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μSR spectroscopy of the Kondo insulators $Lu_{1-x}Yb_xB_{12}$ ☆

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Abstract

Single crystals of $Yb_{1-x}Lu_xB_{12}$ ($x = 0, 0.125, 0.5, 1$) were measured between 1.8 and 300 K. We found similar spectral shapes in all compounds studied over the whole temperature range scanned. In zero field the spectra alter their appearance around 20, 100 and 150 K. In a longitudinal field of 100 G, which largely suppresses the contribution from ^{11}B nuclear moments the relaxation rate remained constant up to ~150 K where it suddenly peaks. These findings exclude magnetic correlations as the origin of muon spin relaxational behavior. The dominant features of the μSR spectra at low temperatures arise from lattice dynamics, probably involving the B_{12} clusters, rather than from spin dynamics. Higher temperature features are most likely muon related.

Keywords: Kondo insulator; Lattice dynamics; μSR

YbB_{12} is a well-established Kondo insulator [1]. Its susceptibility shows Curie behavior down to ~75 K with $\mu_{eff} = 4.4\mu_B$, then falls off rapidly due to the opening of a gap in the density of states at the Fermi energy. The ground state is considered non magnetic. We have performed μSR spectroscopy on high purity single crystalline samples of $Lu_{1-x}Yb_xB_{12}$ ($x = 0, 0.125, 0.5, 1$) between 1.8 and 300 K in zero (ZF), longitudinal (LF) and transverse field (TF) using the MORE facility of the Paul Scherrer Institute (Switzerland). This apparatus allows to record μSR spectra up to ~8 muon lifetimes ($\tau_\mu = 2.2$ μs). In contrast to pulsed beam μSR facilities it offers a good time resolution (~1 ns). Spectral distortions may occur in the first 0.25 μs. We have checked their absence by comparing spectra in normal and MORE mode. The samples consisted of mosaics of small discs not oriented with respect to their crystalline axes.

Previously published μSR data [2] on polycrystalline YbB_{12} were interpreted in terms of extremely weak slowly dynamic (~60 MHz) magnetic correlations of Yb moments of less than $10^{-2}\mu_B$ developing at low temperatures. This conclusion was based on LF = 200 G relaxation data which showed a collapse of muon spin relaxation rate above 10 K. The ZF spectra are dominated by dipolar couplings to the nuclear moment of ^{11}B. The original aim of the present study was to look for changes in local magnetic properties of YbB_{12} due to the gap closure initiated when Yb is partially replaced by Lu [3]. It was found, however, that our results, covering a wider temperature range and different members of the series $Lu_{1-x}Yb_xB_{12}$, cast doubt on the presence of magnetic correlations in YbB_{12}. We observed a rather similar μSR response in all four compounds, but the ZF spectra showed distinct variations with temperature. For example, as shown in Fig. 1 for the case of YbB_{12}, the asymmetry (μSR signal) decays for $t > 6$ μs more rapidly

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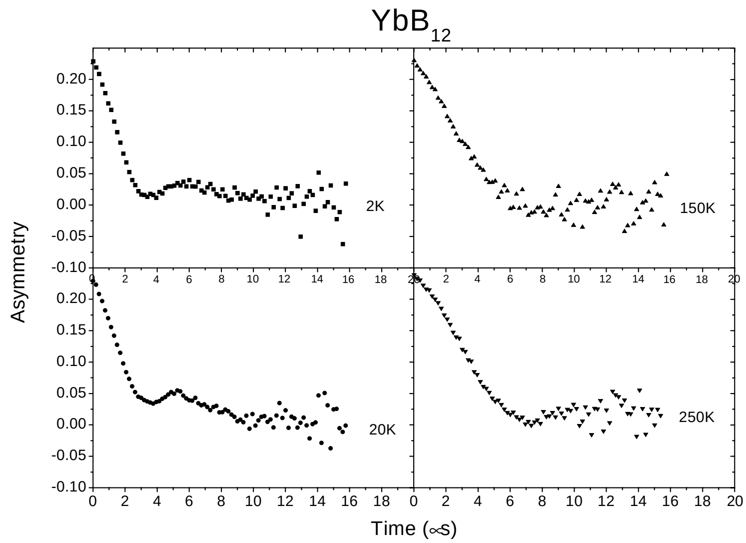

Fig. 1. Raw ZF-μSR spectra of YbB_{12} at various temperatures.

