Study of the 1-K Phase Transition in the Heavy-Electron Compound UCu_5 by Muon Spin Resonance and Neutron Scattering

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The 1-K phase transition in UCu_5, showing up in specific-heat data within the antiferromagnetic state below $T_N \cong 15$ K, was investigated. Neither the average internal fields seen by the μ^+ nor the magnetic and nuclear Bragg reflections in the neutron-diffraction data reflect the phase transition while the muon relaxation rates increase drastically below 1.2 K. These results are interpreted in terms of some additional small-moment magnetic order or spin-density-wave phenomenon, implying the coexistence of two rather independent electronic subsystems: one involving "heavy" electrons, associated with the weak magnetism, and another one associated with the "conventional" antiferromagnetic order.

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Low-temperature specific-heat measurements characterize the intermetallic compound UCu_5 as a heavy-electron system whose heavy-electron state, surprisingly, develops at temperatures well below the onset of antiferromagnetic order which is observed at $T_N \cong 15$ K.[1] Specific-heat data $c_p(T)$ have further shown that a second phase transition occurs at ~ 1 K, displaying hysteretic behavior but no latent heat.[1] The nature of this phase transition has not been identified yet. The absence of a latent heat in what looks like a first-order phase transition and a strongly increasing resistivity below 1.2 K (Ref. 1) are, however, reminiscent of similar problems arising in the transition between commensurate and incommensurate charge-density-wave (CDW) states.[2] The second phase transition, including the rise in resistivity, is only seen in high-quality UCu_5 samples.

Earlier neutron-diffraction measurements on UCu_5 allowed one to identify the antiferromagnetic structure of the U sublattice as consisting of ferromagnetically ordered (111) planes coupled antiferromagnetically with alternating moment directions along the body diagonal of this fcc compound possessing the $AuBe_5$ crystal structure.[3] The U moment was determined in this earlier study to be $\sim 0.9\mu_B$/U.[3] Previous muon-spin-resonance (μSR) studies on a polycrystalline UCu_5 sample of apparently lesser quality, which did not show the 1-K specific-heat anomaly, revealed two spontaneous precession signals below T_N with saturation frequencies of $\nu_1 = 19.75$ MHz and $\nu_2 = 13.65$ MHz, corresponding to internal fields of 0.146 and 0.100 T, respectively.[4] Both components and a third, nonprecessing one ($\nu \equiv 0$) displayed an essentially temperature-independent spin-relaxation rate of ~ 0.4 μs^{-1} from 30 mK up to 10 K.[4] No distinct features showed up around 1 K.

The present measurements, intended to probe into the nature of the 1-K phase transition, were performed on a high-quality polycrystalline sample, produced at LANL (Los Alamos) which showed the 1-K anomaly in the electrical resistivity. The neutron-scattering work was carried out on the multidetector powder diffractometer[5] DMC located at the reactor Saphir of the Paul Scherrer Institute (PSI) using a neutron wavelength $\lambda = 1.703$ Å selected by means of a vertically focusing Ge monochromator (311). The sample was contained in a Cu can mounted on the cold finger of an Oxford dilution refrigerator 200 NS. The μSR work was performed with the low-temperature μSR facility on the πM3 beam line of the PSI 600-MeV proton accelerator.

As an example of our results we show in Fig. 1(a) neutron-diffraction intensities for UCu_5 at 10 mK. In addition to the expected nuclear reflections from UCu_5 and Cu (sample container), we clearly observe antiferromagnetic superlattice peaks corresponding to a wave vector $\mathbf{k} = [\frac{1}{2}\ \frac{1}{2}\ \frac{1}{2}]$ implying a doubling of the chemical unit cell. The absence of the $(\frac{1}{2}\ \frac{1}{2}\ \frac{1}{2})$ peak near 12° indicates that the magnetic moments are predominantly

