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μSR studies of the hexaboride system $Eu_xCa_{1-x}B_6$

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Abstract

We report the results of transverse field (TF) and zero-field (ZF) μSR measurements on the hexaboride system $Eu_xCa_{1-x}B_6$. EuB_6 is a semimetallic ferromagnet that magnetically orders via two transitions at $T_m \approx 15$ K and $T_c \approx 12$ K, with colossal magnetoresistance accompanying the higher temperature transition. New TF μSR measurements on EuB_6 allow us to follow the temperature evolution of the local magnetic field distribution through the two magnetic transitions. Substitution of Ca for Eu dilutes the magnetic sublattice, causing a substantial suppression of the transition temperature, with the transition completely removed when the doping level approaches the three-dimensional site percolation limit, $x_p = 0.31$. ZF experiments on doped samples, $0.35 \leqslant x \leqslant 1$, enable a sensitive local-probe exploration of the order and dynamics across the phase diagram.
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Europium hexaboride has attracted recent interest because it exhibits colossal magnetoresistance (CMR) [1] and it has been suggested that its semiconductor–semimetal transition results from the overlap of magnetic polarons [2]. EuB_6 crystallises into a simple cubic structure (space group Pm3m) with divalent Eu ions ($^8S_{7/2}$) at the corners of the unit cell and B_6-octahedra at the body-centred positions, and is a ferromagnet at low temperatures [1]. Specific heat and magnetization measurements reveal that this state is reached via two distinct transitions at $T_m = 15.5$ K and $T_c = 12.6$ K [2,3]. The magnetic ordering is accompanied by a sharp drop in the resistivity which is strongly field dependent [4] and gives rise to a large negative magnetoresistance [2]. This transition from a semiconductor at high temperatures to a semimetal [5–7] at low temperatures is reminiscent of the metal–insulator transition seen in manganites exhibiting CMR. Detailed measurements of resistivity and magnetization [2] show that this transition is associated with T_m. It is thought that magnetic polarons could be responsible for this behaviour, with the upper magnetic transition and drop in resistivity caused when the bound carriers overlap and percolate, and the lower transition caused by a true transition to a bulk ferromagnetic state [2]. This explanation is supported by the observation of polaronic features below ~30 K in Raman-scattering spectra [8], and is consistent with results of a zero field muon-spin rotation study that revealed the co-existence of a Gaussian signal with an oscillatory signal for $T_m \lesssim T \lesssim T_c$. Upon substituting Ca for Eu, T_c decreases substantially with increasing dilution of the magnetic sublattice, and leads to significant changes of the electronic properties across the $Eu_xCa_{1-x}B_6$ series [9,10].

Transverse field muon-spin rotation measurements were performed on a polycrystalline sample of EuB_6 (the same sample used for the zero-field study reported in Ref. [11]) using the GPS spectrometer at the Swiss Muon Source, Paul Scherrer Institute. An applied field of 0.5 T (corresponding to a muon oscillation frequency of 67.75 MHz) was used. To describe the field distribution experienced by the muon, the data were transformed into the frequency domain, using both a cosine Fourier transform and a

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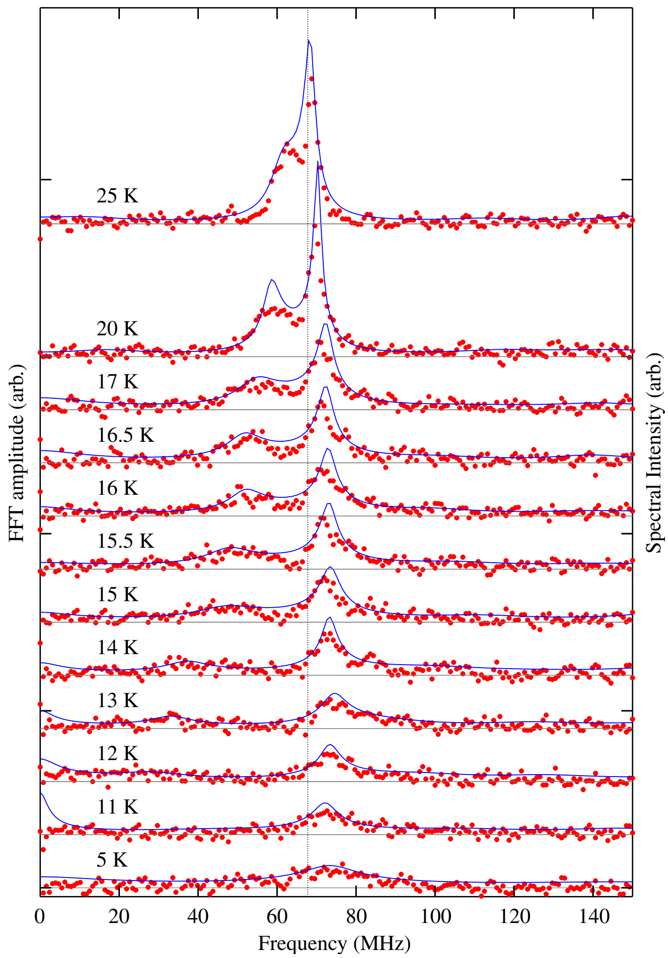

