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Observation of high-temperature spin fluctuations in UBe_{13} by nuclear-spin relaxation

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We report the extension of measurements of the ^{9}Be spin-lattice relaxation rate in UBe_{13} from 300 up to 1000 K. They reflect the fluctuating local magnetic field at the site of the ^{9}Be nuclei. Above about 100 K, there appears a new contribution in addition to the rate observed at lower temperatures. The high temperature contribution is modeled in terms of the thermal excitation of low-lying states of the U^{3+} ion along lines used by Felton *et al.* to explain their measurements of the specific heat. Our results indicate a splitting of 200 ± 20 K between the ground and excited states of the system, and a lifetime of 8×10^{-14} s for the excited states (width $= 95$ K).

I. INTRODUCTION

The attempt to understand the mechanism that produces "heavy fermions" in actinide and rare-earth intermetallic compounds and alloys has been one of the most actively pursued challenges in condensed-matter physics over the past few years. An important key to this understanding is their magnetic properties. One useful probe of the magnetic behavior is nuclear magnetic resonance (NMR), as it constitutes a *microscopic* probe of both the static and the dynamic behavior of the local magnetic field. In particular, it is known from its large heavy fermion enhancement[1,2] and its behavior in the superconducting state[3] that the ^{9}Be resonance is sensitive measure of the magnetic properties of the heavy fermions in UBe_{13}.

In this paper we describe measurements of the ^{9}Be nuclear spin lattice relaxation rate $1/T_1$ in UBe_{13} at high temperatures and analyze them in terms of a simple phenomenological model. The major result is that a new contribution to $1/T_1$ appears above 100 K. From the magnitude and temperature dependence of $1/T_1$, we infer the existence of low-lying excited states of the system, which are split from the ground state by an energy $\Delta E \approx 200(\pm 20)$ K and have a lifetime $\tau \approx 8\times10^{-14}$ s (i.e., a width ≈ 95 K). These results are in substantial agreement with related observations based upon specific-heat measurements,[4] and show in addition the rapidly fluctuating magnetic moment associated with the states.

II. EXPERIMENTAL DETAILS AND RESULTS

The sample was taken from a nominally pure arc-melted polycrystalline ingot, which was ground to a particle size ≈ 0.2 mm and sealed under He gas in a quartz tube. Standard pulsed NMR methods and apparatus were used for the measurements. The furnace used to obtain the elevated temperatures follows designs described elsewhere.[5] Temperature errors were on the order of 10 K, and uncertainties in the value of T_1 at high temperature approximated $\pm 10\%$.

A graph of $1/T_1$ as a function of T over the range 1–1000 K is shown in Fig. 1. The crosses and $\times$'s refer to measurements made at 9.0 and 18.5 MHz, respectively. Some of the low-temperature data have been published earlier.[6] The solid line is a fit to the data of the model to be discussed below, and the dashed line is a continuation of the trend established by the lower temperature data. From Fig. 1 it can be seen that the data below 40 K follow the solid curve, and that if it were to be extended to higher temperatures without knowledge of the higher-T data, the dashed line would be followed.

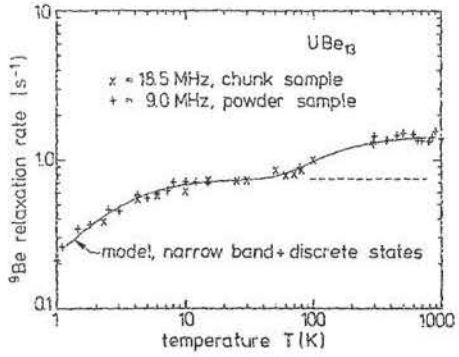

FIG. 1. ^{9}Be relaxation rate in UBe_{13} as a function of temperature. The solid line is the model developed in the text and the dashed line is a continuation of the low temperature behavior. The difference between the solid line and the dashed line at high temperature is attributed to thermal excitation of states 200 K above the ground state and whose spin correlation time is 8×10^{-14} s.

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The interpretation presented below identifies the low-T solid line and its dashed continuation as relaxation of the ^{9}Be nuclei by itinerant electrons, and treats the high-T rise of the solid line as an additional process (called the "additional relaxation") that is thermally excited above 100 K.

III. DISCUSSION

In order to discuss our result, we use a formulation of nuclear relaxation that can be applied to both itinerant and localized electrons. When nuclear magnetic moments are relaxed in a metal by electrons, $1/T_1$ can be written as[7,8]

$$\frac{1}{T_1} \approx \gamma_n^2 \langle \delta H^2 \rangle n_{\text{eff}} J(\omega) \approx \gamma_n^2 \langle \delta H^2 \rangle n_{\text{eff}} \tau, \tag{1}$$

where γ_n is the nuclear gyromagnetic ratio, $\langle \delta H^2 \rangle$ is average of the square of the amplitude of the fluctuating magnetic field at the nucleus associated with one electron, n_{eff} is the (effective) number of electrons (normalized to the unit cell) that contribute to $\langle \delta H^2 \rangle$, $J(\omega) \approx \tau$ is the spectral density of the fluctuations at the NMR transition frequency ω, and τ is the correlation time for the fluctuations. In Eq. (1), we assume the short correlation time limit $\omega\tau \ll 1$, as discussed below. When dealing with itinerant conduction electrons, $n_{\text{eff}} \approx k_B T/\Delta$ and $\tau \approx \hbar/\Delta$ (k_B is Boltzmann's constant; $\hbar$ is Planck's constant), and the usual Korringa relaxation rate is obtained.[7,8] For direct relaxation by localized electrons, n_{eff} is the local moment concentration and τ is the lifetime for the z component of the electron spin. The solid line below 40 K and the dashed line extension is the temperature dependence that one calculates using the model of Korringa relaxation, but in a narrow, Lorentzian-shaped band with a half width at half maximum $\Delta = 7.5$ K. The implications of this result will be discussed in a different publication; it is introduced here only to establish the *extra* relaxation that appears above 100 K.