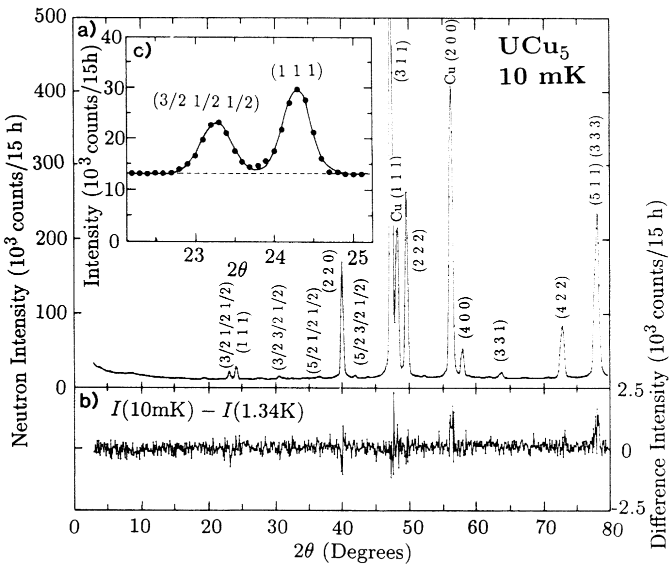

FIG. 1. (a) Neutron-diffraction pattern at $T = 10$ mK. The neutron wavelength is 1.703 Å. The solid lines are guides to the eye. (c) A blowup of the magnetic ($\frac{3}{2}$ $\frac{1}{2}$ $\frac{1}{2}$) reflection next to a nuclear peak. (b) Difference neutron diagram $I(10\text{ mK}) - I(1.34\text{ K})$.

directed along the [111] direction. These findings confirm the experimental results by Murasik *et al.*[3]

The temperature dependence of the antiferromagnetic order parameter indicates conventional behavior of the sublattice magnetization. In particular, there is no anomaly near 1 K. The difference scan in Fig. 1(b) shows that the intensity of the magnetic scattering does not change when the temperature increases from 10 mK to 1.34 K within the accuracy of our measurements, i.e., within $\pm 5\%$. The intensity of the nuclear Bragg peaks is constant to within 0.1%. By means of a standard Rietveld profile analysis we confirmed the known nuclear and magnetic structure and obtained for the moment a value $(1.55 \pm 0.05)\mu_B$/U, which is significantly larger than the value reported in Ref. 3. In the fitting procedure we varied the position of the Cu atoms at the positions (xxx), $x \simeq \frac{5}{8}$. We obtained for all temperatures (10 mK $< T <$ 25 K) $x = 0.623 \pm 0.001$.

In summary, the neutron-diffraction data indicate neither a structural nor a magnetic phase transition near 1 K. However, the ordered moment $\mu = (1.55 \pm 0.05)\mu_B$/U is significantly larger than the value quoted in Ref. 3. We do not understand the origin of the discrepancy. There is the possibility that μ depends rather strongly on the crystal quality. A strong influence of sample perfection on magnetic behavior is well known in other heavy-electron compounds.

Next we turn to the μSR results.[6] The zero-field measurements were performed from 0.4 to 1.95 K by scanning between these temperatures in order to detect possible hysteretic features. No such features were seen. Compared to previous results in Ref. 4 both similarities