Fig. 1. Cosine Fourier transform (dots) and maximum entropy all-pole power spectrum (solid lines) of typical TF μSR spectra for EuB_6. The vertical dotted line shows the expected position of a diamagnetic response to the 0.5 T applied field.

maximum entropy all pole estimation of the power spectrum. Typical transformation results are displayed in Fig. 1.

The two estimation techniques seem to give consistent results, and demonstrate that there are two peaks in the field distribution: a narrower one with a frequency higher than the expected diamagnetic frequency, and a broader one with a lower frequency that shifts significantly to lower frequency as the temperature is lowered, until it is no longer visible near 11 K. The changes in the structure and positions of these peaks were parameterized by fitting in the time-domain to a model with two relaxing frequencies:

$$A(t) = A_1 \exp(-\lambda_1 t)\cos(2\pi v_1 t + \phi_1) + A_2 \exp(-\lambda_2 t)\cos(2\pi v_2 t + \phi_2). \quad (1)$$

All parameters were allowed to vary freely during fitting, and the resulting oscillation frequencies, $v_{1,2}$, and relaxation rates, $\lambda_{1,2}$, are shown in Fig. 2. The trends seen by eye in Fig. 1 are captured well by these fits. The higher frequency peak is seen to shift upwards on cooling until it saturates at around 10 K, possibly following the magnetic susceptibility of the sample. The lower peak moves down until it is no longer observed near 11 K. The relaxation rate plot shows the two peaks broadening on cooling. As the width of the field distribution becomes comparable to the peak position our parameterization becomes unreliable, so we cannot accurately describe the behaviour in this model at lower temperatures.

