Muon spin precession in ferromagnetic $DyAl_2$

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Abstract. The Laves phase intermetallics $REAl_2$ were studied by μSR over several decades. Results had mainly been obtained for the temperature and field dependencies of relaxation rates in the paramagnetic state. However, it turned out that spontaneous precession signals in the ordered magnetic state were very difficult to observe. We report here the observation a weak precession signal in a single crystalline sample of $DyAl_2$. The precession signal seen has low asymmetry and is heavily damped. This is understood with the notion that the magnetic structure of $DyAl_2$ splits the internal field at the interstitial muon stopping site into several components, and that of those only one of the field components leads to the visible precession signal. The observed temperature dependence of the precession frequency is compatible with the change of easy axis of magnetization at $T = 40$ K.

1. Introduction

Like all rare earth (RE) di-aluminates, $DyAl_2$ crystalizes in the C15 Laves phase structure. Other members of this crystallographic group are the analogeous compounds with transition metals and actinides. A fair number of these Laves phase intermetallics have been studied by μSR [1] on account of their fundamental magnetic behavior. This holds in particular for the $REAl_2$ intermetallics, mainly because (with the exception of RE = Ce) they show only one (second order) magnetic phase transition into a simple ferromagnetic state. Nearly all of the earlier $REAl_2$ studies concentrated on the paramagnetic range, since the precession signals in the ferromagnetic state could not be seen (again with the exception of $CeAl_2$ and one recently found case). Those extensive studies of $REAl_2$ compounds in the paramagnetic range [2, 3, 4], including prominently $DyAl_2$, were undertaken with the aim to elucidate the fluctuations of paramagnetic 4f-moments, especially on approach to the Curie point. The μSR spectra show single pure exponential decay of asymmetry with rate λ_{pm}. Foremost established was the divergent rise in $\lambda_{pm} \propto \tau_{4f}$ (with $1/\tau_{4f}$ being the fluctuation rate of the 4f moments) over a fairly wide temperature range above T_C, i.e., much wider than the common 'critical range' in mean field theory. This established regime of strong rise in λ_{pm} is commonly termed 'critical slowing down' and reflects the growing size of spin clusters due to increasing spin-spin correlations. The larger the spin cluster, the longer the time needed to turn the full cluster by thermal agitation (see also the discussion in [5] relating to iron and some of its intermetallics). Also explored in the previous study of $REAl_2$ materials was the temperature dependence of the paramagnetic frequency shift which can have magnitudes of several percent and shows as well a strong rise on aprroaching T_C, quite compatible with the picture for λ_{pm}. The above mentioned recent exception where a signal in the ferromagnetic state has been seen is $SmAl_2$ [6], which has a comparatively small magnetic moment. Its μSR spectra display a clear precession pattern with normal asymmetry.

In this report we present new results on DyAl$_2$ using a spherical single crystalline sample. We were able to observe a weak precession signal in the ferromagnetic regime ($T_C \approx 65\,\mathrm{K}$), a first in a high moment REAl$_2$ compound.

2. Experimental

We have carried out our studies on single crystalline DyAl$_2$ at different facilities. With surface muons at M13 of TRIUMF and DOLLY of PSI as well as with backward muons at GPD of PSI. Zero field (ZF) and (in the paramagnetic range) weak tranverse field (TF) data were taken from 190 K down to 10 K in different orientations between [100] and [111]. A previous run sequence with the single crystalline sample at ISIS extended the higher temperature region to 1000 K. The low temperature range T<T_C includes in particular the change in easy axis of magnetization being [111] close to T_C and [100] at low temperatures which, according to [7], occurs around 40 K.

