PHYSICAL REVIEW B VOLUME 25, NUMBER 7 1 APRIL 1982

Spin-glass—ferromagnetic transitions and critical lines in $Fe_xPd_{82-x}Si_{18}$ metallic glasses

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(Received 16 November 1981)

Magnetization measurements on a series of $Fe_xPd_{82-x}Si_{18}$ metallic glasses are reported, $9 \leq x \leq 20$. The experimental results resemble closely the predictions of the Sherrington-Kirkpatrick model. Scaling analysis is applied to the Curie and the spin-glass—ferromagnetic transitions. It shows that both are similar in nature with, however, different critical exponents. The magnetic phase diagram of amorphous $Fe_xPd_{82-x}Si_{18}$ is obtained. It differs significantly from the one obtained previously on the basis of low-field dc and ac measurements alone.

The magnetic properties of Fe-Pd-Si metallic glasses were studied extensively by several authors during the last decade.[1–7] Recent work[5–7] on the (amorphous) a-$Fe_xPd_{82-x}Si_{18}$ system $(2 \leq x \leq 25)$ revealed a gradual transition with increasing Fe content from spin-glass (SG), for $x \lesssim 5$, to ferromagnet (FM) at $x \gtrsim 20$. Intermediate compositions $(5 < x < 20)$ are spin-glass-like at low temperature, becoming ferromagnetic with increasing temperature.[5–7] A magnetic phase diagram of a-$Fe_xPd_{82-x}Si_{18}$, $2 \leq x \leq 25$, was constructed[6] by plotting Curie temperatures T_C and SG "freezing" temperatures T_{SG} versus composition. Arrott plots were used to obtain T_C while T_{SG} was determined by the position of a broad maximum in the thermal variation of low-field ($H = 100$ Oe) dc magnetization and, for a-$Fe_2Pd_{80}Si_{18}$ and a-$Fe_5Pd_{77}Si_{18}$, also by ac susceptibility. Thus obtained, *both* T_C and T_{SG} increase with x and the low-temperature FM-SG phase boundary, presumably around $x = 15$, could not be determined.[6]

The purpose of the present report is to establish and examine the FM-SG critical line in the magnetic phase diagram of a-$Fe_xPd_{82-x}Si_{18}$. The analysis of the experimental $M(H,T)$ data follows a procedure which has been employed recently[8–10] to determine the nature of magnetic transitions in a-$(M_{1-x}M'_x)_{75}P_{16}B_6Al_3$ alloys ($M =$ Fe,Co; $M' =$ Mn,Ni) and to obtain the critical exponents. It was concluded[8–10] that the FM-SG transition in those alloys is continuous, resembling the Curie transition, and that the phase boundary is described qualitatively by the Sherrington-Kirkpatrick (SK) model.[11] Similar conclusions are reached in the present work and the previously reported phase diagram[6] is modified.

The a-$Fe_xPd_{82-x}Si_{18}$ alloys differ from the a-$(M_{1-x}M'_x)_{75}P_{16}B_6Al_3$ system[8–10] in that they contain a single metalloid element and up to only 25 at. % of, essentially, a single magnetic element, as the paramagnetism of Pd is strongly suppressed in the glass.[1–7] Being available in the glassy state over a wide range of compositions,[5–7] the a-$Fe_xPd_{82-x}Si_{18}$ alloys offer advantages over other materials for the study of FM-SG transitions. Although evidence for the occurrence of both SG and FM phases has been reported in a number of other systems,[12] these have not been closely examined with regard to their critical behavior.

Ribbons of $Fe_xPd_{82-x}Si_{18}$ metallic glasses $(9 \leq x \leq 20)$ were prepared by rapid quenching from the melt as described in detail elsewhere.[5,6] The temperature dependence (4.2—320 K) of the magnetization M was measured at a number of increasing fields, from 40—9900 Oe, using a vibrating-sample magnetometer. The samples were cooled in zero field between runs, following warm-up well into the paramagnetic (PM) regime to eliminate hysteresis effects. The field was applied parallel to the plane of the ribbon to minimize demagnetization.

