Magnetic and electronic properties of the magnetically ordered quasicrystalline alloys $Al_{70-x}Pd_{15}Mn_{15}B_x$

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The magnetic susceptibility, magnetization and the temperature and field dependence of the Hall coefficient have been measured for the magnetically ordered icosahedral quasicrystals $Al_{70-x}Pd_{15}Mn_{15}B_x$ ($x=0,2,4,6,8,10$). The temperature dependence of the real and imaginary part of the complex ac susceptibility indicates re-entrant magnetic behavior and dc magnetization measurements are interpreted in the context of this behavior. The present Hall resistivity measurement shows the existence of both normal and anomalous Hall effects. The normal Hall coefficient is independent of temperature in all the alloys and changes from negative to positive with increasing boron concentration. This can be explained by the effects of $s-d$ hybridization. The anomalous Hall coefficient is also found to be temperature independent and has a compositional dependence correlated to the spin–orbit scattering rate.
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I. INTRODUCTION

Since the first discovery of quasicrystals in Al–Mn alloys,[1] the electronic and magnetic properties of these materials have been studied extensively. Although early reports indicated that most quasicrystals are weakly magnetic,[2,3] some recently developed materials have shown strong magnetic behavior. Yokoyama *et al.*[4,5] have reported the synthesis and characterization of ferromagnetic $Al_{70-x}Pd_{15}Mn_{15}B_x$ icosahedral alloys. These alloys have Curie temperatures between 300–500 K and exhibit saturation magnetization up to 18 emu/g which disappears after crystallization. In the present work the Hall effect, magnetic susceptibility and magnetization of $Al_{70-x}Pd_{15}Mn_{15}B_x$ have been investigated and these complement our previous transport studies of this series.[6]

II. EXPERIMENTAL TECHNIQUES

Single phase icosahedral $Al_{70-x}Pd_{15}Mn_{15}B_x$ alloys were prepared in the form of ribbons by the melt spinning method.[7] The quasicrystalline nature of these samples was confirmed from x-ray diffraction patterns. The temperature dependent ac susceptibility was measured by a standard mutual inductance technique[8] at a frequency of 127 Hz in an ac magnetic field of ~80 A/m. dc magnetization measurements were carried out using a conventional superconducting quantum interference device (SQUID) magnetometer. Hall effect measurements were performed using a six-point dc technique in applied magnetic fields up to 4.4×10^6 A/m.

III. RESULTS AND DISCUSSION

Figure 1 shows the room temperature ac susceptibility of $Al_{70-x}Pd_{15}Mn_{15}B_x$ ($x=0-10$). The measured susceptibility increases with increasing x and is consistent with previous magnetization, nuclear magnetic resonance (NMR) and ferromagnetic resonance (FMR)[4,5,9] results for these alloys. This behavior can be described by the model in which the ferromagnetic coupling originates from the Mn–B bonding.[5]

A typical measurement of the real and imaginary parts of the ac susceptibility for the present alloys is shown in Fig. 2. These measurements show a knee in $\chi'(T)$ and, to a lesser extent, in $\chi''(T)$, for $6\leqslant x\leqslant10$. No anomalous features have been observed for the susceptibility of samples with $x\leqslant4$. Results may be compared with the previous data of Hattori *et al.*[10] which showed that the ac susceptibility of $Al_{70}Pd_{15}Mn_{15}$ showed a cusp at low temperature which was attributed to the existence of a reentrant spin glass (RSG) transition. There is some ambiguity in the interpretation of features in the ac susceptibility[10,11] which can arise from experimental conditions such as the exciting field frequency

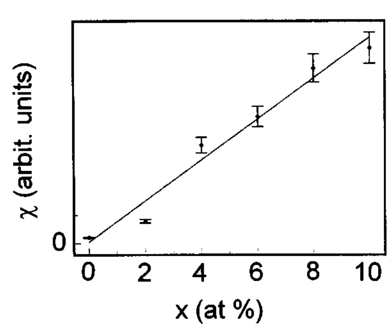

FIG. 1. The room temperature ac susceptibility of icosahedral $Al_{70-x}Pd_{15}Mn_{15}B_x$ alloys as a function of x.