3. Results

3.1. Paramagnetic range

The μSR spectra for $T > T_C$ show in ZF as well as in TF single simple exponential decay of muon spin polarization with full initial asymmetry confirming a unique single interstitial muon stopping site. This conclusion is further corroborated by high field (up to 300 mT) TF measurements which still show a single signal shifted by a well defined paramagnetic Knigt shift [2]. The single site approach holds for all members of the REAl$_2$ series and also in μSR studies of other C15 Laves phase intermetallics [1], especially for the results in magnetically ordered CeAl$_2$ and SmAl$_2$. Examples of paramagnetic ZF spectra taken within this work are depicted in Fig. 2-left (top curves).
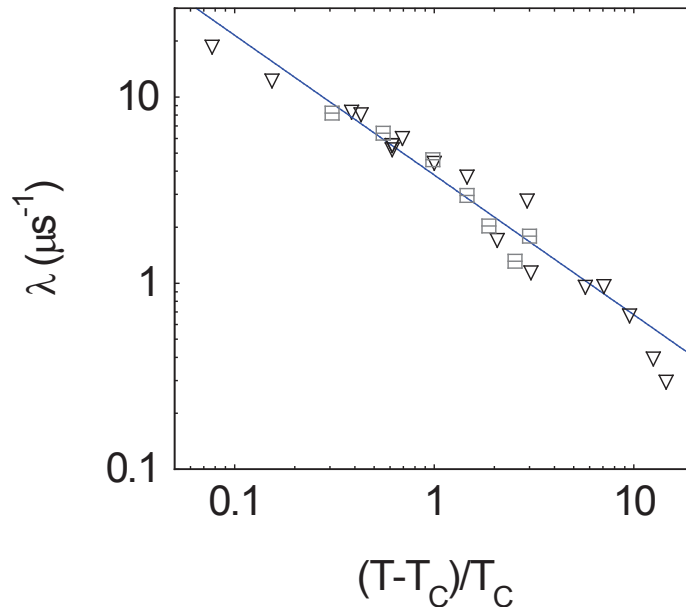

Figure 1. Dependence on reduced temperature of the paramagnetic relaxation rate in DyAl$_2$ from ZF and low TF measurements. Triangles: ZF data from the single crystalline sample; Squares: Older weak TF data from a polycristalline sample. The line is a least squares fit explained in text.

The temperature dependence of the paramagnetic relaxation rate λ_{pm} is displayed in Fig. 1 including earlier weak TF data [2] on a polycrystalline sample. The new and the old data agree

well with each other. The solid line is a least squares fit giving $\lambda_{\text{pm}} \propto [(\text{T-}T_{\text{C}})/T_{\text{C}}]^{-w}$ with $w = 0.76 \pm 0.06$, in agreement with values found in earlier studies [3, 4].
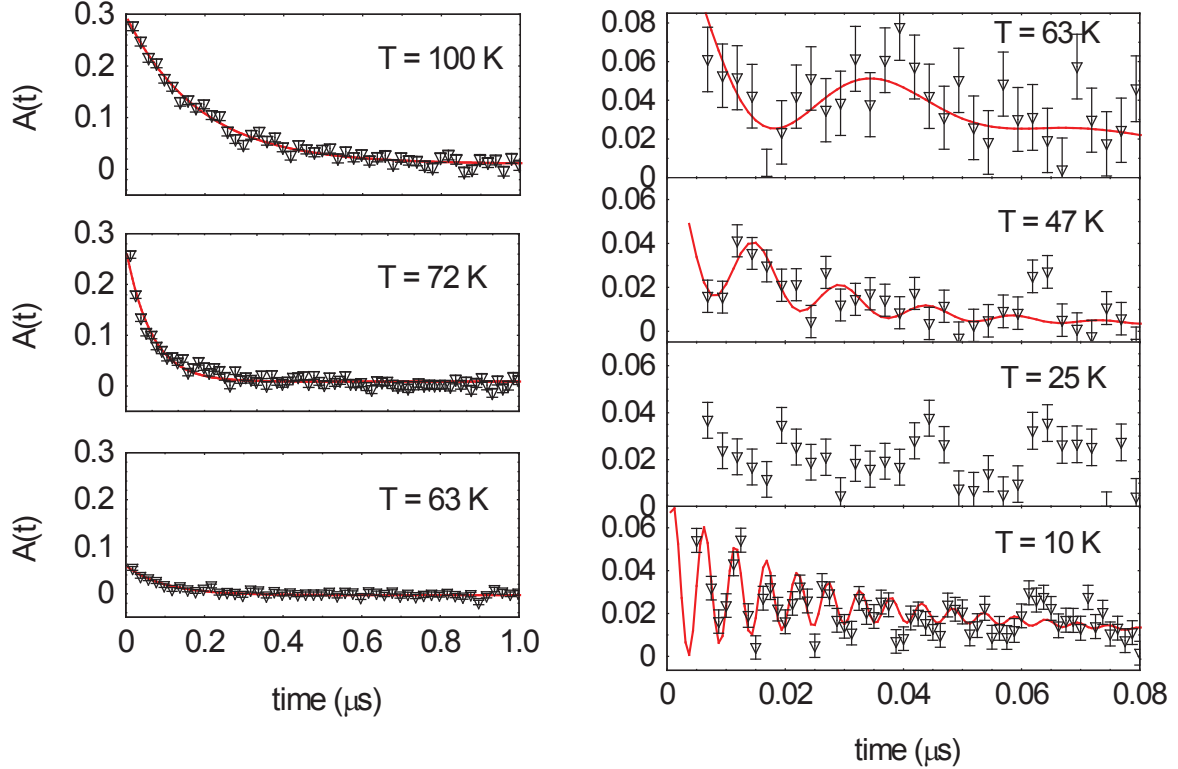
Figure 2. Left: ZF spectra at $100\,\text{K}$, $72\,\text{K}$ (paramagnetic) and $63\,\text{K}$ (ferromagnetic). Right: Muon precession patterns in the ferromagnetic regime at T = $63\,\text{K}$, $47\,\text{K}$, $25\,\text{K}$, and $10\,\text{K}$. Note the different scales of the x and y axes between the left and right panels. The lines are least squares fits discussed in text. The $25\,\text{K}$ spectrum shows no distict precession pattern and cannot be fitted in the usual manner as discussed in text

