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Studies of the time dependence of the magnetization of T-Al$_3$Mn(Pd,Fe) compounds

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Abstract. The structure of the complex intermetallic compound T-Al$_3$Mn, an approximant of the decagonal quasicrystal, persists by substitution of Pd or Fe for Mn. Magnetically spin glass behaviour is obtained. Relative to the mother compound substitution of Pd (Fe) decreases (increases) the freezing temperature $T_f$. The time dependence of the magnetic moment points to broad activation energy distributions with similar shape but different centre of gravity. For comparable temperatures the Fe containing compounds exhibit larger height of the barriers than those without and with Pd substitution. For the decagonal quasicrystal the mean effective activation energy is reduced compared to the one for the T-phase compounds.

1. Introduction

The Taylor phases, in which the complex intermetallic compound T-Al$_3$Mn is formed, contain 156 atoms in the orthorhombic unit cell (space group Pnma). Some of the sites are either too close in space to be occupied simultaneously or show mixed Al/Mn occupancy. This leads to inherent chemical and spatial disorder in the lattice, and is further pronounced by substitution of Pd or Fe for Mn, viable without destroying the structure [1]. Spatial disorder and random distribution of magnetic and nonmagnetic elements in intermetallics are important ingredients for spin glass behaviour, which manifests in the appearance of irreversibilities, asymmetric hysteresis loops and strong time dependence of the magnetization as well as after effects below a characteristic freezing temperature $T_f$. Reason for this behaviour is the highly degenerate free energy landscape, with distributions of barriers between local minima. Measurement of the time dependence of the magnetization allows gaining information about the barriers. A detailed study of magnetic after effects on Al$_{75}$Mn$_{25}$-x(Pd,Fe)$_x$ was reported by Dolinsek et al [2]. In this work we focus on the influence of a further extended substitution of Pd and Fe on both $T_f$ and the barrier heights characterized by the mean activation energy using compounds with slightly different Al content.

2. Experimental

Five different samples were prepared for this investigation: Al$_{75}$Mn$_{25}$ (AM), Al$_{75}$Mn$_{20}$Pd$_5$ (AMP), Al$_{69}$Mn$_{23}$Fe$_8$ (AMF8), Al$_{71}$Mn$_{19}$Fe$_{10}$ (AMF10), and Al$_{68}$Mn$_{20}$Fe$_{12}$ (AMF12). With the exception of AMF10 all samples show the T-Al$_3$Mn structure, where Pd and Fe occupy the 8d and 4c sites [1-3]. AMF10 crystallizes as decagonal quasicrystal. Details of sample preparation are given in [1, 2].
Magnetic measurements were performed in fields up to 9 T and temperatures between 2 and 300 K by cooling in zero field to the base temperature and then applying the respective field (ZFC) and by cooling in the respective field cooling (FC) using a QD-PPMS vibrating sample magnetometer. Freezing temperatures $T_f$ were determined from the cusp in the ZFC magnetization curves. The time dependence of the magnetic moment was recorded at temperatures between 2 K and temperatures slightly above $T_f$ in external fields of 0.50 T and 2 T after zero field cooling from temperatures above the freezing temperature. Waiting time $t_w$ before applying the field was always 240 s.

3. Results and Discussion
The irreversibilities appearing between ZFC and FC curves and the typical temperature dependence of the magnetization recorded in fields between 0.8 mT and 9 T (example given in figure 1) indicate spin glass behavior. A fit according to $T_f(B_a) = T_f(0) + a*(B_a)^b$ for the field dependence of $T_f$ for the different samples (figure 2) leads to values for $b$ between 0.10 and 0.15, which are far away from 0.66, typical for a Thouless – d’Almeida line [4]. Figure 4 gives an overview of the influence of the different Fe/Pd substitutions in comparison with results obtained from Dolinsek et al [2] taking into account the difference in the Al concentration of the two sets of samples. As typical for spin glasses, $T_f$
Figure 4. Dependence of $T_f$ determined at 0.8 mT for samples with different Al content (numbers close to symbols) on the concentration of Pd/Fe. Lower straight line connects decagonal phase samples. The lines are guide for the eye.

decreases with substitution of nonmagnetic Pd, whereas the freezing temperature increases by substitution of magnetic Fe atoms. The samples showing the Taylor phase (2, 4, 8, and 12 at% Fe) have slightly higher $T_f$ values than the compounds crystallizing as decagonal quasicrystal (6 and 10 at% Fe).

