March 1984

Spin-glass-like freezing in disordered $MnPd_3$ and $CrPd_3$ alloys

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Spin-glass-like freezing in disordered $MnPd_3$ and $CrPd_3$ alloys[a)]

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We report studies of dc and ac susceptibility and high-field magnetization on disordered samples of $MnPd_3$ and $CrPd_3$. Both samples undergo spin-glass-like transitions, as is evident from susceptibility peaks at 45 and 37 K, respectively. The effective paramagnetic moments deduced from Curie–Weiss fits of the susceptibility in the paramagnetic region are $0.38\mu_B$/Mn and $0.4\mu_B$/Cr. Both materials show saturation effects at high applied fields and a flat $\chi(T)$ below the spin-freezing temperature in the field-cooled state. The data are discussed in terms of a cluster-glass model with local antiferromagnetic interactions.

PACS numbers: 75.30.Hx, 75.30.Kz

INTRODUCTION

Pure palladium is strongly paramagnetic. But ordered $FePd_3$ and $CoPd_3$ are ferromagnetic.[1] Ordered $MnPd_3$ is antiferromagnetic[2] with $T_C = 170$ K. Pd(Mn) alloys with a very low Mn concentration (0.05 to 3.0 at. %) are ferromagnetic,[3–5] with Mn having a so-called "giant moment" ($\mu_{\text{eff}} \sim 7.5\mu_B$). A spin-glass phase appears at a higher Mn concentration (> 4 at. %). Mn impurities form clusters with a strong antiparallel coupling between Mn nearest neighbors.[6,7] In the case of Pd(Cr) alloys, on the other hand, Cr has only a weak magnetic moment in the dilute limit[3,8] (Kondo temperature ~ 200 K). The Cr moment becomes stable ($\mu_{\text{eff}} \sim 4.8\mu_B$) for a Cr concentration that is large enough to provide a suitable Cr local environment. Resistivity measurements of Pd(Cr) alloys have shown spin-glass-like behavior for Cr concentrations between 11 and 18 at. %.[9] Recently we have been studying the occurrence of itinerant magnetism in ordered compounds of the form TPd_3 and related compounds, where T represents a 3D magnetic element.[10] In this paper we report on the magnetic properties of relatively concentrated *disordered* $MnPd_3$ and $CrPd_3$ alloys.

EXPERIMENTAL METHODS

Alloys of $MnPd_3$ and $CrPd_3$ are prepared by arc-melting the constituent elemental metals (purity: 99.9 + % Mn, 99.999% Cr, and 99.99% Pd) to obtain "buttons" of approximate mass of two grams. The samples are arc-melted several times to obtain a good mixing of the constituents. The "buttons" are then cleaned with proper etchants, distilled water, and then with methanol. They are then heated in a low-pressure argon atmosphere ($\sim 1/4$ atm) in quartz tubes at a temperature of 700 °C for 15 days and quenched in air to achieve homogeneity.

The crystal structures and lattice parameters are determined by a horizontal plane diffractometer, calibrated by known quartz lines. The dc susceptibility is measured by the Faraday method, and the ac susceptibility is measured at 270 Hz in a field of 0.3 Oe. High-field magnetization measurements are made using a homemade vibrating-sample magnetometer[11] which employs an 80-kOe superconducting magnet.

RESULTS AND DISCUSSION

Our $MnPd_3$ sample is analyzed to have a disordered face-centered-cubic (fcc) crystal structure with the lattice parameter, $a = 3.890 \pm 0.003$ Å. Results of the Faraday susceptibility at 8.0 and 1.2 kOe and the ac susceptibility of $MnPd_3$ are shown in Fig. 1. χ_{dc} at 1.2 kOe and χ_{ac} have broad peaks near 45 K. Sharp susceptibility peaks at a temperature where no long-range magnetic order is present are characteristic of spin-glass systems.[12] For example, these types of peaks have been observed in Au(Fe) alloys[13] and in the disordered $MnNi_3$ system.[14] The peak in χ_{dc} for the 8.0-kOe field run is flattened. A Curie–Weiss fit in the temperature range 80–300 K gives a Curie constant C of 0.0177 emu/gK and a Weiss temperature θ of -50 K. The Curie constant corresponds to a moment of $0.38\mu_B$ per Mn atom. This small paramagnetic moment, along with a negative θ value, indicates that at least some of Mn moments are coupled antiferromagnetically. This is consistent with the model that the orgin of the spin-glass phase in dilute Pd(Mn) alloys is a competition between the antiparallel coupling of the Mn nearest neighbors and the long-ranged ferromagnetic giant moment interactions.[3] Low-field (10 Oe) magnetization measurements in disordered $MnPd_3$ were reported by Rainford[15] as showing a "sharp cusp" at $T_g = 30$ K. In addition

