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Pressure Dependent Magnetism in $Y_{1.05}(Mn_{0.95}Al_{0.05})_2$

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Abstract

The Laves phase compound YMn_2 exhibits a discontinuous transition from a spin fluctuating Pauli paramagnetic state above 110 K to an expanded, localised moment helical antiferromagnetic state below 110 K. Substitution of Al for Mn in the pseudo-binary compound $Y(Mn_{1-x}Al_x)_2$ exerts a negative chemical pressure, expanding the unit cell and further localizing the Mn moment. Here we present the results of a μSR study of spin fluctuations in $Y(Mn_{1-x}Al_x)_2$ ($x = 0.05$) in which external mechanical pressure (4.5 kbar) is applied to counteract the Al induced chemical pressure and destabilize the local Mn moment.

Keywords: $Y(MnAl)_2$; itinerant electron magnetism; muon spin relaxation

1. Introduction

The cubic C15 Laves phase compound, YMn_2, is Pauli paramagnetic down to 110 K at which temperature a discontinuous 5 % volume expansion occurs and the Mn moment spontaneously localises. Neutron powder diffraction has shown that this transition is accompanied by a tetragonal distortion of the unit cell and the onset of long range antiferromagnetic ordering consistent with a spin arrangement in which the moments lay in the (1,0,0) plane, propagating helically along both the [1,0,0] direction with a period of 430 Å and the [0,1,0] direction with a period in the order of 2500 Å [1]. Further studies have shown that the average Mn-Mn separation in YMn_2 plays a significant role in the localization of the 3d manganese moments and, through the partial substitution of Mn for larger or smaller atoms, the Mn moment can be stabilised or made to collapse entirely [2]. YMn_2 and its ternary derivatives are therefore interesting model systems with which to explore the mechanisms responsible for moment localisation in

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3d spin fluctuating systems, for example within the framework of Moriya's Self Consistent Renormalisation (SCR) theory of spin fluctuations [3].

The substitution of Al for Mn in YMn_2 increases the Mn-Mn separation and leads to a gradual transition from the itinerant behaviour of YMn_2 to more localised behaviour. While the volume expansion and moment localisation remains discontinuous for $x < 0.03$, the increasing Mn-Mn distance resulting from the negative chemical pressure exerted by the Al atoms, together with the disruption of the long range anti-ferromagnetic order caused by the non-magnetic aluminium, leads to a continuous phase change for concentrations greater than $x = 0.03$. Between $x = 0.03$ and $x = 0.10$, a highly frustrated spin glass-like magnetic state is observed [4].

Previously μSR has proved extremely useful in the extensive characterisation of the spin dynamics and the collapse of long range magnetic order resulting from inverse *chemical* pressure on $Y(Mn_{0.9}Al_{0.1})_2$ [5]. Here, we present the results of a μSR study of $Y(Mn_{1-x}Al_x)_2$ with $x = 0.05$ in which we have applied *external* mechanical pressure (P = 4.5 kbar) to counteract the chemically induced expansion of the lattice in order to explore whether the Mn moment can once again be destabilised and a pure YMn_2-like state recovered.

2. Experimental Details

The $Y_{1.05}(Mn_{0.95}Al_{0.05})_2$ sample was prepared by melting stoichiometric proportions of pure (99.995 %) constituent materials in a water cooled argon arc furnace. Excess yttrium was added to prevent the formation of the highly ferromagnetic Y_6Mn_{23} impurity phase. The polycrystalline ingots were sealed in quartz ampoules under vacuum and annealed at 800 °C before quenching in liquid nitrogen. The resulting ingots were then crushed into a fine powder.

The high pressure (P = 4.5 kbar) μSR experiments were carried out at the RIKEN-RAL muon facility using the ARGUS spectrometer with the sample loaded into a Copped-Beryllium (CuBe) pressure cell. The pressure was applied using a helium gas intensifier and monitored as the temperature was reduced. Any pressure drop, resulting from helium contraction, was compensated for to ensure that the pressure remained at a constant 4.5 kbar even at the lowest temperatures. Simplification of the background response arising from nuclear dipole relaxation associated with the pressure cell material was achieved by applying a 110 Gauss longitudinal field, as shown in Figure 1(a). As a reference, ambient pressure (i.e. P = 0 kbar) measurements were collected using the MuSR spectrometer at the ISIS facility, with the sample loaded onto a silver plate. All spectra were analysed using the data analysis program WIMDA [6].

3. Results and Discussion

Under ambient pressure conditions (P = 0 kbar), all spectra collected in the paramagnetic regime are well described using a muon spin relaxation function, $G_z(t)$, of the form,

$$G_z(t) = \exp(-(\lambda t)^{\beta}) \tag{1}$$

with a stretching parameter, β, that is predominantly less than unity (Figure 1(b) insert); β falling from unity at room temperature to approximately 1/3 by 85K. Such behaviour is a characteristic of many concentrated spin glass systems and, for β values less than 1, generally indicative of the muon sensing a distribution of μ^+ spin relaxation processes within the sample [7, 8]. The associated effective relaxation rate, λ(T), is seen to increase slowly as the temperature decreases from 290 to 100 K before diverging.

