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Spin glass behavior in $FeAl_2$

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Magnetic and transport measurements indicate $FeAl_2$ to be an ordered intermetallic spin glass, with canonical behavior including a susceptibility cusp at $T_f = 35$ K and frequency-dependent susceptibility below T_f. The field-cooled and zero-field-cooled magnetization diverge below T_f, with hysteresis characteristic of a spin glass. A resistivity minimum just above T_f is explained in terms of coherent magnetic scattering. This behavior is common to spin glasses with short-range interactions among *f*-electron moments and indicates a similar spin configuration in these materials.

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I. INTRODUCTION

An increasing recent interest in spin glasses has been focused on the observation of spin glasses having ordered crystal structures,[1–4] as well as the possibilities of quantum spin glasses[5] and related effects in these materials. Spin-glass behavior is identified most prominently with dilute metallic alloys, in which the long-range Ruderman-Kittel-Kasaya-Yosida (RKKY) interaction dominates, leading to canonical spin-glass effects such as a strong frequency dependence to the magnetic behavior.[6] Only recently has spin-glass behavior been identified in ordered systems; to date such behavior has been identified in insulating oxides and *f*-electron metals. In an ordered spin glass, geometrical frustration due to the lattice configuration must dominate. However, site-occupation disorder has also been shown to be important in the known *f*-electron ordered spin glasses. In this paper, we demonstrate spin-glass behavior in an ordered transition-metal alloy $FeAl_2$. This observation in a *d*-electron intermetallic contrasts the previous observation of this behavior in *f*-electron intermetallics. From magnetic and transport measurements we show that the behavior is similar to that of the *f*-electron ordered spin glasses.

In $Fe_{1-x}Al_x$ alloys, studies of the magnetic behavior have focused on compositions near $x = 0.3$, that exhibit reentrant spin-glass behavior.[7,8] $Fe_{70}Al_{30}$ is ferromagnetic below 400 K, then becomes superparamagnetic at around 170 K, and finally freezes into a spin-glass state at 92 K.[7,8] While there is continued debate about the nature of the ordered and glassy states in this system,[8,9] it has been found that a small number of sites with both ferro- and antiferromagnetic bonds can destroy an ordered ferromagnetic state below a characteristic temperature.[10] For higher Al concentrations, $Fe_{1-x}Al_x$ alloys form a series of complex ordered crystal structures, generally with weak magnetic behavior. While no magnetic or spin-glass transitions have been identified in these alloys, a recent site-diluted Ising model has suggested the existence of a spin-glass phase in disordered alloys of this type.[11] In the dilute Fe limit, Fe-Al alloys are nonmagnetic, and there is some uncertainty as to whether the Fe ions lose their moment through spin fluctuations or hybridization.[12]

II. SAMPLE PREPARATION

The sample studied here was prepared from the elemental constituents in an induction furnace under a partial argon atmosphere and homogenized by annealing in a vacuum at 850 °C for four days. Cu $K\alpha$ x-ray analysis of a powdered sample from this ingot showed a pattern identical to that of the reported structure.[13–16] $FeAl_2$ has a triclinic unit cell with $a = 0.4878$, $b = 0.6461$, and $c = 0.8800$ nm; $\alpha = 91.75°$, $\beta = 73.27°$, and $\gamma = 96.89°$. Using these parameters and the atom positions identified by Corby and Black,[13] intensities of the 250 strongest peaks between $2\theta = 10°–110°$ were calculated using standard methods.[17] Calculated intensities provided a good match for all observed peaks, indicating a single-phase sample. The triclinic $FeAl_2$ cell has 18 sites, including 10 Al sites, 5 Fe sites, and 3 sites having mixed Al and Fe occupation.[13] In our calculation we assigned each mixed site 67% occupancy by Al, corresponding to the $FeAl_2$ composition. While an *R* value was difficult to calculate due to overlap within the x-ray spectrum, the calculated intensities gave a good visual match to the spectrum, with the largest difference a factor of 2 in intensity without employing thermal factors.

Part of our initial interest in this material was in looking for $FeAl_2$ in the tetragonal $MoSi_2$ structure, predicted to occur as a hybridization-gap semiconductor by Weinert and Watson,[18] but we have seen no evidence of such a structure for our preparation conditions.

