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Muon spin relaxation in $ErRh_4B_4$

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Data on zero-field μSR in $ErRh_4B_4$ are presented for temperatures from 4 to 300 K. Due to the large erbium moment, most of the muons have a relaxation time of less than 10 ns for temperatures below about 50 K. The data have been analyzed in terms of two distinct muon sites in the material. The deduced erbium relative fluctuation rate versus temperature is related to the crystalline electric field levels of erbium in the $RERh_4B_4$ structure.

INTRODUCTION

$ErRh_4B_4$, one of the isostructural $RERh_4B_4$ (RE = rare earth) series of compounds,[1] is a re-entrant magnetic superconductor. As temperature T is lowered through 8.7 K, it becomes superconducting, but as T continues to be lowered through 0.9 K, the superconducting state is destroyed by the onset of ferromagnetic ordering. Thus there is a competition between superconducting and magnetic ordering interactions in this material.

The magnetic moment dynamics in the ordered states and in the paramagnetic state above both transition temperatures should be affected in interesting ways by the competition between the interactions. There is some indication of this in ^{11}B NMR of $(Y,Er)Rh_4B_4$,[2] but these effects may be complicated (perhaps even hidden) by crystalline electric field (CEF) effects in these materials.[2,3] Muon spin relaxation (μSR) is another technique that can sense internal magnetic field dynamics, and some μSR measurements on $HoRh_4B_4$,[4] and $SmRh_4B_4$,[5] have already been reported.

EXPERIMENT AND RESULTS

The $ErRh_4B_4$ sample was prepared by arc melting and annealing in a manner typical for these materials.[1] Powder x-ray diffraction showed evidence of a small amount of RhB, plus some weak unidentified lines. Impurity phases occupy about 5% of the sample. The midpoint of the superconducting transition of a powdered portion of the ingot in ac susceptibility was 8.6 K. The zero-field (ZF) μSR technique has been described in Ref. 6. The present μSR data were obtained using the M20 "surface" muon beam at TRIUMF, with the apparatus described in Ref. 7. The sample was mounted in a helium-flow cryostat in the center of a set of trim coils that allowed the field at the sample to be set to 0.0 ± 0.1 Oe. The asymmetry $[A(t)]$ spectra, i.e., the experimental spin relaxation functions, discussed below were obtained by subtracting separately measured background rates from each of the backward (B) and forward (F) counter histograms, and then calculating

$$A(t) = [B(t) - F(t)]/[B(t) + F(t)]. \quad (1)$$

Figure 1 shows ZF μSR $A(t)$ spectra of $ErRh_4B_4$ at a selection of temperatures. The muon polarization relaxes rapidly, but not as a simple exponential decay. The relaxation becomes faster as T decreases, until below 50 K it becomes so fast that most of the signal relaxes away between the stopping of the muon in the target (time = 0) and the start of the first good data bin (about 8 ns later in our present apparatus). As T decreases toward the region of the transitions, only a smaller-amplitude signal remains, with little T dependence.