3.2. Ferromagnetic range
A muon signal is visible in all the three sets of data introduced in section 2. Spectra at select temperatures are shown in Fig. 2-b. They all have a low initial asymmetry of 0.03-0.05, in contrast to the paramagnetic spectra with $A_0 \sim 0.2$. Clearly, only a part of the full muon signal is observable below T_{C}. The remaining signal relaxes extremely fast and is in most cases hidden within the initial dead time (approximately between 5 and 10 ns) of the μSR spectrometer. In the majority of cases, a spontaneous muon spin precession signal could be seen, yet the oscillations were heavily damped (up to ~ 50 μs^{-1}). The spectra were fitted with a single cosine oscillation having exponential damping. It cannot be excluded that some spectra contain more than one oscillatory frequency. Some of the data points can be succesfully fitted with a set of free parameters, but in others, expecially for the GPD data, the phase and the initial asymmetry (set to 0.04) had to be kept fixed. In the temperature region between 40-15 K the data cannot be fitted reliably in that way, neither with a single oscillation nor with two oscillation frequencies. The signal is just too heavily damped for the fit procedure to lock on to oscillations. Nearly all of the signal is within the dead time range and not observable. A typical example is the spectrum at $25\,\text{K}$ in Fig. 2-right. The high damping rate also prevents a location of precession frequencies in a FFT analysis. The interstitial fields derived from the observed frequencies in the cosine oscillation fits are summarized in Fig 3.
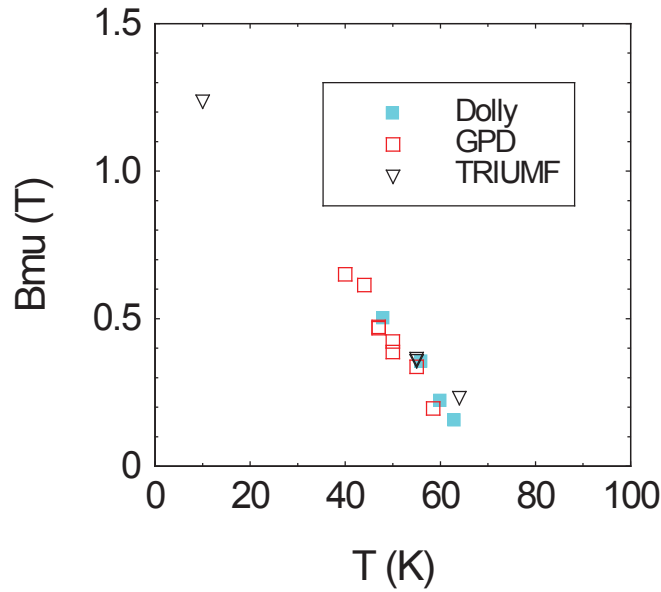
Figure 3. Observed interstitial fields in $DyAl_2$. The region between $40\,K$ and $15\,K$ is excluded since reliable fits could not de obtained (see text)