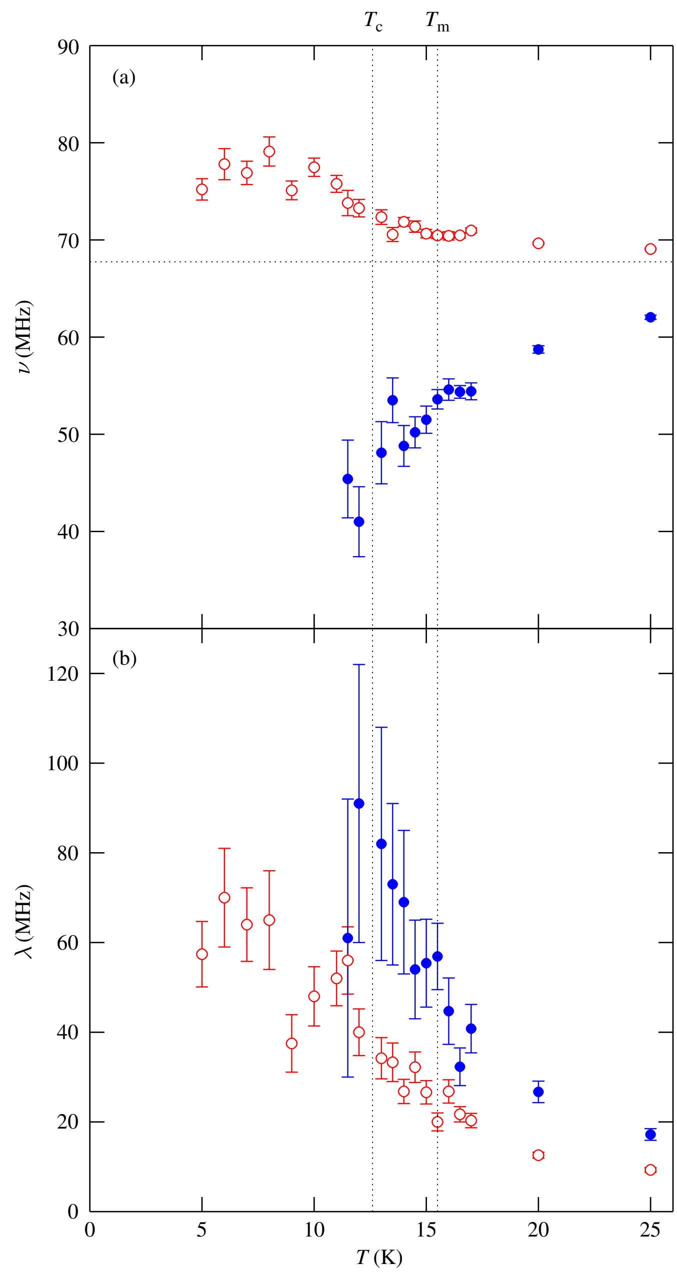

Fig. 2. (a) Oscillation frequency and (b) relaxation rates determined from time-domain fits of Eq. (1) to TF EuB_6 data. Dotted vertical lines mark the temperatures of the two transitions.

Zero field muon-spin rotation measurements were performed on three polycrystalline samples in the series $Eu_xCa_{1-x}B_6$, with $x = 0.9, 0.6$ and 0.35, again using the GPS spectrometer. The early time behaviour at low temperatures revealed a heavily damped oscillation for the $x = 0.9$ sample, demonstrating a bulk magnetic ground

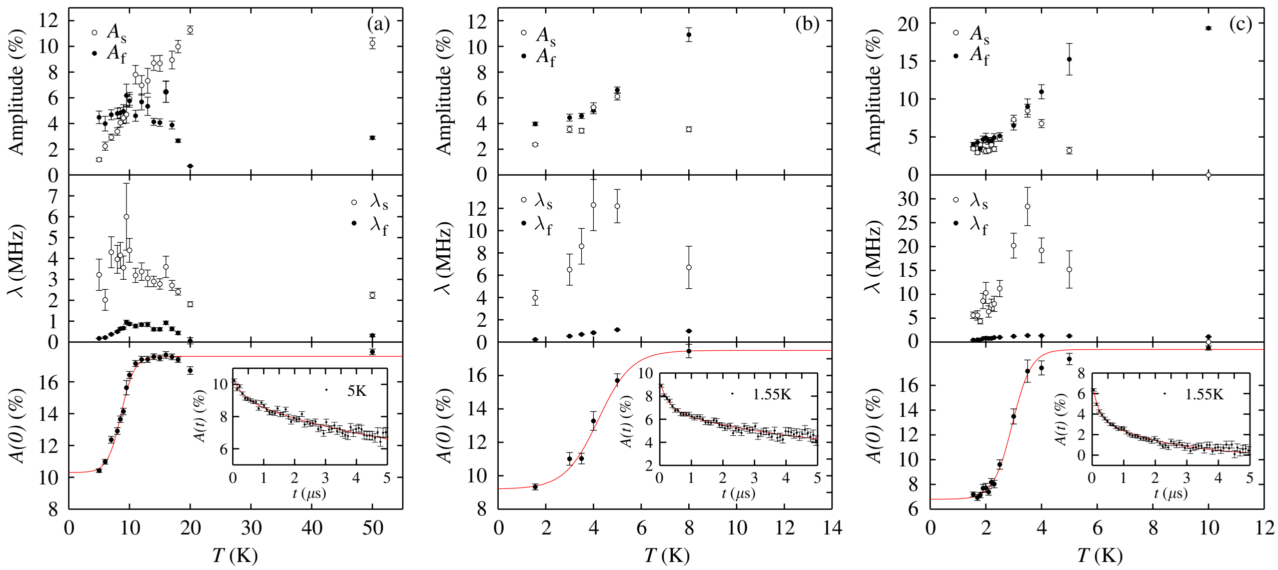

Fig. 3. Parameters resulting from fits to Eq. (2) of $Eu_xCa_{1-x}B_6$ with $x =$ (a) 0.9, (b) 0.6 and (c) 0.35. The top panels show amplitudes of the two fractions, the middle panels show the two relaxation rates and the lower panels show the initial asymmetry, with solid lines showing fits to Eq. (3). The small high-temperature value of $A(0)$ (~18%) is caused by running with the beamline in transverse field mode, so that the muon spin is rotated by ~50°. The inset to each lower panel shows the lowest temperature raw data with fit.

