Magnetism of decagonal $Al_{69.8}Pd_{12.1}Mn_{18.1}$

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Abstract

We report the results of measurements of ^{27}Al NMR spectra and the DC susceptibility of polycrystalline decagonal quasicrystalline $Al_{69.8}Pd_{12.1}Mn_{18.1}$. The temperature-variation of the magnetic susceptibility $\chi(T)$ and the ^{27}Al NMR-spectra imply that a fraction of the order of 20% of the Mn-atoms carry a magnetic moment, and that a spin-glass-type freezing of these Mn moments occurs at $T_f = 12$K. The NMR spectra reveal two partially resolved lines for the ^{27}Al nuclei, indicating that there are two different sets of local environments for the Al-sites. Below 100K and upon decreasing temperatures, the ^{27}Al-NMR line-width Δ increases with a progressively increasing negative slope $d\Delta/dT$, as it is often observed in relation with spin-glass freezing processes.

Key words: magnetism; quasicrystal; NMR; spin glass

1. Introduction

Since the discovery of quasicrystalline systems by Shechtman et al. in 1982 there has been lively interest in the peculiar magnetic properties of these compounds. While the initial studies were mainly performed on metastable AlMn quasicrystals (QC), the discovery of the stable icosahedral *i*-AlPdMn (QC) family motivated experimental and theoretical studies of the influence of the icosahedral structure on the *d*-electron magnetism of the Mn-ions. In spite of the variety of magnetic properties found in these QC's, such as diamagnetism, (unconventional) paramagnetism[1], or spin-glass freezing[2,3], they seem to exhibit a common feature, i.e., only a small fraction of the manganese ions carry a magnetic moment[1–3]. In comparison with the icosahedral AlPdMn family, the situation is much less clear for decagonal AlPdMn alloys. This work aims at giving insights into the magnetic properties of a sample of *d*-$Al_{69.8}Pd_{12.1}Mn_{18.1}$.

2. The sample

The structure of this alloy consists of periodically stacked decagonal quasiperiodic layers. One translational period with a length $d = 12.56$Å is built up by two different types of layers: a puckered layer P and a flat layer F[5]. The ratio of Al in P to Al in F is approximately 2:1; for Mn this ratio is about 1:8. Decagonal AlPdMn is stable only in a very narrow range of composition in the phase diagram. Our sample was obtained by annealing melt-spun flakes with the nominal composition $Al_{69.8}Pd_{12.1}Mn_{18.1}$. After rapidly quenching the flakes contain a metastable icosahedral phase, that can be transformed, without long-range diffusion, into *d*-$Al_{69.8}Pd_{12.1}Mn_{18.1}$[4]. The resulting tapes contain polycrystalline *d*-$Al_{69.8}Pd_{12.1}Mn_{18.1}$ with no linear phason strains. Selected area electron diffraction confirmed the high perfection of the decagonal symmetry in the planes and the periodicity along the *c*-axis. Scanning electron microscopy sets the upper limit of the admixture of a second phase Al_3(Mn,Pd) to about 1%.

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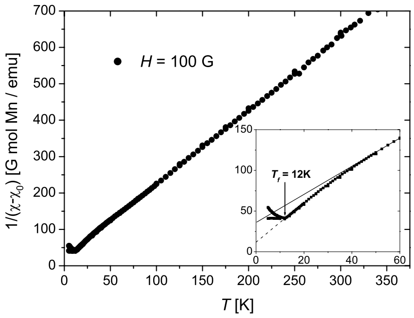

Fig. 1. $\chi^{-1}(T) = H/M(T)$ measured in an external field $H = 100$G. The inset inset emphasizes the temperature range below 60K

3. DC-Susceptibility

We measured the DC-susceptibility $\chi(T)$ of d-$Al_{69.8}Pd_{12.1}Mn_{18.1}$ with a DC-SQUID magnetometer between 340K and 2K, at fields between 50G and 5T. Figure 1 shows a representative curve of $\chi(T)$ measured at 100G between 5K and 340K.

We measuered a field cooled and a zero field cooled set of data. For these measurements the sample was cooled to low temperatures in zero field. The field was then switched on and the data were collected while increasing and subsequently reducing the temperature in the presence of the applied field. A preliminary analysis of the data shows that above 70K the data follows a Curie-Weiss type behaviour $\chi(T) = C/(T - \Theta) + \chi_0$ with a constant background contribution of $\chi_0 = 2.7 \cdot 10^{-4}$ emu/Gauss mol Mn. The Curie temperature $\Theta = -11$K indicates a predominant but weak antiferromagnetic coupling between the Mn d-moments. The Curie constant $C = 0.495$ emu/Gauss mol Mn is compatible with 10 to 25% of the Mn ions carrying a moment, depending on their valence state. The deviations of $\chi(T)$ from the Curie-Weiss behaviour below 50K may be interpreted as a precursor of a spin-glass freezing of the Mn moments[6], which occurs at $T_f = 12K$. This is evidenced by the minimum of $1/(\chi - \chi_0)$ at $T_f = 12$K and the different temperature dependence of the zero-field, and field-cooled data.

4. NMR Spectra

Field-sweep spectra were recorded at various temperatures and frequencies. Figure 2 shows the NMR field-sweep spectrum recorded by means of pulsed NMR at the excitation frequency $\nu_0 = 70$MHz. The main feature of the spectra is the very wide ^{27}Al signal with the characteristic broad distribution of quadrupolar wings observed in QC's. The ^{27}Al central transition $1/2 \leftrightarrow -1/2$ is by almost a factor of 10 broader than expected from considering a second order quadrupolar perturbation of the Zeeman line. It also reveals a shoulder at higher fields, suggestive for a second even broader signal. We assume that these two signals represent the ^{27}Al central transitions in two different Al-environments. This assumption is supported by the results of simulations of the spectra, shown in the inset of figure 2, and by recalling that the structure of d-$Al_{69.8}Pd_{12.1}Mn_{18.2}$ contains two different types of layers with different amounts of Mn. More detailed studies are necessary to clarify this issue.

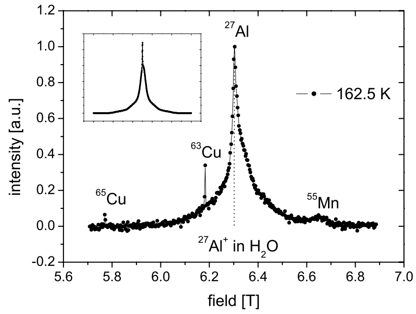

Fig. 2. NMR spectrum of d-AlPdMn at 167.5K measured with an excitation frequency of 70MHz, revealing signals due to ^{63}Cu, ^{65}Cu, ^{27}Al and ^{55}Mn nuclei. The inset shows a spectrum that was obtained by a computer simulation.

At higher fields the spectra exhibit a pronounced maximum around 6.64T, originating from the ^{55}Mn ions that carry no magnetic moment. The total integrated intensity of that signal, compared with the total integrated intensity of the ^{27}Al signal suggests that only 25±10% of the Mn-ions carry a local magnetic moment. From this result and from $\chi(T)$ we conclude that the magnetic Mn ions must be either in the Mn^{3+} or Mn^{4+} oxidation state. Below 100K and with decreasing temperature, the width of all the lines increases with a progressively increasing negative slope, indicating a slowing down of the moment fluctuations, as it is often observed in relation with spin-glass freezing processes.

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