at 20 K than at 2 K. Other distinct variations in shape are seen around ~100 and ~150 K. This contradicts the previous interpretation of the μSR patterns using a single weakly dynamic nuclear Kubo–Toyabe (KT) relaxation with $\Delta \approx 0.5\ \mu s^{-1}$ independent of temperature (covering $T \leqslant 50$ K). In addition, this parameter set is unable to reproduce the spectra in low LF (5–50 G). Our results on the temperature dependence of the relaxation rate in LF = 100 G, which largely suppresses the influence of nuclear dipole coupling, are shown in Fig. 2 for all four compounds. Although the relaxation rate of YbB_{12} drops around 10 K (in agreement with Ref. [2]), it recovers its low temperature value for $20\ K \leqslant T \leqslant 100$ K. Most notably, at high temperatures (above ~200 K) this value of relaxation rate persists. Given the band of experimental uncertainties, Fig. 2 shows similar results for all four $Lu_{1-x}Yb_xB_{12}$ compounds. Their rate remains effectively constant with temperature until it suddenly peaks sharply at 150 K and then drops back. These results excludes magnetic couplings as the agent (LuB_{12} is a diamagnet). For LF $\geqslant 1000$ G the relaxation rate is found to be close to zero at all temperatures (in agreement with Ref. [2]), which means that one deals indeed with comparatively slow dynamical effects. It is difficult to find a theoretical spectral function which allows to least squares fit the ZF and low LF data up to 300 K. Below ~100 K, acceptable results were obtained with $A(t) = a_1 G_{KT}(\Delta_1, \tau) + a_2 G_{KT}(\Delta_2, \tau)$, i.e. a sum of two weakly dynamic Gaussian KT functions of nuclear origin with widths $\Delta_1 = 0.52(2)\ \mu s^{-1}$, $\Delta_2 = 0.15(3)\ \mu s^{-1}$ and $1/\tau \approx 1$ MHz. The relative strengths a_1, a_2 are temperature, but not field dependent. Details vary somewhat

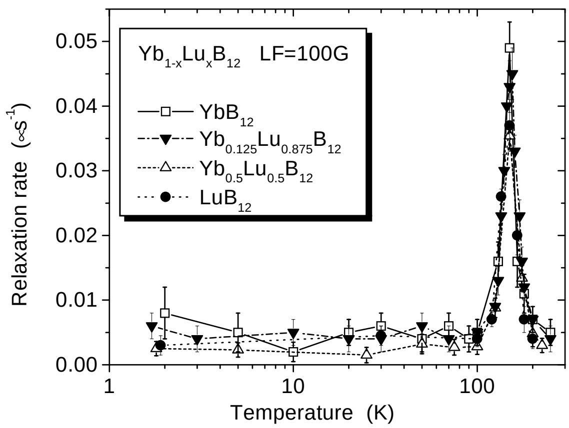

Fig. 2. Temperature dependence of the muon spin relaxation rate λ in LF = 100 G for $Lu_{1-x}Yb_xB_{12}$ obtained from fits to the function $A(t) = a\exp(-\lambda t)$.

from compound to compound without any clear correlation to the Lu concentration. Above ~100 K a single site KT function with roughly the weighted average of widths at 100 K ($\Delta = 0.22(1)\ \mu s^{-1}$) and the sum of intensities ($a = a_1 + a_2$) fits the spectra well. All differences in appearance in this regime are due to changes in the weak fluctuation rate $1/\tau$. The likely explanation is fast hopping between the two muon sites producing a single relaxation function by motional narrowing. Above 200 K this leads to a quasi-static KT relaxation pattern ($1/\tau_{eff} = 0$). A change from a two site to new single site muon stopping position is another possible scenario. It might be initiated by molecular rearrangement which would explain the peak in LF = 100 G relaxation rate.

In summary, our data show: (a) The μSR spectral features of $Y_{1-x}Lu_xB_{12}$ must arise from lattice and not spin dynamics. One probably deals with atomic motions within the B_{12} clusters. (b) The total set of relaxation data in LF = 100 G (Fig. 2) renders no evidence for a detectable moment at low temperatures on Yb. Conclusion (a) is supported by recent ^{171}Yb NMR measurements [4], where a minimum of $1/T_1$ was seen around 15 K which ties in with the change in ZF-μSR spectral shape seen around 20 K. The NMR results on ^{171}Yb differ from those from ^{11}B requiring an additional relaxation process for the B ions. Fluctuation in the field at the muon site generated by the ^{11}B moments leads to muon spin relaxation. This is the only contribution to the dynamic spectral shape in case no moment is present at Yb or if all of Yb is replaced by Lu. The features at 100 and 150 K are most probably muon related (e.g. hopping between muon sites) and not due to intrinsic properties of $Lu_{1-x}Yb_xB_{12}$.

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