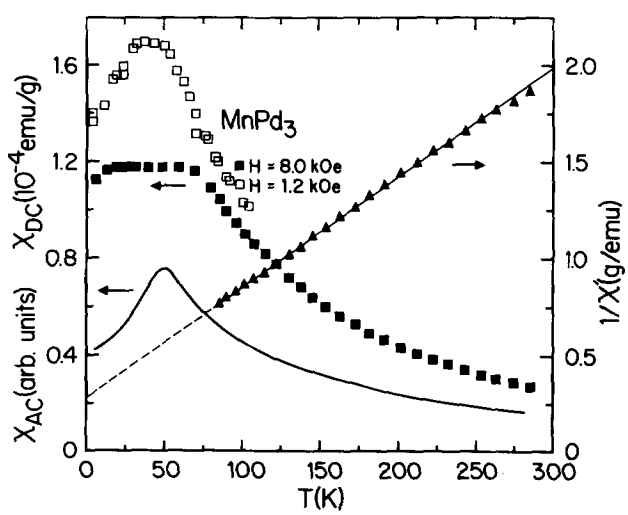

FIG. 1. $\chi_{\text{dc}}(T)$, $1/\chi'_{\text{dc}}(T)$, and $\chi_{\text{ac}}(T)$ for $MnPd_3$ ($\chi'_{\text{dc}} = \chi_{\text{dc}} - \chi_0$, where χ_0 is the temperature-independent part of χ_{dc}).

[a)] Research supported by NSF Grant DMR-8110520.

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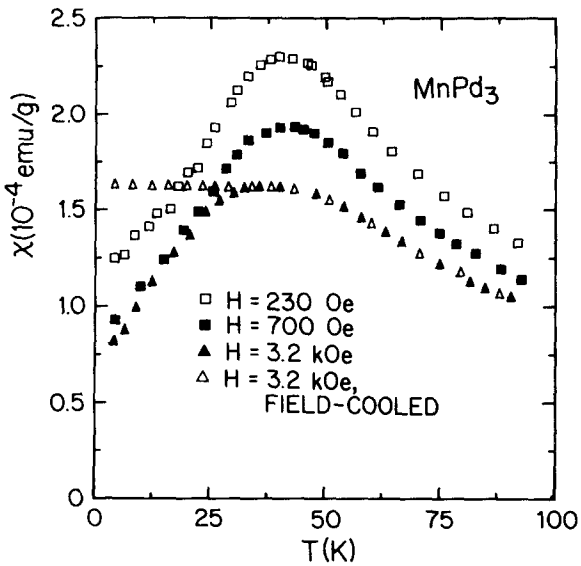

FIG. 2. $\chi_{dc}(T)$ for $MnPd_3$ at different applied fields.

Rainford reports neutron measurements which show only short-range magnetic order below T_g.[15] Our T_g value of 45 K suggests perhaps a somehwat higher degree of short-range order in our sample.

Saturation effects are observed in $MnPd_3$ at finite fields resulting in a broader and smaller maximum with an increase of the applied field, as shown in Fig. 2. Similar behavior has been observed in Au(Fe) alloys.[13] A rounded susceptibility maximum, even in the zero-field limit, can be explained in terms of a "distribution of freezing temperatures" due to different cluster environments and coupling strengths.[13] At 3.2 kOe, a flat $\chi(T)$ is observed below the spin-freezing temperature in the filed-cooled state (Fig. 2), which is another characteristic of a spin glass.

Figure 3 shows the field-cooled hysteresis loop at 4.2 K for $MnPd_3$. The loop is slightly open at the maximum positive field. It shows no noticeable displacement from the origin. The remanent magnetization is 0.044 μ_B/Mn at 4.2 K. The remanence drops off sharply with the increase of temperature, and goes to zero above the spin-glass transition.

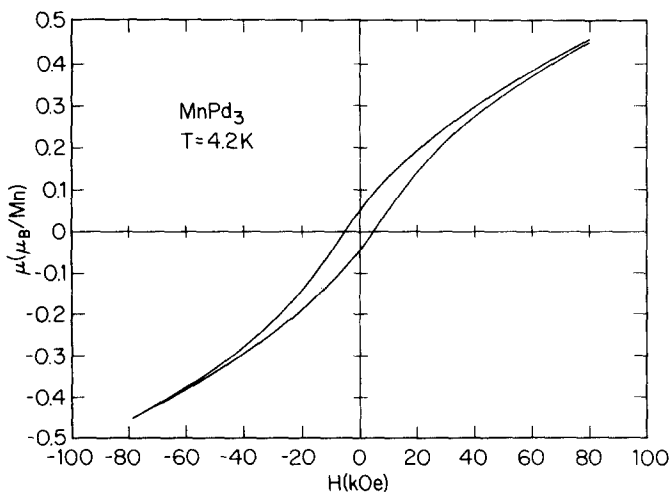

FIG. 3. Hysteresis loop at 4.2 K for $MnPd_3$.