Figure 1 shows the temperature dependence of the magnetization of a-$Fe_{12}Pd_{70}Si_{18}$ at several fields. Demagnetization effects which become significant only at fields below ~ 100 Oe—as indicated by the distorted shape of the 40- and 90-Oe curves in Fig. 1—have not been corrected for. The broad maximum which shows in the M vs T curves at low fields (Fig. 1) becomes less pronounced with increasing field; its position shifting to lower temperature it finally disappears above ~ 2000 Oe for a-$Fe_{12}Pd_{70}Si_{18}$ (Fig. 1). Similar behavior was observed in other a-$Fe_xPd_{82-x}Si_{18}$ alloys that are spin-glass-like at low temperature,[5,6] $x < 15$. The composition dependence of the thermal variation of the magnetization is illustrated in Fig. 2 showing M vs T/T_C curves of several $Fe_xPd_{82-x}Si_{18}$ metallic glasses ($x = 9,13,15,20$) at $H = 300$ Oe. The position of the maximum shifts to lower T/T_C with increasing x until simple FM behavior is observed at $x = 20$ (Fig. 2).

The SK magnetic phase diagram[11] is given in

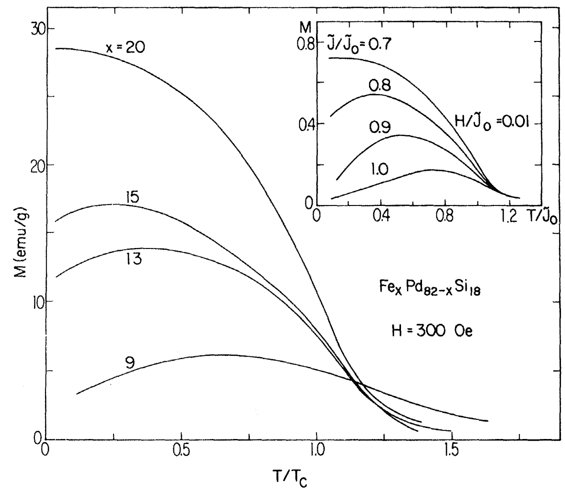

FIG. 2. Magnetization vs reduced temperature curves at 300 Oe of $Fe_9Pd_{73}Si_{18}$, $Fe_{13}Pd_{69}Si_{18}$, $Fe_{15}Pd_{67}Si_{18}$, and $Fe_{20}Pd_{62}Si_{18}$ metallic glasses. Values of T_C were determined by scaling analysis. Inset shows solutions of the SK equations at a fixed field, $H/\tilde{J}_0 = 0.01$, for several values of $0.7 \leq \tilde{J}/\tilde{J}_0 \leq 1$.

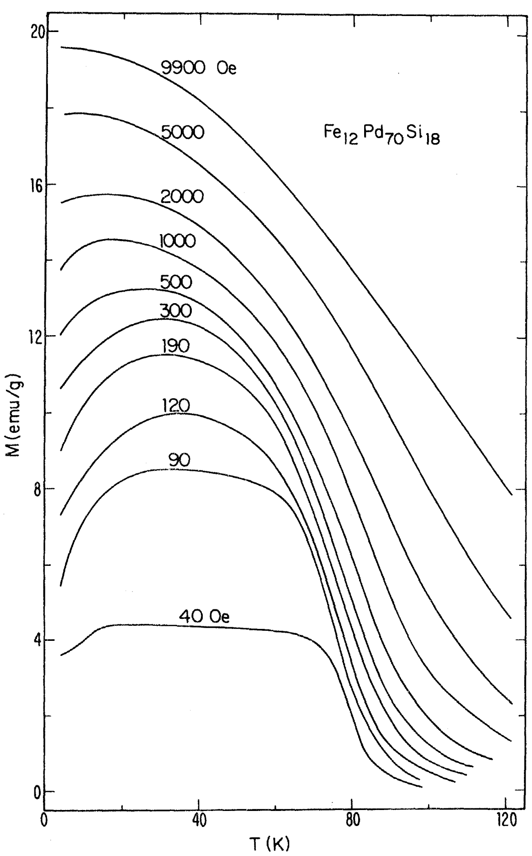

FIG. 1. Temperature dependence of the magnetization of a-$Fe_{12}Pd_{70}Si_{18}$ at several applied fields between 40 and 9900 Oe.