III. MAGNETIC MEASUREMENTS

All magnetic measurements were carried out using a commercial superconducting quantum interference device magnetometer (Quantum Design). The ac susceptibility (χ) was obtained with an ac field amplitude of 1 Oe and a frequency of 125 Hz. The results, shown in Fig. 1, exhibit a cusp at 35 K with a maximum at lower temperatures. Above the cusp, the data follow a Curie-Weiss law $\chi = C/(T - \theta)$, as shown by the straight-line fit to $1/\chi$ in the inset to Fig. 1. From this fit we obtain an effective magnetic moment $p_{eff} = 2.55\mu_B$ per iron, indicating strong local-moment magnetism in this material. The extrapolated Weiss temperature $\theta \approx -42$ K indicates antiferromagnetic interactions, presumably superex-

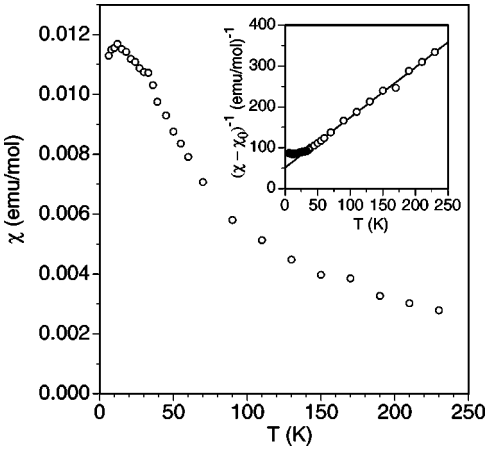

FIG. 1. The real part of the ac susceptibility (χ) obtained at 125 Hz. Inset: inverse χ vs temperature, with the straight line corresponding to a Curie-Weiss fit described in the text.

change mediated by Al. An early measurement[19] indicated a somewhat smaller moment $p_{eff}=1.32$ per iron, but this report is also at variance with recent measurements[20] of Fe_2Al_5 and Fe_4Al_{13} ($p_{eff}=0.73$ and 0.44, respectively).

To further understand the apparent transition at 35 K, we measured the frequency dependence of the susceptibility in the range 1.25–1250 Hz. As shown in Fig. 2, the susceptibility becomes frequency dependent just above the cusp at 35 K. At lower temperatures, χ has a reduced amplitude and the broad maximum shifts to higher temperatures with increasing frequency. Such behavior is commonly seen in classical spin-glass systems,[6] and we associate the observations with a spin-glass transition having a freezing temperature $T_f=35$ K. The low-temperature maximum in χ is not typical of spin-glass systems, but we believe that some Fe sites in the structure are more weakly coupled magnetically than others, so that they begin to freeze at lower temperatures, leading finally to the decrease in χ below 12 K. The final decrease of χ shows that eventually all sites participate in the spin-glass condensate.

Measurements of dc magnetization under zero-field-cooled conditions (M_{ZFC}) and under field-cooled conditions (M_{FC}) demonstrate the irreversible behavior of the spin-glass state. For the curves shown in Fig. 3, the sample was

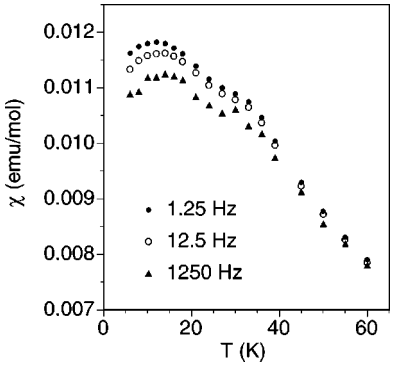

FIG. 2. Frequency dependence of the susceptibility measured at 1.25, 12.5, and 1250 Hz.

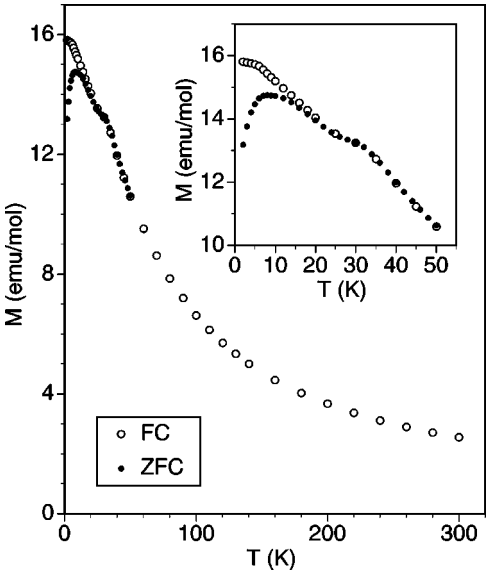

FIG. 3. Field-cooled (open circles) and zero-field-cooled (dots) magnetization in a dc field of 0.1 T. The inset displays both curves on an expanded scale.