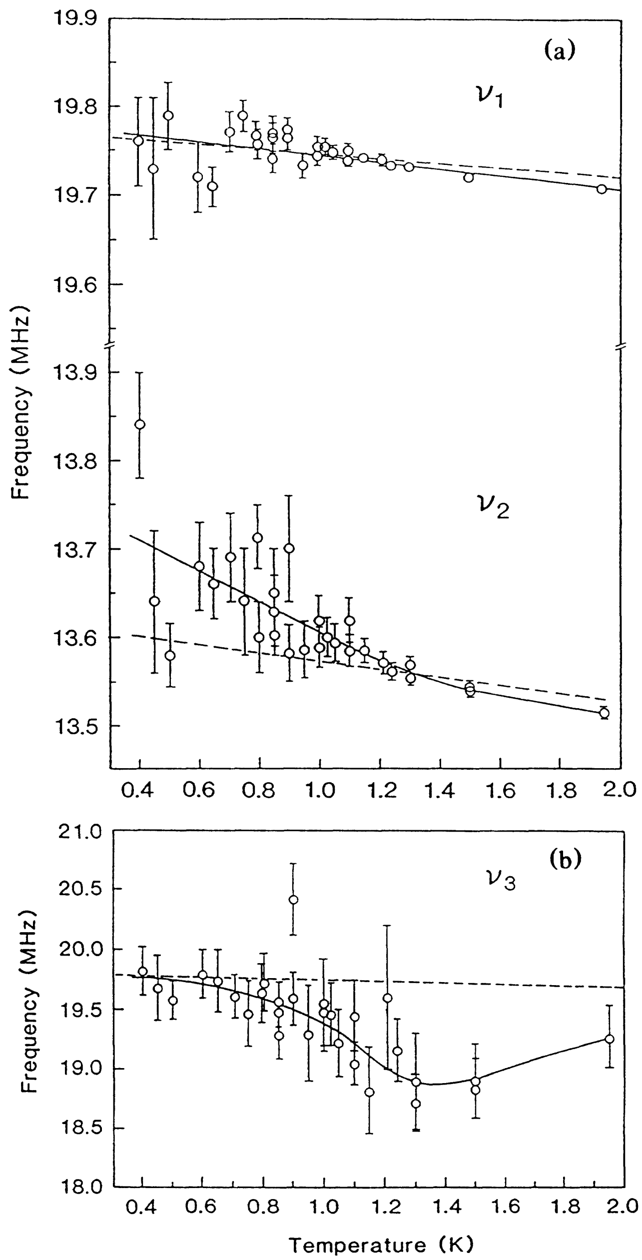

FIG. 2. Temperature dependence of the μ^+-spin precession frequencies (a) ν_1 and ν_2 and (b) ν_3. The dashed lines in (a) represent the earlier data of Ref. 4; the dashed line in (b) the present ν_1 values. The solid lines are guides to the eye.

and interesting differences have to be noted. Again the $\nu_1 = 19.75$ MHz, the $\nu_2 = 13.65$ MHz and the $\nu = 0$ component are observed. The temperature dependence of ν_1 and ν_2 are displayed in Fig. 2(a). Essentially the new data agree well with the earlier ones. There is a slight deviation between the new and the old ν_2 below ~ 1 K which is less than 0.7%. At present it is not clear whether it should be considered as significant in view of the limited statistical accuracy and in view of the absence of such a deviation in the better determined ν_1 component.

Figure 2(a) shows in particular that no discontinuous features appear around 1 K. This implies that the average fields at the μ^+ position, as manifested in each component, is not affected by the phase transition, at least not within the present level of accuracy. (For all components the μ^+ position is suggested to be at the center of the small Cu tetrahedron, to be discussed in a future paper.) This seems to be consistent with the neutron results, supporting the conclusion that the antiferromagnetic structure is not involved in the 1-K phase transition.

The analysis of the data leads to the identification of a third precessing component with $\nu_3 \simeq 19.5$ MHz, which is close to ν_1 [see Fig. 2(b)]. This component is, in fact, related to ν_1 as follows from the behavior of the signal amplitudes $A(\nu_i)$ of the various components ν_i: While $A(\nu_2) \simeq 2.3\%$ and $A_4(\nu=0) \simeq 3.8\%$ independent of temperature and $A(\nu_1) \simeq A(\nu_3) \simeq 5\%$ for $T \leq 1.15$ K, $A(\nu_1)$ and $A(\nu_3)$ show a complementary temperature dependence for $T > 1.15$ K such that $A(\nu_1) + A(\nu_3) \simeq 10\%$ = const. At 1.9 K we find $A(\nu_1) \simeq 6.5\%$ and $A(\nu_3) \simeq 3.5\%$. This remarkable feature seems to be a first indication of the 1-K phase transition. Details of the rather involved multicomponent analysis will be reported in a future full account of this work.