terms of the mean (value of the distribution of exchange energies $\tilde{J}_0$ and its standard deviation $\tilde{J}$. In the diagram the FM phase gives way to a SG phase at low temperature when $0.8 \leq \tilde{J}/\tilde{J}_0 \leq 1$. With $\tilde{J}/\tilde{J}_0$ in this range, the SK model predicts a maximum in the temperature dependence of the spontaneous magnetization, the field dependence of which closely resembles the results of Fig. 1.[9,10] Solutions of the SK equations for several $0.7 \leq \tilde{J}/\tilde{J}_0 \leq 1$ and a small fixed field, $H/\tilde{J}_0 = 0.01$, are plotted in the inset of Fig. 2. There is a remarkable resemblance between the experimental and calculated curves, with the increase in Fe content corresponding to a decrease in $\tilde{J}/\tilde{J}_0$ (Fig. 2).

A decrease in $\tilde{J}/\tilde{J}_0$ sufficient to drive the SG-FM transition in the SK model could be provided by an increase in Fe-Fe exhange energies alone, if it is similar to the observed increase in the moment per Fe, from 2.3 to 2.8μ_B between $x = 9$ and 20 at. % Fe in a-$Fe_xPd_{82-x}Si_{18}$.[5,6] Alternatively, as was proposed elsewhere,[6] if the SG-FM transition is due to an increasing concentration of magnetic clusters such that intercluster coupling overcomes local random anisotropy in the clusters, then the decreasing $\tilde{J}/\tilde{J}_0$ with increasing x, as Fig. 2 implies, could be associated with a decreasing width of the distribution of indirect intercluster interactions.[7] These appear to be Ruderman-Kittel-Kasuya-Yosida (RKKY)-like, showing strong "self-damping" with increasing x.[7]

In the following we apply scaling analysis to both FM-PM and FM-SG transitions in a-$Fe_xPd_{82-x}Si_{18}$. For a second-order FM-PM transition, the magnetic equation of state in the critical region is given by $M(H,T)=t^{\beta}m^*(y)$, where $t=T/T_C-1$ and $y=\mathrm{sgn}(t)(H/|t|^{\beta\delta})$. The scaling function m^* has two branches, m^*_- for $t<0$ and m^*_+ for $t>0$. The asymptotic behavior is: $\lim_{y\to 0} m^*_- =\text{const}$, $\lim_{y\to 0} m^*_+ =y$, and $\lim_{y\to\infty} m^*_\pm =y^{1/\delta}$. A completely analogous equation of state is applicable along the FM-SG phase boundary[8–10] with $\tilde{t}=1-T/T_{fg}$ and the critical exponents $\tilde{\beta}$ and $\tilde{\delta}$ replacing t, β, and δ, respectively; T_{fg} is the FM-SG transition temperature. For a proper choice of parameters, the $M(H,T)$ data collapse into two branches in a M/t^{β} vs y plot. This is illustrated in Figs. 3 and 4 for a-$Fe_{10}Pd_{72}Si_{18}$ in the vicinity of its FM-PM and FM-SG transitions, respectively. For clarity, only data recorded between $H=100$ and 1000 Oe are included in Figs. 3 and 4, although the scaling remains valid up to the highest applied field of 9900 Oe. The magnetization at fields below ~ 100 Oe, however, did not scale properly as a result, it appears, of demagnetization (Fig. 1). The critical exponents given in Figs. 3 and 4 are accurate to within 8% and the critical temperatures to within 1 K. The errors represent the range of values over which adequate scaling is achieved.