cooled in fields of zero and 0.1 T, followed by magnetization measurements upon warming at 0.1 T. M_{ZFC} falls below M_{FC} at temperatures below T_f, behavior observed in typical spin-glass materials.[6] The canonical behavior associated with spin glasses, constant M_{FC} while M_{ZFC} drops toward zero, is observed below about 6 K. The structure observed between 6 K and T_f is consistent with some sites in the $FeAl_2$ lattice remaining decoupled from the condensate in this temperature range, as described above. For the zero-field-cooled case, we observed hysteresis at low temperatures. A trace of the magnetization versus H at 4 K is shown in Fig. 4, illustrating the relatively small coercivity commonly observed in spin glasses.[6]

A Curie-Weiss fit to the M/H results above T_f yields $p_{eff}=2.8$ and $\theta=-38$ K, agreeing with the susceptibility results and indicating no tendency for saturation. This result confirms that the magnetic behavior is due to atomic-scale moments rather than superparamagnetic clusters or small inclusions of a ferromagnetic phase below the detectability limits of the x-ray measurement.

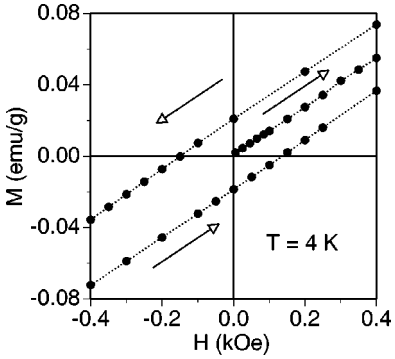

FIG. 4. Low-field region of the hysteresis loop for $FeAl_2$ at 4 K.

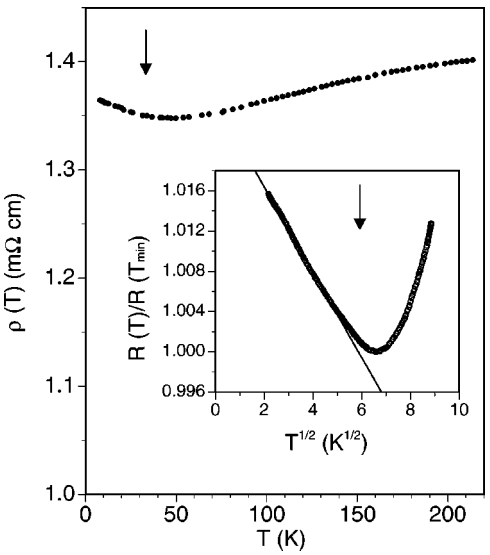

FIG. 5. Temperature variation of resistivity for $FeAl_2$. The arrow indicates the freezing temperature T_f. The inset shows the resistivity normalized with the value at T_{min} versus $\sqrt{T}$, with the data below 25 K following a $\sqrt{T}$ dependence.

IV. TRANSPORT MEASUREMENTS

The sample for transport measurements was cut from the same ingot used for magnetic studies. The resistivity (main plot in Fig. 5) was obtained with a dc technique; the results are dominated by a large temperature-independent term of about 1.3 $m\Omega$ cm. A resistivity minimum is seen at about 43 K, just above the freezing temperature, with no additional anomaly seen near the temperature of the susceptibility maximum 12 K. No sharp features are observed at T_f, confirming the absence of true magnetic ordering at this temperature. Because of the relatively small resistivity changes, we obtained additional data using an ac bridge having greater relative accuracy, as shown in the inset to Fig. 5. While a resistance minimum could be an indication of the Kondo effect arising from the strong magnetic impurity scattering,[6,21] the results do not fit to the logarithmic behavior, ρ increasing as $-\ln T$, as expected for such a model. Instead, the low-temperature resistivity scales well to $-\sqrt{T}$, as plotted in the figure inset.

