUSSION

A. Local-field distribution, zero and low applied field

The observed increase of σ_μ below T_N is strong evidence for magnetic ordering associated with this transition. The same is true of the onset of the AF Bragg neutron scattering observed by Broholm *et al.*,[7] which, in addition, gives the ordered AF structure. Neither result rules out a charge-density wave[2] or other structural modification, although a CDW with no accompanying spin-density variation is inconsistent with the data.[14]

The increase of σ_μ below T_N shown in Fig. 2 is qualitatively similar to Brillouin-function behavior of the sublattice magnetization, as was also observed[9] in U_2Zn_{17}. This behavior is reminiscent of a mean-field transition, and is consistent with the mean-field-like specific-heat discontinuity,[1,2] although the μSR data are not precise enough to determine the magnetization critical exponent β.

Experimental uncertainty also places a lower bound of ~5 K on the width of the transition at T_N. This is, however, considerably sharper than the onset of neutron Bragg scattering,[7] which persists to temperatures above 30 K.

The value of σ_μ for $T \ll T_N$ is much less than expected from typical μ^+-U atom dipolar fields and U moments of ~$0.03\mu_B$ in the AF structure of the neutron results. This could be due to a symmetric $(\frac{1}{4},\frac{1}{4},\frac{1}{4})$ μ^+ stopping site. Alternatively, the presence of a charged interstitial muon might locally depress the AF ordering tendency in this heavy-fermion system. Similar low values of ordered moment have previously been observed[9] in the heavy-fermion antiferromagnetic U_2Zn_{17}. Identification of the mechanism for such a depression remains work for the future.

At zero and low applied fields there is no evidence for a change of the asymmetry or (in nonzero field) the average μ^+ frequency of more than a few percent. These results suggest that no muon sites experience local fields much larger than those which contribute to the observed line. The fact that the mean local field does not change below T_N is consistent with dipolar anisotropic broadening as the origin of the observed linewidth increase, and further implies that the sample is a good polycrystal with little ($<10\%$) preferential orientation. Otherwise one would expect a frequency shift below T_N of the same order of magnitude as the linewidth increase.

B. Knight shifts and high-field linewidths

We noted above that the high-field μ^+ linewidth is consistent with anisotropic broadening from U-moment dipolar fields for several candidate μ^+ stopping sites (Table I). In addition, the absence of any sign of the transition at T_N in high field implies that the field-induced paramagnetic spin polarization dominates the μ^+ local field. The breakdown of the linear relation between K_i and χ_m (Fig. 5) and the reduction of σ_μ for temperatures $\gtrsim 50$ K (Fig. 6) are both consistent with motional narrowing due to the onset of muon diffusion at high temperatures.

We conclude from the low muon linewidth in the ordered state of URu_2Si_2 that either the μ^+ site is nearly symmetric, or that the interstitial muon locally suppresses the antiferromagnetic spin ordering. Such suppression may occur in more than one heavy-fermion antiferromagnet,[9] and therefore should be studied further in an attempt to understand its origin.

ACKNOWLEDGMENTS

We acknowledge, with thanks, useful discussions with Z. Fisk and G. M. Kalvius. This work was supported in part by National Science Foundation Grant No. DMR-8413730 and by the University of California, Riverside, Academic Senate Committee on Research, and was performed under the auspices of the U. S. Department of Energy.

---

[1] T. T. M. Palstra, A. A. Menovsky, J. van den Berg, A. J. Dirkmaat, P. H. Kes, G. J. Nieuwenhuys, and J. A. Mydosh, Phys. Rev. Lett. **55**, 2727 (1985).

[2] M. B. Maple, J. W. Chen, Y. Dalichaouch, T. Kohara, C. Rossel, M. S. Torikachvili, M. W. McElfresh, and J. D. Thompson, Phys. Rev. Lett. **56**, 185 (1986).

[3] W. Schlabitz, J. Baumann, B. Pollit, U. Rauchschwalbe, H. M. Mayer, U. Ahlheim, and C. D. Bredl, Z. Phys. B **62**, 171 (1986).

[4] For a review of the μSR technique see A. Schenck, *Muon Spin Rotation Spectroscopy* (Hilger, Bristol, 1985). A review of μSR in metals is given by E. Karlsson, Phys. Rep. **82**, 272 (1982).

[5] Strictly speaking H_L can vary in time and still yield well-defined Larmor frequencies, as long as the *quasistatic* condition (variation of H_L negligible over a time of the order of the inverse μ^+ linewidth) is fulfilled. The linewidth will, of course, reflect the distribution of Larmor frequencies in time.

[6] For a review of NMR Knight-shift theory and measurements, see G. C. Carter, L. H. Bennett, and D. J. Kahan, Progr. Mater. Sci. **20**, 1 (1977).

[7] C. Broholm, J. K. Kjems, W. J. L. Buyers, P. Matthews, T. T. M. Palstra, A. A. Menovsky, and J. A. Mydosh, Phys. Rev. Lett. **58**, 1467 (1987).

[8] Such a low moment could be a sign of a singlet U crystal-field ground state (Ref. 7) or, alternatively, of a form of itinerant heavy-fermion AF ordering.

[9] S. Barth, H. R. Ott, F. N. Gygax, A. Schenck, T. M. Rice, and Z. Fisk, Hyperfine Interact. **31**, 397 (1986).

[10] S. Barth, H. R. Ott, F. Hulliger, F. N. Gygax, A. Schenck, and T. M. Rice, Hyperfine Interact. **31**, 403 (1986).

[11] R. S. Hayano, Y. J. Uemura, J. Imazato, N. Nishida, T. Yamazaki, and R. Kubo, Phys. Rev. B **20**, 850 (1979).

[12] The U electronic structure is expected to be nearly atomic or tight-binding in nature, and the wave functions are therefore well localized. To this extent the approximation of point dipoles at U sites is justified.

[13] Y. J. Uemura, W. J. Kossler, B. Hitti, J. R. Kempton, H. E. Schone, X. H. Yu, C. E. Stronach, W. F. Lankford, D. R. Noakes, R. Keitel, M. Senba, J. H. Brewer, E. J. Ansaldo, Y. Oonuki, T. Komatsubara, G. Aeppli, E. Bucher, and J. E. Crow, Hyperfine Interact. **31**, 419 (1986).

[14] A structural transition could modify internuclear distances and, hence, the linewidth σ_{nuc} from the Van Vleck second moment (Table I). This is ruled out by x-ray measurements (Ref. 1). To obtain an increase of σ_{nuc} by the observed factor of 3 (Fig. 1) would require a decrease of lattice parameters by a factor of $\sim(3)^{1/3}=1.4$, whereas the experimental values at 4.2 and 294 K differ by no more than 1 part in 10^4.