The most dramatic effect of the phase transition, however, is seen in the relaxation rate data, which are displayed in Fig. 3. In sharp contrast to the earlier μSR data,[4] the relaxation rates of the components with ν_1, ν_2, and $\nu_4=0$ rise drastically as the temperature is lowered through 1.15 K. In parallel the relaxation function changes in *appearance* from Gaussian (or Kubo-Toyabe type[6] for $\nu=0$) above 1.15 K to exponential below 1.15 K. The latter is not taken as evidence for dynamically induced relaxation, since there is still an unchanged time-independent fraction of μ^+ polarization. This fraction associated with those μ^+ spins which—in a polycrystalline sample—happen to be aligned along the internal field direction and hence do not precess. Fluctuating internal fields would also render this fraction time dependent. The increase in relaxation rate below 1.2 K must, therefore, reflect an increased inhomogeneous line broadening, i.e., an increased static field spread experienced by the μ^+ ensembles contributing to each component. The fact that use of an exponential relaxation function provides a better fit (in terms of χ^2) does not necessarily imply a Lorentzian field distribution. Rather on the basis of similar observations made, e.g., in the mixed phase of the high-T_c superconductors, it seems to signal the presence of a more complicated than Gaussian field distribution. But, as emphasized before, the average fields remain unchanged. The observed Gaussian relaxation rates above 1.15 K can be attributed to just the Cu nuclear dipole fields as indicated by second moment calculations.

The relaxation rates associated with ν_3 are much larger in the whole temperature range and must be related to the magnetic state. One possibility is that this component is associated with the magnetic domain structure, i.e., with regions in which the antiferromagnetic propagation vector **k** changes from, e.g., ⟨111⟩ to