V. DISCUSSION

In the $FeAl_2$ structure, each Fe site is surrounded by a cage of 10–11 nearest neighbors consisting mostly of Al, with an average 1.6 neighboring Fe sites and 1.8 neighboring mixed sites. In the cubic $Fe_{1-x}Al_x$ mixed phases observed for larger Fe concentrations, the magnetic moment is significantly reduced at Fe sites having fewer than four near-neighbor Fe atoms.[7] On this basis one would expect $FeAl_2$ to be a low-moment structure. Furthermore, the Fe-Al distances are the shortest for each Fe site in $FeAl_2$ (average 0.25 vs 0.28 nm mean Fe-Fe distance), indicating the importance of Fe-Al hybridization, which can lead to a nonmagnetic configuration as observed in the hybridization-gap semimetal[22] Fe_2VAl and in the Fe-Al quasicrystalline phases.[23]

One can also compare Al-rich Mn-Al alloys,[24] in which the stable, ordered $MnAl_6$ structure is nonmagnetic due to *s-d* hybridization, while metastable cubic alloys have high moments and are spin-glass-like, with the quasicrystalline and liquid alloys somewhere in-between. There is currently considerable interest in the unusual electronic features observed in ordered hybridization-gap alloys composed of magnetic constituents.[25] $FeAl_2$, however, is a system with the highest moment of the Fe-Al alloys, apparently stabilized by entropy over the $MoSi_2$ structure[18] rather than hybridization. In this case we have the interesting result that the encaged-Fe structure leads to local-moment behavior and spin-glass behavior on an ordered lattice.

The spin-glass behavior in $FeAl_2$ can be due to both frustration on the complex lattice structure of this material, as well as disorder due to occupation of the mixed sites. This case is thus quite similar to that of the Ce and U intermetallics that exhibit spin-glass behavior, such as[1,2] U_2PdSi_3 and URh_2Ge_2. Normally, the moments in conducting transition-metal systems are more extended and do not exhibit the behavior attributed to rare-earth and actinide systems. $FeAl_2$, however, clearly shows local-moment behavior leading to a spin-glass freezing at low temperatures.

The resistivity of this material is large, comparable in size to that of the *f*-electron spin-glass materials,[1] and due to scattering from local moments in this system. The resistivity minimum we attribute to the development of short-range spin correlations in the spin-glass phase. The low-temperature resistivity rise does not have the logarithmic behavior characteristic of a Kondo effect, nor does it follow an activated curve, as would be expected for a narrow gap at the Fermi level; such an observation might be an indication of quantum spin-glass behavior.[5]

A resistivity minimum near T_f has been observed in a number of other concentrated *d*- and *f*-electron spin glasses. Similar behavior was observed near the spin-glass transition in NiMn and NiMnPt alloys,[26,27] both for reentrant and nonreentrant compositions, and somewhat above the transition in U_2PdSi_3.[1] This behavior is in contrast to the resistivity maximum typically observed in traditional diluted spin glasses dominated by the RKKY interaction.[21] For the NiMn case the behavior has been associated with remanent domains that exist into the spin-glass state. For the present case there is no tendency for ferromagnetic domain formation, but above T_f there will be antiferromagnetic correlations corresponding to the observed negative Weiss temperature, and these become progressively enhanced and frozen in below T_f. Enhancement of spin-spin correlations at the wave vector $2k_f$ gives an increase in the coherent magnetic scattering, as shown for amorphous ferromagnets,[28] which exhibit a similar resistivity minimum. Such an enhancement in the spin correlation will be particularly significant in conducting materials such as these, with short-range interactions leading to antiferromagnetic correlations at low temperatures. The resistivity shows a minimum somewhat above the freezing temperature observed by low-frequency techniques, however it is well known that high-frequency measurements such as the resistivity overestimate T_f by neglecting low-frequency fluctuations. Indeed, the frequency-dependent susceptibility re-

ported above exhibits just such an effect. Thus we deduce the spin configuration in $FeAl_2$ to be similar to that of the *f*-electron ordered spin glasses.

VI. CONCLUSIONS

We have demonstrated that $FeAl_2$ is a high-spin material that exhibits spin-glass behavior, with a freezing temperature of $T_f = 35$ K. The magnetic hysteresis observed below T_f is similar to that exhibited by canonical spin glasses. A minimum in the resistivity near T_f indicates that the spin configuration has features common to other concentrated ordered spin glasses.

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[1] D.X. Li, Y. Shiokawa, Y. Homma, A. Uesawa, A. Dönni, T. Suzuki, Y. Haga, E. Yamamoto, T. Honma, and Y. Onuki, Phys. Rev. B **57**, 7434 (1998).