Within the chosen time interval the magnetization exhibits in first approximation logarithmic time dependence (figure 3). With increasing temperatures small deviations from this behaviour are obtained. (e.g. data recorded at 9 K in figure 3). Above $T_f$ no relaxation is observed within the experimental resolution. If the time dependence is described in the simplest way as a thermally activated process with one characteristic barrier, an exponential decrease according to a simple Debye process $M(t) = M(0) \times \exp(-t/\tau)$ is expected, where the characteristic relaxation time $\tau$ is given by the Arrhenius equation $\tau = \tau_0 \times \exp(E/kT)$. $E$ is the effective barrier height, which has to be overcome by

Figure 5. Temperature dependence of the normalized creep rate ($S$) for the investigated samples measured at 0.5 T. The lines are guide for the eye.

Figure 6. Temperature dependence of the mean barrier height $E$ obtained at 0.5 T and 2.0 T for sample AFM8. The lines are guide for the eye.

Figure 7. Temperature dependence of the mean barrier height $E$ obtained at 0.5 T for the investigated samples. The lines are guide for the eye.
thermal activation. A logarithmic instead of the before mentioned exponential time dependence is an indication that not one energy barrier but a broad distribution of barrier energies is present. In that case the centre of gravity of the distribution leads to the mean activation energy \( E \). The logarithmic behavior allows the determination of a creep rate \( \Gamma = \frac{dM}{d\ln t} \). In figure 5 the temperature dependence of the creep rate normalized to the magnetization at the starting time \( S = \Gamma/M(t_0) \) is shown. Uncertainties of the creep rate, due to aging of the samples, are small, as the aging effects in this type of samples are below 5% [2]. For all investigated samples \( S \) shows strong temperature dependence with a maximum appearing at approximately \( T_f/4 \) for 0.5 T and much lower values for 2.0 T. It can be shown that \( \Gamma \) is related to the effective barrier height by \( E = kT_{\ast}(dM/d\theta)/\Gamma \), where \( \theta = T/T_f \) [5].

With increasing temperature the mean effective activation energy increases for both external fields 0.5 T and 2.0 T (figure 6). This is in contradiction to what is expected, as both the barriers which evolve with decreasing temperature should decrease and the time dependence of the magnetization should become stronger with increasing \( T \). Interpreting, however, \( E \) as the centre of gravity of an energy distribution this increase can be understood. For a given temperature, part of the relaxations on the low energy side of the distribution function does not contribute to the time dependence of the magnetization, because already having reached thermal equilibrium within the experimental time scale. This can be expressed by introducing a cut-off energy \( E_c = kT_t\ln(t/\tau) \), which shifts to higher values for higher temperatures, shifting also the centre of gravity to higher energies. The steepness of the increase of \( E(T) \) is a measure of the width of the energy distribution. Increase of the applied field reduces the effective barrier height, leading to smaller \( E \) values (figure 6). The appearance of similar temperature dependences of \( E \) (figure 7) points to similar shapes of the barrier distributions for the different samples, although differences of the absolute values are present. Small values are found for samples AM and AMP which amongst themselves show no difference within the measuring accuracy. Higher values are found for the Fe-substituted samples, with negligible differences for 8 and 12% Fe, but lower values for the decagonal quasicrystal AMF10. An interpretation of the time dependence within a classical droplet model allows an interpretation of \( E \) in terms of magnetic cluster sizes. In that sense the magnetic entities rotating in the external field are larger in the Fe-substituted than in the Pd- and the unsubstituted sample, which is not unexpected as Fe increases magnetic coupling. Although the T-phase is an approximant of the decagonal quasicrystal – that means that the structural building blocks are very similar – the intrinsic energy landscape is clearly different. It is surprising that the barriers are higher in the T-phase, although the symmetry is lower in the decagonal phase.

In summary, we have investigated the evolution of the freezing temperature with substitution of Pd and Fe in the complex metallic alloy T-Al\(_3\)Mn. \( T_f \) decreases with increasing Pd and increases with increasing Fe content. Relaxation measurements point to broad distributions of activation energies. Although the shape of these distributions is very similar for the investigated samples, the centre of gravity is different. Higher values are obtained for the Fe containing compounds, indicating, that magnetic correlations are stronger.

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