We use the following phenomenological model for the additional relaxation. For the sake of concreteness, the same microscopic picture used to explain the specific heat[4] is adopted: The ground state of the U^{3+} ion, which would otherwise be degenerate, is split by the crystal field into a ground-state doublet (Γ_6) and a low-lying excited quartet (Γ_8). At low T only the Γ_6 states are occupied. As T is increased, there is a thermal population of the Γ_8 states. They have a different $\langle \delta H^2 \rangle$ (and perhaps τ) from the ground state, so that as they become populated, $1/T_1$ changes accordingly. This model is put on a quantitative basis by assuming that there is a splitting ΔE between the ground and first excited states of the system and that the states obey Boltzmann statistics. Then the relaxation rate is a linear combination of the rate R_g associated with the group of ground states and the rate R_e associated with the group of excited states:

$$1/T_1 = R_g P_g + R_e P_e, \tag{2}$$

where P_g and P_e are the probability that the system occupies the ground or the excited states respectively. These probabilities are

$$P_g = \frac{1}{1 + 2e^{-\Delta E/k_B T}}, \quad P_e = \frac{2e^{-\Delta E/k_B T}}{1 + 2e^{-\Delta E/k_B T}}. \tag{3}$$

The solid line on Fig. 1 is then obtained with the assignments $R_g = 0.75(\pm 0.05)\ \text{s}^{-1}$, $R_g = 1.88(\pm 0.18)\ \text{s}^{-1}$, and $\Delta E/k_B = 200(\pm 20)$ K.

The model just described provides a good fit to the data, and yields a value $\Delta E/k_B$ that is quite close to the one (180 K) obtained from the specific heat.[4] In addition, from the value of R_e we can estimate τ for the excited states. By using the theory of direct relaxation of a nucleus by a localized electron spin via dipolar coupling[9] we estimate

$$1/T_1 \approx (0.8/r^6)\gamma_n^2 \mu_e^2 \tau, \tag{4}$$

where μ_e is the fluctuating moment associated with the excited states, r is the typical distance from a localized excited state to the Be nucleus, and we have included the fact that each Be nucleus has two near-neighbor U ions. If we use the values $r = 2.8$ Å (a typical U-Be spacing) and $\mu_e = 3.4$ Bohr magnetons (from the high-T susceptibility[10]), Eq. (4) gives $\tau = 8.0 \times 10^{-14}$ s, or $\hbar/k_B\tau = 95$ K. The picture that then emerges from this interpretation is that there is a set of 4 excited states (such as a $U^{3+}\Gamma_8$) 200 K above a doublet ground state (such as a $U^{3+}\Gamma_6$) and the excited state system has a lifetime of 8×10^{-14} s, which corresponds to a width of 95 K. The smallness of τ justifies the use of the short correlation time limit in Eqs. (1) and (4).

The picture obtained from this experiment agrees closely with what was inferred from the specific-heat measurements of Felton *et al.*[4] The main difference compared to their work is that they introduced an additional crystal field splitting (two levels 115 K above the ground state and two levels 270 K above it for an excited state splitting of 155 K) of the excited states to fit the observed width of their Schottky anomaly. On the basis of our work, we identify that width as a lifetime broadening of the levels. The energy scale for the excited state observed in our experiment is also the same as that of a feature seen (but not identified) in the inelastic scattering of neutrons by UBe_{13}.[11] For a further discussion of the neutron results, see Felton *et al.*[4]

There are several aspects of the model we have used that require comment. Implicit in the treatment is the notion that the additional relaxation at high T is due to *orientational* relaxation of a local moment. The microscopic physical situation could as well be dominated by transitions *between* the electronic levels, or a combination of the two. At the present level of understanding, we cannot distinguish among these cases on the basis of the nuclear relaxation rate; all of them would generate a fluctuating magnetic field that would relax the nuclei, and it is reasonable to expect that similar values of ΔE and τ would be obtained. More detailed microscopic calculations are needed to improve this aspect of the interpretation.

A second point is related to the choice of the Γ_6 doublet and Γ_8 quartet suggested by Felton *et al.*[4] The temperature dependence obtained with our nuclear relaxation model is not sensitive to the total number of degenerate states, but rather to the ratio of the degeneracy of the excited state to that of the ground state. We have used a ratio of 2. A similar model in which the ratio is 1 fits the data almost as well and gives similar parameters (we did not try other ratios). From this observation we conclude that the model used is consis-

tent with our experimental results, but that it is not uniquely so.

The final point to be made is that the picture of a purely localized set of spins being responsible for the relaxation is probably too simplified to apply to the real physical situation in UBe_{13}. We expect, instead, that ΔE represents the difference in energy between a many-body ground state and a many-body excited state (or states) for which localized states of the U ion $5f$ electrons play a major role, but that the states responsible for the physical properties cannot be described simply in terms of fully localized ionic states.

IV. CONCLUSIONS

We have reported high-temperature measurements of the ^{9}Be relaxation rate in the heavy fermion intermetallic compound UBe_{13}. An additional relaxation component is observed above 100 K. It is interpreted in terms of dipolar coupling to a localized U^{3+} moment with a doublet ground state and a quartet first excited state. The deduced splitting between the ground and excited states is $200(\pm 20)$ K. The correlation time for the excited states is 8×10^{-14} s (width $= 95$ K).

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