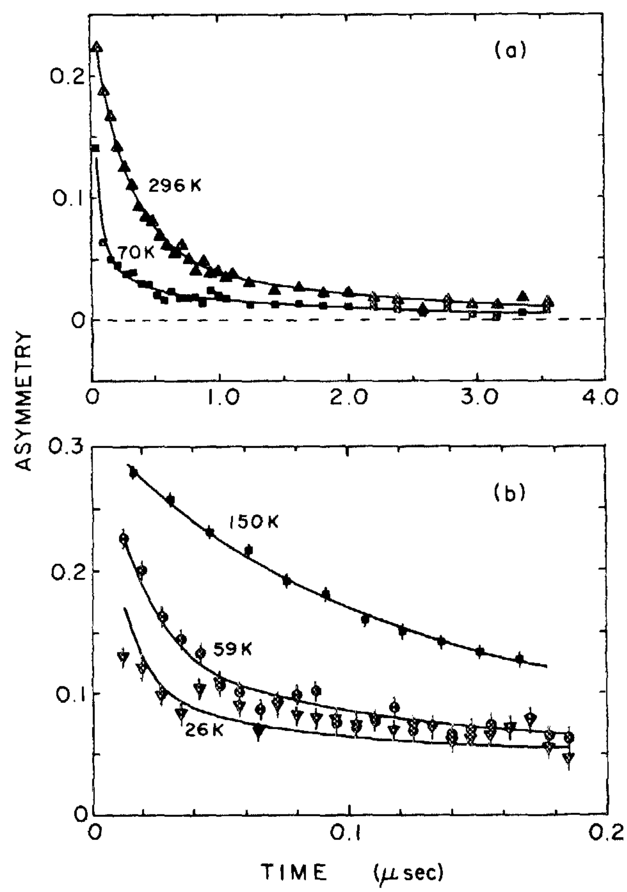

FIG. 1. ZF μSR asymmetry spectra of $ErRh_4B_4$ at a selection of temperatures: (a) over two muon lifetimes and (b) at early times and best time resolution, showing the loss of asymmetry below 60 K. Solid lines are the 3-site least-squares fits discussed in the text.

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No dramatic effect is seen in going through the superconducting transition temperature.

The interactions that cause relaxation of the muon polarization in conductors containing magnetic moments should be representable as (primarily) a distribution of effective magnetic fields at the muon sites, each muon spin precessing about the field at its site. In general, the field at a particular muon will fluctuate randomly at some average rate. This fluctuation may be due to the muon hopping from site to site in the material, or to the fluctuation of the magnetic moments causing the local field at the muon site. This kind of interaction in zero applied field results in the "dynamic Kubo–Toyabe" relaxation function.[6] In polycrystalline samples with ordered lattices of magnetic moments, the local field distribution is normally taken to be isotropic and Gaussian, and the field is assumed to have zero self-correlation across a fluctuation.

To assess the effect of ^{11}B moments on the muon spins in the $RERh_4B_4$ compounds, independent of rare earth atomic moments, we have taken ZF μSR data of YRh_4B_4.[5] There we observe the static Kubo–Toyabe relaxation function for all temperatures below 200 K, and deduce an rms field at the muon, generated by static ^{11}B nuclear moments, of less than 10 G. The static nature of the local field also indicates that the muons are not hopping (hop rate $<0.2\ \mu s^{-1}$) at temperatures below 200 K. (The hop rate at 300 K is of order $2\ \mu s^{-1}$.) Similar results[4] in $LuRh_4B_4$ indicate that in all members of the series muons do not hop below about 200 K. The muon relaxation in $ErRh_4B_4$ is so much faster than this that the local field at the muon must be completely dominated by the contribution from the erbium ions.

The Er^{3+} moment ($10.0\ \mu_B$ free ion), besides being orders of magnitude larger than the ^{11}B nuclear moment, must be expected to be fluctuating (on the time scale observable by μSR) in the paramagnetic state. The observed spectra's monotonic decay suggests that in the dynamic Kubo–Toyabe description, we are in the fast fluctuation regime. In this regime, the relaxation (for a single type of muon site) reverts to simple exponential, with decay rate

$$\lambda = 2\Delta^2/\nu\,, \tag{2}$$

where Δ is the rms value of a component of the field (stated as the corresponding Larmor precession rate) and ν is the average fluctuation rate. In this case, one can only place lower limits on Δ and ν individually.

Simple exponential relaxation does not fit the data, except perhaps near room temperature. The sum of two exponential signals can fit the data at all temperatures measured. In the temperature range in which both exponentials are well resolved (50–160 K), the ratio of the two fit relaxation rates (λ_1/λ_2) was roughly independent of temperature (with a value near 12). The amplitude of the weaker (slow relaxing) signal was about one quarter of the stronger one, too large to be from impurity phases.