4. Discussion

As said in the introductory chapter, the exponent w of the temperature dependence of the paramagnetic relaxation rate cannot be related directly to theoretical treatments of phase transitions since it is not the critical parameter in their sense. This issue is discussed extensively in [8], and also in [5]. The value of w varies markedlly throughout the literature and often turns out to be different for different samples of the same compound. Clearly more macroscopic parameters enter like crystal faults, grain sizes and domain volume enter. It is usualy also method sensitive, even within the various nuclear methods.

In all $REAl_2$ compounds we expect the muons to reside at the 2-2 site (g-site) of the C15 Laves phase structure that is shown in Fig. 4. It is the preferred site for low concentration of hydrogen (see discussion in [2]), and was also found for muons in $CeAl_2$ [9]. Furthermore the muons are likely to be stationary below $T = 80\,K$ as seen in non-magnetic $LaAl_2$ [10].

That only a part of the muon signal is observed in the ferromagnetic range is presumably due to the fact that the magnetic structure of the compound will split the local fields at the
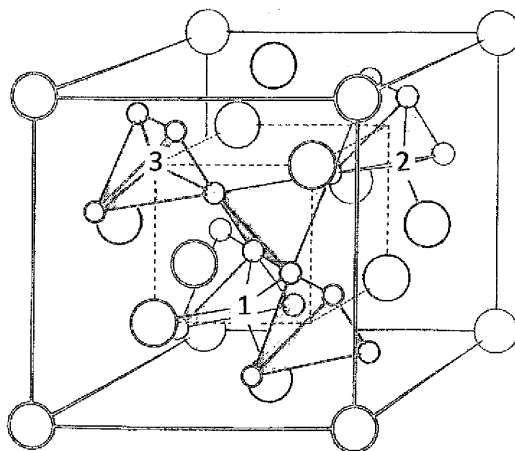

Figure 4. Structure of the C15 $REAl_2$ lattice (RE = large circles, Al = small circles). The 3 types of interstitial sites are marked 1 ,2 ,3 in the figure with 1 being the likely muon position (2-2 site).

interstitial sites into several components. As shown in Fig. 4, the 2–2-site forms a tetrahedron with 2 RE and 2 Al atoms, but the distances from the muon to the RE and the Al atoms are not identical. There are 96 different 2–2 sites in the unit cell and the distribution of dipolar fields on these sites depends on the magnetization direction. Calculations of the magnitude of the internal fields are shown in Fig. 5 for muons on the 2–2 sites, and also for a slightly shifted 2–2-site where the distances to the 4 neighboring atoms are all equal. In the standard 2–2 site there are 4 or 6 different fields, but they are pairwise only of opposite signs, reducing the numbers to 2 and 3 respectively. If the muon position is offset even further splittings occur as displayed in Fig. 5-c and -d.
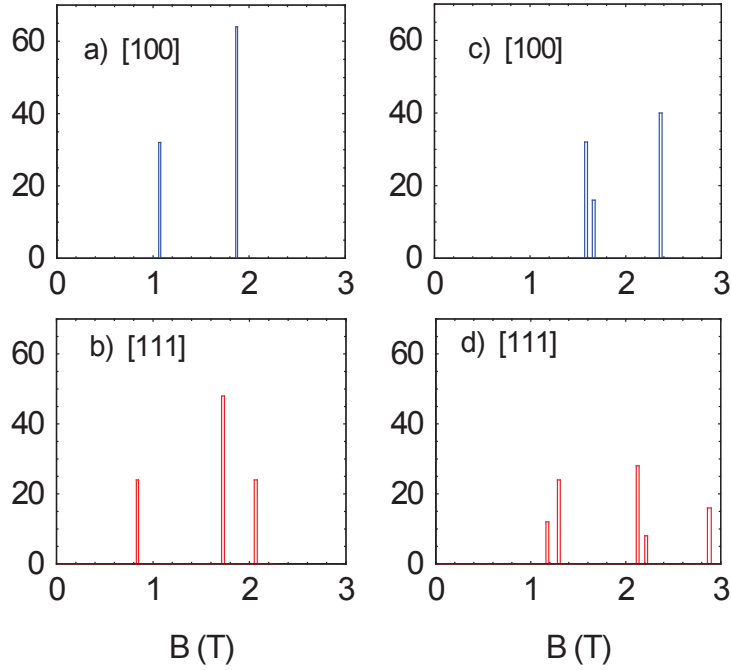

Figure 5. Relative magnitude of calculated dipolar fields at T=0 in $DyAl_2$ for [100] and [111] magnetization directions in regular 2-2 sites (a and b), and muon shifted 2-2-sites (c and d). See text for details.