state. However, only a missing asymmetry fraction and the hint of a fast relaxation was seen in the more Ca-doped samples. This strong suppression of the oscillations previously observed in the $x = 1$ sample [11] is consistent with the increased magnetic disorder induced by the doping. It was possible to fit the longer time behaviour of the relaxation functions for all three samples with a two exponential function,

$$A(t) = A_{bg} + A_f \exp(-\lambda_f t) + A_s \exp(-\lambda_s t), \tag{2}$$

where A_{bg} represents a time-independent background arising from muons that halt in the silver surrounding the sample, and was fixed for each sample.

Fig. 3(a) shows the results of these fits for the $x = 0.9$ sample: the lower panel shows the initial asymmetry, $A(0)$, beginning to drop near 9 K and the middle panel shows a small peak in the relaxation rates at the same temperature. This is a clear signal that the material becomes magnetically ordered at this temperature, but the transition is unusually broad, suggesting that some regions of the sample order before others. This is consistent with a non-homogeneous distribution of Ca dopant [9]. The relative amplitudes of the two signals (upper panel) present a complicated picture: the amplitude of the faster relaxing fraction, A_s (driven by *slower* dynamics assuming the fast-fluctuation limit), grows steadily in amplitude with increasing temperature. The slower relaxing fraction, A_f, is constant at the lowest temperatures, but decreases as the temperature is raised further. The low temperature "missing" fraction is not really missing: it corresponds to the damped rapidly oscillating signal which is ignored by this long-time fitting procedure.

Fig. 3(b) demonstrates that the transition in $A(0)$ for the $x = 0.6$ sample is again extremely broad, and is accompanied by a broad peak in the two relaxation rates. The amplitude of the faster relaxing signal, A_s, is always non-zero, and peaks at the same temperature as its relaxation rate peaks. The amplitude of the slower relaxing signal, A_f, grows as we cross the transition with increasing temperature. This looks similar to the behaviour seen in the $x = 0.9$ sample, but in this case, it is the amplitude of the slower relaxing signal, A_f, that grows on warming and dominates at high temperature.

Fig. 3(c) shows the $x = 0.35$ sample having behaviour similar to that seen in the $x = 0.6$ sample: peaks in the relaxation rates with the amplitude of the faster relaxing signal shrinking on warming through the transition, and a slowly growing amplitude for the slower relaxing signal.

The previous muon study on EuB_6 fitted the long time signal with two exponentials above the transitions at least up to 55 K, with relaxation rates of ~2.5 and 0.08 MHz at the highest temperature. With increased Ca doping, the two relaxation rates tend to increase, which is again consistent with increasing disorder. The presence of two timescales suggests that phase separation may be important in all these samples.

The temperature and width of the transition were followed by fitting a Fermi-like function,

$$A + \frac{B - A}{1 + \exp\left(\frac{T_c - T}{\Delta}\right)}, \tag{3}$$

to the initial asymmetry data, where A and B represent the low- and high-temperature values of the initial asymmetry, T_c is the transition temperature and Δ is a measure of the width of the transition. The results are displayed in Table 1. The derived T_c values match well with those determined previously [9]. The width of the transition varies significantly with doping. We might expect that, as more disorder

Table 1
Results of fitting Eq. (3) to initial asymmetry data for $Eu_xCa_{1-x}B_6$ with $x = 0.9$, 0.6 and 0.35

	$x = 0.9$	$x = 0.6$	$x = 0.35$
T_c (K)	8.6(2)	4.2(2)	2.95(6)
Δ (K)	1.1(2)	0.7(2)	0.34(5)

is introduced with the addition of Ca, the width of the transition should increase, but the trend goes in the other direction.

In summary, TF μSR measurements allowed us to follow the temperature evolution of the local magnetic field distribution through the two magnetic transitions in EuB_6. They reveal a distribution with two peaks, a higher one that increases on cooling, possibly following the magnetic susceptibility of the sample, and a lower one that decreases significantly on cooling. ZF μSR measurements on samples of $Eu_xCa_{1-x}B_6$ with $x = 0.9$, 0.6 and 0.35 allow us to follow the suppression of the transition temperature with increased Ca doping, and reveal that the transition becomes sharper as this happens.

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