Two distinct muon sites in $ErRh_4B_4$ must, therefore, be considered. In the Kubo–Toyabe description, two sites are likely to differ in the size of the average local field (Δ_1,Δ_2), which will be governed by the distance to the nearest erbium. The sites are unlikely to differ in field-fluctuation rate ν, which is a property of the erbium ensemble. When $\nu \gg$ both Δs, then, the muon spin relaxation should be a sum of two exponentials with λ_1/λ_2 equal to Δ_1^2/Δ_2^2, which ratio should not change (to first order) with temperature, even when the erbium moment decreases somewhat at low temperature.[3] This model, with the ratio of λs independent of temperature, fits the data well at 55 K and above. To get acceptable fits below 50 K, however, it was necessary to add a third, slowly relaxing ($\lambda \sim 0.5\ \mu s^{-1}$), T-independent background signal to the model (at all temperatures), which we attribute to muons that missed the (relatively small) sample and stopped in the sample holder and surrounding material.

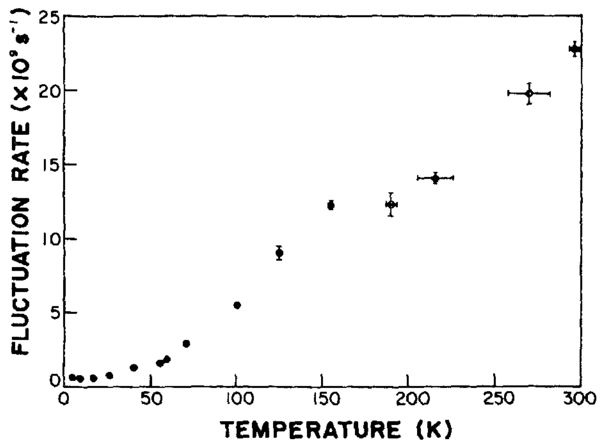

FIG. 2. Fluctuation rate of the local field at the muon in $ErRh_4B_4$ deduced from 3-site fits to the ZF μSR data assuming high-field $\Delta_1 = 200\ \mu s^{-1}$ and low field Δ_2 near $65\ \mu s^{-1}$.

The model thus involves three exponentially relaxing signals, the parameters of which are naturally highly correlated in fitting, so that interpretation must be done carefully. The assumption that the fast initial relaxation, already hard to resolve at 55 K, becomes faster still at lower T implies that the high-field site has an rms local field of at least 1 kOe and a fluctuation rate at 55 K of at least $5\times 10^8\ s^{-1}$. The muon relaxation slows down as the field fluctuation rate rises with temperature, as shown in Fig. 2.

The least squares fits imply the ratio of the low field site Δ_2 to the high field site Δ_1 to be in the range of 0.32 to 0.45. The initial asymmetry of the high-field signal is about 0.23, while that of the low field signal is about 0.04, and that of the "background" signal about 0.03, for a total initial asymmetry of 0.30, typical for this apparatus.

DISCUSSION

The relative asymmetries of the signals indicate that a muon is five to six times more likely to stop in a high-field site than a low field one in $ErRh_4B_4$. While the crystal structure is complicated enough to easily support more than one type of muon site, such a disparity in stopping probability is surprising. Detailed consideration of possible muon sites in this material is now in progress.

The local field fluctuation rate should be proportional to the erbium moment fluctuation rate. A rate near the lower bound set above, with the temperature dependence of Fig. 2, is roughly consistent with a T-independent rate at low temperatures (but still above 5 K) due to the RKKY exchange interaction,[8] plus T dependence due to spin-lattice fluctuation[9] among the CEF-split states of the erbium ion, which are distributed up to about 120 K.[3] The break in the smooth rise of the fluctuation rate between 150 and 200 K apparent in Fig. 2, however, is difficult to explain in terms of rare-earth fluctuations. It may instead indicate the onset of muon hopping in $ErRh_4B_4$ at temperatures slightly lower than in YRh_4B_4. If muons begin to hop from one type of site to another, then the present model, which does not allow for this possibility, will begin to produce incorrect parameter values. In this case, the points of Fig. 2 for $T > 150$ K only indicate the general trend. Results of detailed modelling of the erbium fluctuation rate in this material will be the subject of a future publication.

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