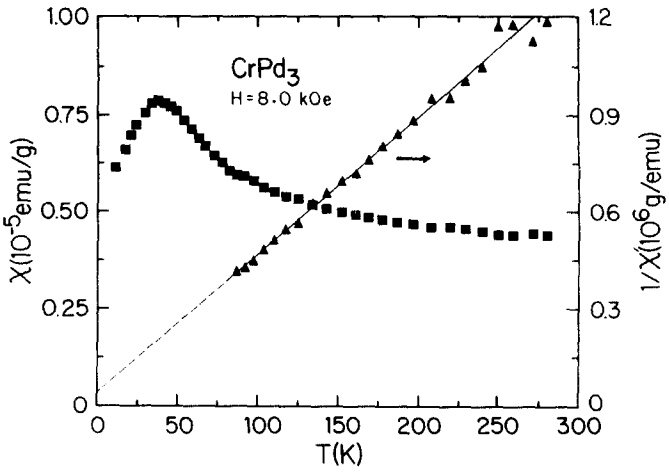

FIG. 4. $\chi_{dc}(T)$ and $1/\chi'_{dc}(T)$ for $CrPd_3$ ($\chi'_{dc} = \chi_{dc} - \chi_0$).

Also, as the sample is field-cooled from 150 to 4.2 K in 80 kOe and the field is cycled between $\pm$ 10 kOe, the resulting hysteresis loop is displaced *upward* from the orgin. These characteristic behaviors have been observed in a number of spin-glass systems.[12,16] Theoretical work aimed at understanding hysteretic properties of spin glasses has been published recently by Soukoulis *et al.*[17] These workers employed mean-field Ising and Heisenberg models and computed hysteresis loops below T_g whick look very similar to our data. It was found that in order to achieve some squareness in the loops a ferromagnetic tendency in the average exchange interactions (J_0) had to be included. They also found that in order to achieve an upward displacement of the loop, one for which the magnetization goes positive while the field is still negative, they had to assume a Heisenberg interaction and include anisotropy of the Dzyaloshinsky–Moriya (DM) form. Thus to the extent that it can be applied to our fairly complex alloys, the theory suggests that in $MnPd_3$ J_0 is rather small and a relatively large DM anisotropy is present.

The Bravais lattice of $CrPd_3$ is also fcc with $a = 3.589 \pm 0.003$ Å. Figure 4 shows the measured Faraday

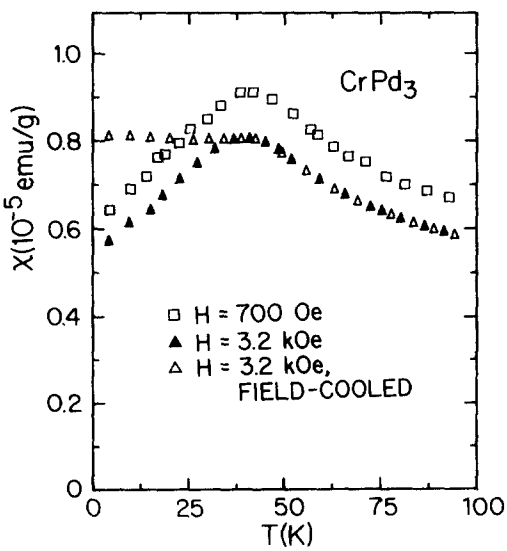

FIG. 5. $\chi_{dc}(T)$ for $CrPd_3$ at different applied fields.

susceptibility versus temperature for $CrPd_3$. There is a peak in susceptibility at 37 K. A Curie–Weiss fit in the temperature range of 80–300 K gives the values, $C = 0.235 \times 10^{-3}$ emu/gK and $\theta = -10$ K. The derived magnetic moment per Cr atom is only 0.04 μ_B. The field dependence of the susceptibility peak is shown in Fig. 5. As in the case of $MnPd_3$, the susceptibility peak of $CrPd_3$ becomes larger and sharper as the applied field is lowered. A small value of effective Cr moment and a negative θ value may indicate the origin of the spin-glass-like behavior in $CrPd_3$ is the same as in $MnPd_3$. But an alternate argument might be advanced, according to which the Cr moment is highly dependent on local environment with "weak" clusters forming which are then coupled via the conduction electrons at longer distances. This model could account for the extremely small moment per Cr atom in the paramagnetic state.

Ordered $MnPd_3$ is antiferromagnetic. But disordered $MnPd_3$ and $CrPd_3$ form what might best be described as cluster glasses. It is clear that a detailed understanding of these disordered materials will be dependent upon microscopic information on local environment effects on moment formation and coupling, and upon a detailed knowledge of the chemical short-range order.

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