The scaling behavior of alloys with $x=9$ and 12 is identical to that of a-$Fe_{10}Pd_{72}Si_{18}$ with the same critical exponents (Figs. 3 and 4). All show "re-entrant" ferromagnetism with T_C increasing while T_{fg} decreases with increasing x. Within the framework of the SK model, the decreasing M at low

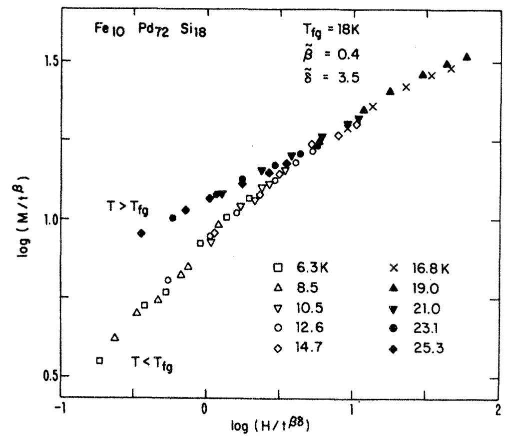

FIG. 4. Scaled magnetization vs scaled field of a-$Fe_{10}Pd_{72}Si_{18}$ in the vicinity of T_{fg}.

temperature for $x=13$ and 15 (Fig. 2) indicates that these alloys also undergo a FM-SG transition. However, T_{fg} values for the $x=13$ and 15 alloys could not be determined by scaling on the basis of the present experimental data available down to only 4.2 K. For a-$Fe_{13}Pd_{69}Si_{18}$ we estimate $T_{fg}\approx 5$ K, and for a-$Fe_{15}Pd_{67}Si_{18}$ it appears that $T_{fg}<4.2$ K. At the FM-PM transition, the scaling behavior for $x=13$ and 15 as well as for $x=20$ is very similar to that of a-$Fe_{10}Pd_{72}Si_{18}$ (Fig. 3) with the same critical exponents. The T_C values thus obtained increase linearly with $x \gtrsim 9$, reaching 170 K in ferromagnetic a-$Fe_{20}Pd_{62}Si_{18}$. These results, along with phase boundaries obtained elsewhere[1,4,6] up to $x=7$, are summarized in the magnetic phase diagram of a-$Fe_xPd_{82-x}Si_{18}$ shown in Fig. 5. It is in good qualitative agreement with the SK model.[8–10] In contrast with the previously published diagram,[6] T_{fg} decreases with increasing $9\le x\le 15$ (Fig. 5), indicating that it does not coincide with the "freezing" temperature as determined by the position of the maximum in the dc low-field M vs T curve,[6] or by ac susceptibility for a-$Fe_{13}Pd_{67}Si_{20}$ and a-$Fe_{20}Pd_{60}Si_{20}$.[3] Also, somewhat lower T_C values are obtained here (Fig. 5), in particular at high x. The dashed lines in Fig. 5 are extrapolations based on the present analysis and on earlier data indicating that a-$Fe_xPd_{82-x}Si_{18}$ are nonmagnetic below $x\approx 1$ and that the multicritical point is close to $x=7$.[1,2,4–6]

Within accuracy of the present scaling analysis, the FM-PM and FM-SG transitions in a-$Fe_xPd_{82-x}Si_{18}$ are each described by a single set of critical exponents regardless of composition, $9\le x \le 20$. Along the FM-PM line we find $\delta=5.0$

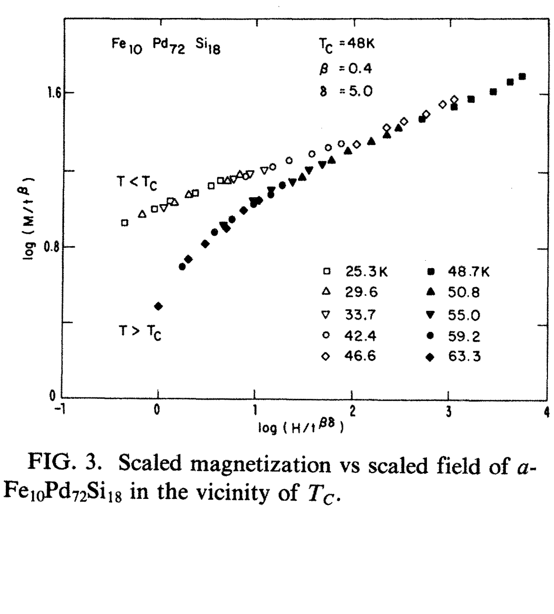

FIG. 3. Scaled magnetization vs scaled field of a-$Fe_{10}Pd_{72}Si_{18}$ in the vicinity of T_C.