[2] S. Sullow, G.J. Nieuwenhuys, A.A. Menovsky, J.A. Mydosh, S.A.M. Mentink, T.E. Mason, and W.J.L. Buyers, Phys. Rev. Lett. **78**, 354 (1997).

[3] Cheng Tien, Chung Hsing Feng, Ching Shui Wur, and Jenq Jong Lu, Phys. Rev. B **61**, 12 151 (2000); A. Kimura, D.X. Li, and Y. Shiokawa, Physica B **281&282**, 247 (2000).

[4] M.J.P. Gingras, C.V. Stager, N.P. Raju, B.D. Gaulin, and J.E. Greedan, Phys. Rev. Lett. **78**, 947 (1997).

[5] Subir Sachdev and N. Read, J. Phys.: Condens. Matter **8**, 9723 (1996); R. Oppermann and B. Rosenow, Phys. Rev. Lett. **80**, 4767 (1998).

[6] K. Binder and A.P. Young, Rev. Mod. Phys. **58**, 801 (1986).

[7] Prabodh Shukla and Michael Wortis, Phys. Rev. B **21**, 159 (1980).

[8] Wei Bao, S. Raymond, S.M. Shapiro, K. Motoya, B. Fȧk, and R.W. Erwin, Phys. Rev. Lett. **82**, 4711 (1999).

[9] A. Das, L.K. Paranjpe, S. Honda, S. Murayama, and Y. Tsuchiya, J. Phys.: Condens. Matter **11**, 5209 (1999).

[10] G.N. Parker and W.M. Saslow, Phys. Rev. B **38**, 11 718 (1988).

[11] E. Mina, A. Bohorquez, Ligia E. Zamora, and G.A. Perez Alcazar, Phys. Rev. B **47**, 7925 (1993); J. Restepo, G.A. Perez Alcazar, and J.M. Gonzalez, J. Appl. Phys. **83**, 7249 (1998); J.A. Plascak, Ligia E. Zamora, and G.A. Perez Alcazar, Phys. Rev. B **61**, 3188 (2000).

[12] D. Guenzburger and D.E. Ellis, Phys. Rev. Lett. **67**, 3832 (1991); P.G. Gonzales, L.A. Terrazos, H.M. Petrilli, and S. Frota-Pessoa, Phys. Rev. B **57**, 7004 (1998).

[13] R.N. Corby and P.J. Black, Acta Crystallogr., Sect. B: Struct. Crystallogr. Cryst. Chem. **29**, 2669 (1973).

[14] Mirek Úředníček and J.S. Kirkaldy, Z. Metallkd. **64**, 899 (1973).

[15] M. Khaidar, C.H. Allibert, and J. Driole, Z. Metallkd. **73**, 433 (1982).

[16] B. Predel, in *Phase Equilibria, Crystallographic and Thermodynamic Data of Binary Alloys* (Springer-Verlag, Berlin, 1991), Vol. 5.

[17] *International Tables for Crystallography*, edited by A. J. C. Wilson (Kluwer, Dordrecht, 1996), Vol. C.

[18] M. Weinert and R.E. Watson, Phys. Rev. B **58**, 9732 (1998).

[19] M.A. Taylor, Proc. Phys. Soc. London **78**, 1244 (1961).

[20] F. Müller, M. Rosenberg, W. Liu, and U. Köster, Mater. Sci. Eng., A **134**, 900 (1991).

[21] P. L. Rossiter, *The Electrical Resistivity of Metals and Alloys* (Cambridge University, Cambridge, England, 1991).

[22] C.S. Lue, Joseph H. Ross, Jr., C.F. Chang, and H.D. Yang, Phys. Rev. B **60**, R13 941 (1999).

[23] R.A. Brand, J. Pelloth, F. Hippert, and Y. Calvayrac, J. Phys.: Condens. Matter **11**, 7523 (1999).

[24] J. Hafner and M. Krajci, Phys. Rev. B **57**, 2849 (1998).

[25] L. Degiorgi, Rev. Mod. Phys. **71**, 687 (1999).

[26] S. Senoussi and Y. Öner, Phys. Rev. B **28**, 455 (1983).

[27] Hüseyin Zafer Durusoy and Yildirhan Öner, Phys. Rev. B **42**, 6831 (1990).

[28] A. Fert and R. Asomoza, J. Appl. Phys. **50**, 1886 (1979).