In contrast, application of 4.5 kbar is seen to produce a marked change in both β(T) and λ(T). Not only is the magnitude of λ(T), which peaks at approximately 50 K, significantly reduced when the system is subjected to high pressure, but β tends to unity at all measured temperatures. Such a simple exponential description of the data suggests that the muon senses a single relaxation process. This simple exponential form of $G_z(t)$ is consistent with the response reported for the parent compound, YMn_2 [1]. Figure 1(b) compares λ(T) at ambient pressure and 4.5 kbar.

Preliminary analysis of λ(T, P = 0), by fitting a critical scaling model to the data, namely,

$$\lambda(T) = \lambda_0 \left(\frac{T - T_g}{T_g} \right)^{-\gamma} \tag{2}$$

yields a transition temperature of T_g = 88.8 +/- 0.2 K. Similar parameterisation of the P = 4.5 kbar data, however, is compromised by significant uncertainty in the resulting fit parameters. The failure of equation (2) to model, with precision, the high pressure data may suggest a significant change in the nature of the spin fluctuations with applied pressure.

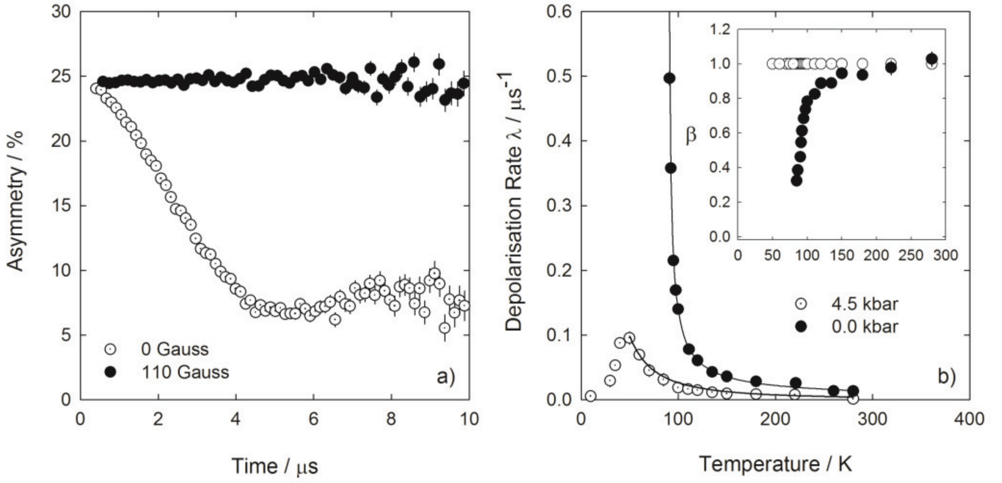

Figure 1: (a) Suppression of nuclear dipole relaxation from the CuBe pressure cell via application of an external longitudinal field of 110 Gauss. (b) Temperature dependence of the muon spin depolarization rate, λ(T), with the $Y_{1.05}(Mn_{0.95}Al_{0.05})_2$ sample at (●) ambient pressure and (○) under 4.5 kbar. The solid lines are, for the P = 0 kbar data, a fit to equation 2 and, for the P = 4.5 kbar data, a guide to the eye. Insert: The temperature and pressure dependence of the stretching exponent, β.

4. Conclusions

The stretched exponential description of muon spin relaxation spectra collected from $Y(Mn_{0.95}Al_{0.05})_2$ at ambient pressure, and in the paramagnetic regime, is consistent with that expected from a concentrated, rather than dilute, spin-glass-like system [7]. A glass transition temperature of T_g = (88.8 +/- 0.2) K has been determined by fitting a critical form to the data. T_g determined in this way is consistent with previously reported values [4]. In contrast, application of 4.5 kbar external pressure not only leads to slower muon spin depolarisation rates, but also suppresses the aforementioned magnetic transition temperature and significantly changes the form of the muon spin relaxation function, $G_z(t)$, required to describe the data; $G_z(t)$ evolving from a stretched to a simple exponential. Such a response is indicative of the muon sensing a single, rather than distributed, relaxation process when the sample is subject to external pressure. It is possible that application of external pressure destabilises the Mn moments, significantly suppressing the spin glass transition and returning the system to a spin fluctuating magnetic state with a simple exponential μ^+ spin relaxation response similar to that observed from the parent compound (YMn_2) under ambient conditions. Based upon the results presented here, it appears that 3d Mn moment localisation, associated with the introduction of inverse chemical pressure arising from Al substitution for Mn, is reversible.

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