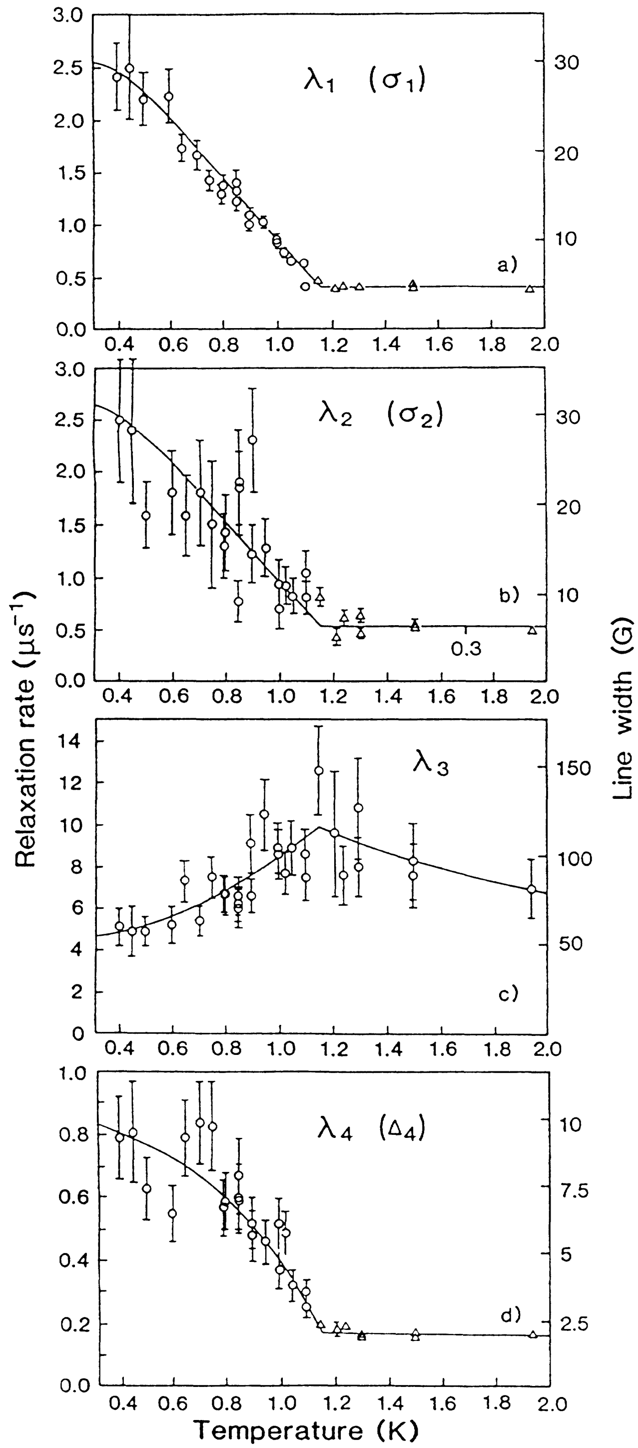

FIG. 3. Temperature dependence of the μ^+ relaxation rates associated with frequencies (a) ν_1, (b) ν_2, (c) ν_3, and (d) $\nu_4=0$. The solid lines are guides to the eye (see text for details).

⟨11−1⟩, etc. An obvious problem with this explanation is the near equality of $A(\nu_1)$ and $A(\nu_3)$ below 1.15 K. It would imply rather small domain sizes while the neutron data do not allow for domain sizes smaller than ~600 Å. Whatever the true explanation might be it must involve a mechanism which is coupled to the 1-K anomaly.

How can the present findings be understood? Obviously, the 1-K phase transition is not associated in a visible way with a change in magnetic structure. Neither does a structural phase transition seem to take place. One possibility to explain the increased field spread below 1.15 K is to postulate the evolution of small random static displacements of the μ^+ position, while keeping the average position unchanged. This would immediately cause a distribution in the dipolar fields at the μ^+ arising from the ordered U moments. Calculations show that random displacements of at least $\delta \simeq 0.07$ Å are required to cause the observed relaxation rates. In line with the discussion of the c_p anomaly (see introductory remarks) one may speculate that these random displacements are caused by the onset of a charge-density wave, producing similar shifts, e.g., in the Cu positions. Such shifts should have resulted in an effective Debye-Waller factor which, given the magnitude of the required shifts, should have been easily seen in the neutron-diffraction data. However, the diffraction data yield an estimate of only $\delta = 0 \pm 0.02$ Å. Hence the absence of any observable effects in the neutron data renders this explanation very unlikely.

The most probable cause for the increased field spread may therefore still be looked for in the framework of magnetism. Previous μSR and neutron studies in $U_{1-x}Th_xBe_{13}$,[7,8] UPt_3,[7,9] and URu_2Si_2[10,11] have provided evidence for the evolution of some sort of small-moment magnetic order at low temperatures, involving effective moments of the order of $(10^{-3}\text{-}10^{-2})\mu_B$. Although the small-moment ground states appear to be different in each case, it may not be unreasonable to assume that—*mutatis mutandis*—some similar phenomenon might also occur in UCu_5, albeit in coexistence with a well established antiferromagnetic order involving much larger effective moments. Placing the weak moments on the Cu sites, their magnitude would have to be of the order of $10^{-2}\mu_B$, i.e., too small to cause any visible effect in the present neutron-scattering data. On the other hand, when placing the weak moments on the U sites their magnitude would have to be about 10 times larger, which would be at the edge of being detectable in the neutron data. Alternatively, the increased field spread below 1.15 K may also be explained in terms of the evolution of a small-amplitude spin-density wave. The latter may indeed be associated with a more complicated field distribution rendering the *apparent* exponential relaxation quite reasonable.

The specific-heat and the μSR data together imply that the "weak magnetism" must be carried by the heavy quasiparticles. Since, on the other hand, the antiferromagnetic order is unaffected by the 1-K phase transition, one is led to speculate that the heavy electrons in this compound form a quasi-independent subsystem which settles into a peculiar ground state below 1.15 K, possessing a perhaps random but static order of very small effective moments or being associated with a static spin-density wave. In this picture the more or less conventional antiferromagnetism of the U-5f moments below 15 K is unrelated to the heavy-electron subsystem. In conclusion, we conjecture that the occurrence of two rather independent types of electron states might be the cause of other anomalous features seen in heavy-electron compounds.

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