The saturation value of the interstitial field B_μ is on the order of $1\,T$ a value considerably larger than that found [6] in $SmAl_2$ ($\sim 0.3\,T$), reflecting mainly the much larger free ion moment of Dy^{3+}. It is, in fact, in roughly the same range as B_μ observed in ferromagnetic dysprosium metal [11, 1]

The temperature dependence of B_μ shown in Fig. 3 is unusual since it shows essentially a linear behavior. Clearly, this relies on the single low temperature data point ($10\,K$), yet this data point is characterized by a fairly clear oscillatory pattern (see Fig. 2-right) defining its frequency rather well. In contrast, the temperature dependence of B_μ in $SmAl_2$ can well be reproduced [6] by the common Bloch spin wave approach (Brilluoin-like behavior). A nearly linear dependence of B_{hf} could possibly result from strong crystalline electric field (CEF) interaction. In a high moment Kramers ion as Dy^{3+} CEF interactions are small. On the other hand, a reorientation transition of the easy axis of magnetization from [111] to [100] at $40\,K$ has been established for $DyAl_2$ from magnetization and heat capacity data on a single crystal [7]. The calculations of the dipolar field component show a shift to higher values when changing the axis of magnetization from [111] to [100], as seen when comparing panels b) and a) in Fig. 5. At $40\,K$, corresponding to $T/T_C \approx 0.6$, one expects the value of B_μ to be close to its saturation value $B_\mu(T \to 0)$ and no large rise in field should occur when lowering the temperature further. The observation of
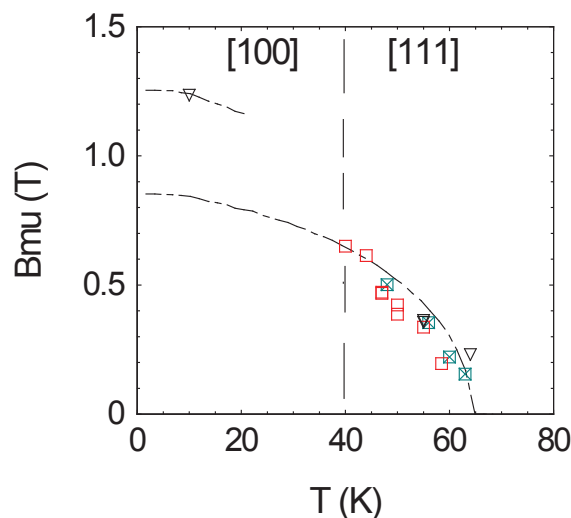
Figure 6. Possible explanation of the dependence of the local field on temperature (see text for details). The broken lines are theoretical magnetization functions for $J = 15/2$.

the opposite behavior then supports the interpretation that the rise in field for T<40 K is the outcome of the change of magnetization direction. Assumed in this conclusion is, however, that we only observe the lowest precession frequencies and that the contribution from the contact field at the muon site is small compared to the dipole field. The latter assumption in particular is quite reasonable (see [1]).

One may finally interpret the dependence of $B_\mu(T)$ shown in Fig. 3 as consisting of two 'normal' (i.e., Brillouin-like) temperature dependences of the interstitial field as displayed in Fig. 6. It has been said before, that unfortunately reliable fits of the spectra for temperatures between 40 and 15 K in terms of one or more cosine frequencies were not possible because of excessive damping rates. The most probable explanation is that the reorientation of magnetization directions is distributed in temperature and consequently, the spectra consist of an overlay of various precession signals. Such a spread of spin turning transition has been oserved previously, for example in the spin reorientation transition in Gd metal [12], albeit over a smaller temperature range.

Acknowledgement

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