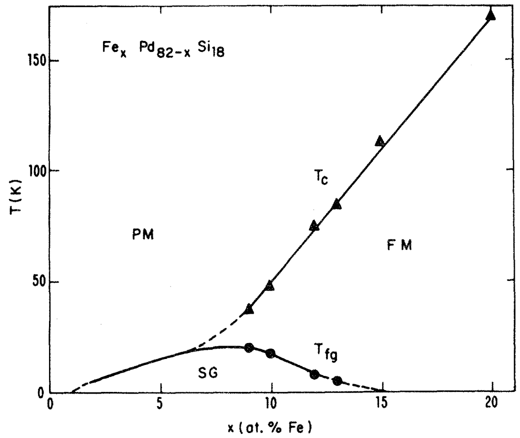

FIG. 5. Magnetic phase diagram of a-$Fe_xPd_{82-x}Si_{18}$. Scaling results are indicated by symbols; dashed lines indicate extrapolated behavior.

± 0.4 and $\beta=0.40\pm0.03$ (Fig. 3); $\tilde{\delta}=3.5\pm0.3$ and $\tilde{\beta}=0.40\pm0.03$ along the FM-SG line (Fig. 4). As in the a-$(M_{1-x}M'_x)_{75}P_{16}B_6Al_3$ alloys[8–10] we note that $\delta > \tilde{\delta}$. The former differs significantly from the Heisenberg value, in agreement with reports on other random alloys.[8–10,13] It is possible that this is due to the magnetic heterogeneity[5–7] of a-$Fe_xPd_{82-x}Si_{18}$ which would, conceivably, affect the sharpness of the transition. The curved Arrott plots of some a-$(M_{1-x}M'_x)_{75}P_{16}B_6Al_3$ also suggest the presence of magnetic heterogeneities.[8–10] The exponents associated with the FM-SG transition (Fig. 4), however, despite its unusual nature, are close to the mean-field values with $\tilde{\gamma}=\tilde{\beta}(\tilde{\delta}-1)=1$, in agreement with earlier studies.[8–10,14] The discrepancy between the critical behavior at T_C and T_{fg} still remains to be resolved.

The existence of a FM-SG critical line in the SK phase diagram has been questioned recently[15] and the presence of mixed phases was proposed.[16,17] The boundary between SG and mixed phase in those recent models is parallel to the temperature axis.[16,17] This, however, is merely a consequence of the mean-field approach taken[16,17] and, in principle, does not rule out temperature-driven transitions. The occurrence of such transitions is clearly indicated by the scaling analysis of a-$Fe_xPd_{82-x}Si_{18}$ here and in other cases.[8–10]

In summary, it was shown that the magnetization of the $Fe_xPd_{82-x}Si_{18}$ metallic glasses closely resembles the predictions of the SK model.[11] The occurrence of FM-SG transitions with temperature and composition was established. The FM-SG and FM-PM transitions are both continuous, each being described by a single set of critical exponents—$\tilde{\delta}=3.5\pm0.3$, $\tilde{\beta}=0.40\pm0.03$, and $\delta=5.0\pm0.4$, $\beta=0.40\pm0.03$, respectively—independent of composition. FM-SG and Curie transition temperatures were also determined by scaling and the magnetic phase diagram reported previously[6] was modified and is in good qualitative agreement with the SK diagram.

Thanks are due to Professor M. B. Salamon, Professor R. D. Parks, and Dr. H. Sompolinsky for valuable discussions. Dr. C.-H. Lin is gratefully acknowledged for the preparation and x-ray analysis of some of the samples. This work was supported in part by the National Science Foundation Grant Nos. DMR-79-25397 and DMR-77-23999.

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