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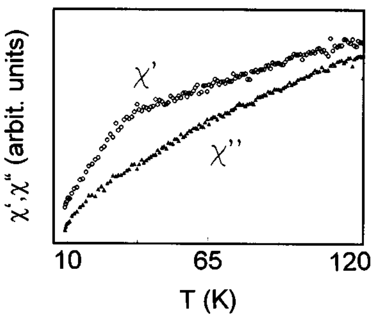

FIG. 2. The temperature dependence of zero-field cooled real and imaginary parts of the ac susceptibility for $Al_{60}Pd_{15}Mn_{15}B_{10}$.

and amplitude. The so-called Hopkinson peak[10] can obscure the characteristic behavior and, as a result, RSG features may be absent or may be shifted to a lower temperature. A comparison with magnetization data is often helpful in understanding the behavior of these materials. In most cases, the transition from ferromagnetism to the RSG state is defined by the onset of deviations from the plateau in the magnetization.[12] Results of these studies are illustrated in Fig. 3 for the present series of alloys. The knee associated with the RSG on lowering the temperature is clear for all alloys with $x \geqslant 2$. The observed temperature dependence is consistent with the behavior as described above.

In magnetically ordered materials both the ordinary Hall effect (OHE) and spontaneous Hall effect (SHE) are present. The latter is not restricted to ferromagnetic materials, but can appear in any material where strong magnetic interactions are present.[13] These two components combine to give the total Hall resistivity as (SI units):

$$\rho_H = R_0 B + R_s \mu_0 M \qquad (1)$$

where B is related to the applied magnetic field by $B = \mu_0(H+M)$, M the magnetization and R_0 and R_s are the ordinary and spontaneous Hall coefficients, respectively.

The field dependence of the Hall resistivity $\rho_H(B)$ for $Al_{70-x}Pd_{15}Mn_{15}B_x$ is shown in Fig. 4. This behavior is virtually independent of temperature. The initial slope gives

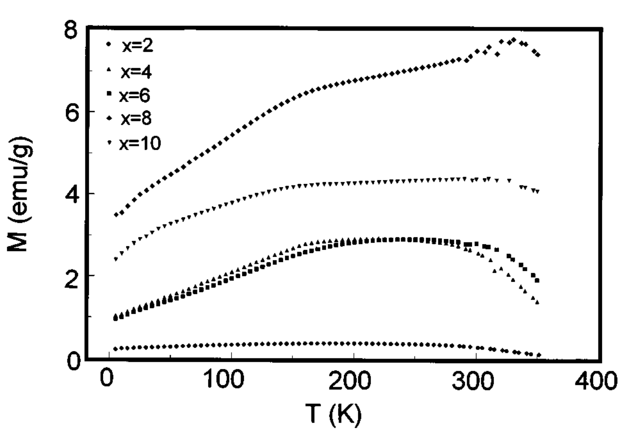

FIG. 3. Temperature dependence of the magnetization of $Al_{70-x}Pd_{15}Mn_{15}B_x$ as a function of x measured in an applied field of 8.0×10^3 A/m.

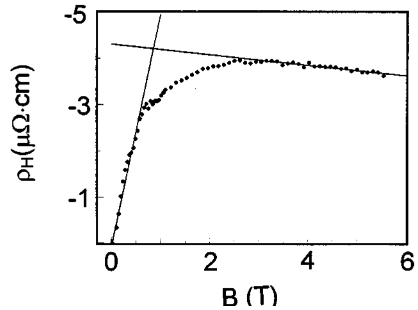

FIG. 4. Hall resistivity as function of field for $Al_{62}Pd_{15}Mn_{15}B_8$ at 4.2 K.

$$R_H = \left(\frac{\partial \rho_H}{\partial B}\right)_{B\to 0} \cong R_s \qquad (2)$$

and the slope at high fields, gives:[14]

$$R_H = \left(\frac{\partial \rho_H}{\partial B}\right)_{B\to \infty} \cong R_0 . \qquad (3)$$

Figure 5 shows that R_0 changes sign as a function of x. This behavior can be explained in terms of the effect of $s-d$ hybridization. This mechanism results from a shift of the Fermi energy and has been used to explain Hall effect results in amorphous alloys as it gives rise to regions of negative group velocity leading to positive R_0. In the present alloys the substitution of B for Al causes the Fermi level to increase as a result of a reduction in the unit cell volume[7] giving rise to the observed behavior.

