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

G. Dublon
Division of Applied Sciences, Harvard University, Cambridge, Massachusetts 02138

Y. Yeshurun*
Department of Physics, University of Illinois, Urbana, Illinois 61801
and Department of Physics, Polytechnic Institute of New York, Brooklyn, New York 11201
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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 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 $2.8\mu_B$ 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]

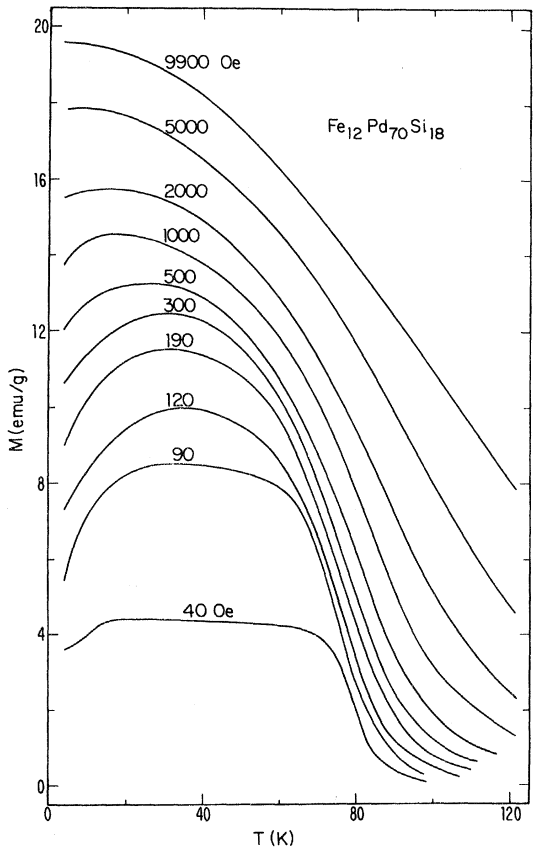

FIG. 1. Temperature dependence of the magnetization of a-$Fe_{12}Pd_{70}Si_{18}$ at several applied fields between 40 and 9900 Oe.

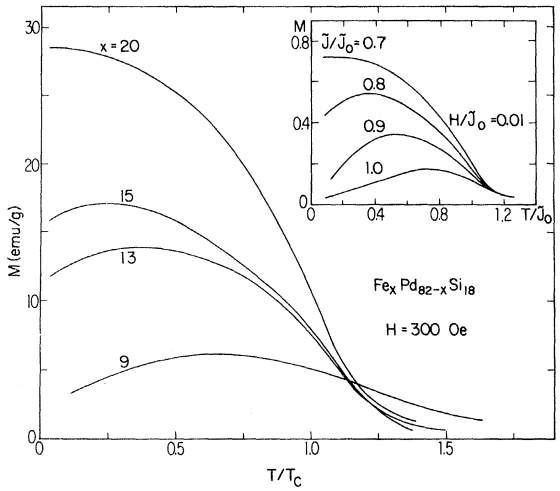

FIG. 2. Magnetization vs reduced temperature curves at 300 Oe of $Fe_9Pd_{73}Si_{18}$, $Fe_{13}Pd_{69}Si_{18}$, $Fe_{15}Pd_{67}Si_{18}$, and $Fe_{20}Pd_{62}Si_{18}$ metallic glasses. Values of T_C were determined by scaling analysis. Inset shows solutions of the SK equations at a fixed field, $H/\tilde{J}_0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

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

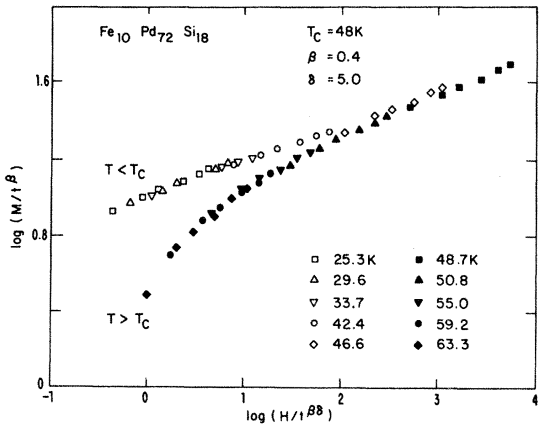

FIG. 3. Scaled magnetization vs scaled field of a-$Fe_{10}Pd_{72}Si_{18}$ in the vicinity of T_C.

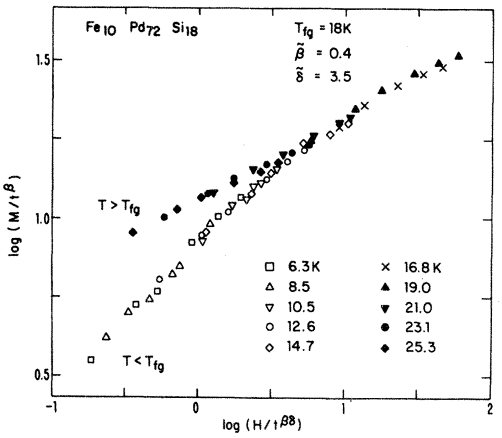

FIG. 4. Scaled magnetization vs scaled field of a-$Fe_{10}Pd_{72}Si_{18}$ in the vicinity of T_{fg}.

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 $\delta=5.0$

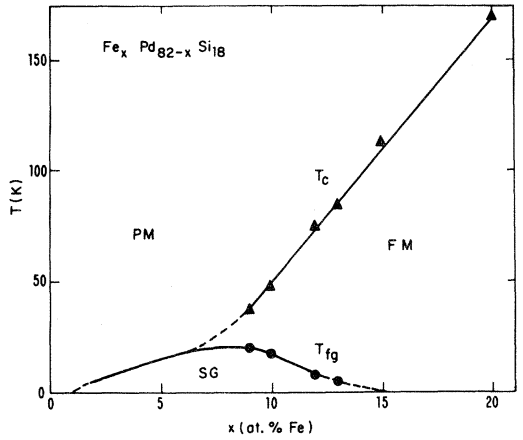

FIG. 5. Magnetic phase diagram of a-$Fe_xPd_{82-x}Si_{18}$. Scaling results are indicated by symbols; dashed lines indicate extrapolated behavior.

± 0.4 and $\beta = 0.40 \pm 0.03$ (Fig. 3); $\tilde{\delta} = 3.5 \pm 0.3$ and $\tilde{\beta} = 0.40 \pm 0.03$ 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—$\tilde{\delta} = 3.5 \pm 0.3$, $\tilde{\beta} = 0.40 \pm 0.03$, and $\delta = 5.0 \pm 0.4$, $\beta = 0.40 \pm 0.03$, 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.

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*Permanent address: Department of Physics, Bar-Ilan University, Ramat-Gan, Israel.

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