In disordered alloys the SHE coefficient may be related to the temperature dependent resistivity, $\rho(T)$, as:

$$R_s(T) = a\rho(T) + b\rho^2(T) \qquad (4)$$

where the first term results from quasiclassical skew scattering and the second term results from the quantum mechanical side jump.[13,15] In amorphous alloys, because of the large number of scattering centers, the quadratic term is believed to dominate. In the present alloys R_s is constant in temperature while $\rho(T)$ changes by as much as about 20%.[7] Therefore, at least to first order, there is no measurable correlation between R_s and ρ. This has been observed in other systems and has been explained by different scattering mechanisms for ρ and ρ_H.[16] The x dependence of R_s is shown in Fig. 6. R_s is negative for all alloys and shows a systematic change

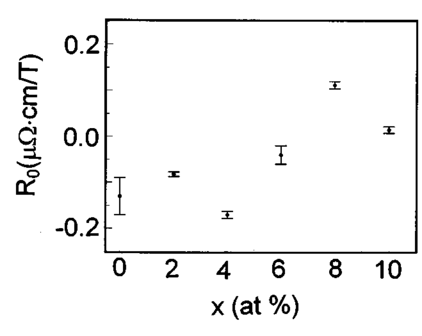

FIG. 5. The ordinary Hall coefficient of $Al_{70-x}Pd_{15}Mn_{15}B_x$ as a function of x at 4.2 K.

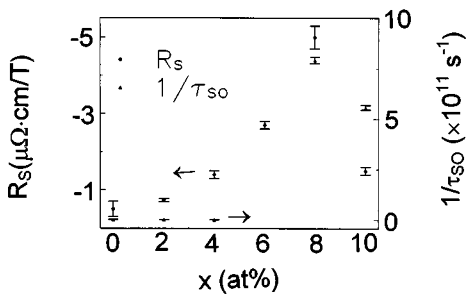

FIG. 6. The spontaneous Hall coefficient and spin–orbit scattering rate of $Al_{70-x}Pd_{15}Mn_{15}B_x$ as a function of x at 4.2 K.

with x. These results can be explained in terms of spin–orbit interactions. In Fig. 6 R_s is plotted along with the spin–orbit scattering rate as reported previously.[6,7] The magnitude of R_s increases with the scattering rate showing that the spin–orbit interaction is indeed the important contributor to the Hall resistivity although the mechanism by which the spin–orbit interaction influences the Hall resistivity is not obvious.

IV. CONCLUSIONS

Magnetic studies of icosahedral $Al_{70-x}Pd_{15}Mn_{15}B_x$ (x =0,2,4,6,8,10) as presented here indicate that these materials are ferromagnetically ordered and show a re-entrant spin glass transition at low temperature. The dc magnetization results indicate a higher transition temperature as expected on the basis of the effects of the Hopkinson peak on ac susceptibility data. The interpretation of these results is consistent with previous magnetic,[4] NMR,[5] and FMR[9] studies.

The data presented show that the Hall resistivity ρ_H is the result of two contributions; the normal term R_0B, due to the Lorentz force and anomalous term $\mu_0 M_s R_s$ due to the spin–orbit interaction. R_0 is independent of temperature but changes sign as a function of x and can be explained by the effects of $s-d$ hybridization. R_s is also found to be temperature independent and suggests that the scattering mechanisms responsible for $R_s(T)$ and $\rho(T)$ are different. The compositional dependence of R_s is similar to that of the spin–orbit scattering rate and indicates that this is the responsible interaction for the spontaneous Hall behavior in these materials.

ACKNOWLEDGMENTS

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