Spin-glass—ferromagnetic transitions and critical lines in Fe$_x$Pd$_{82-x}$Si$_{18}$ metallic glasses

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Magnetization measurements on a series of Fe$_x$Pd$_{82-x}$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$_x$Pd$_{82-x}$Si$_{18}$ is obtained. It differs significantly from that 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$^3$–$^7$ on the (amorphous) Fe$_x$Pd$_{82-x}$Si$_{18}$ system (2 \( \leq x \leq 25 \)) revealed a gradual transition with increasing Fe content from spin-glass (SG), for $x \leq 5$, to ferromagnet (FM) at $x \geq 20$. Intermediate compositions (5 \( \leq x \leq 20 \)) are spin-glass-like at low temperature, becoming ferromagnetic with increasing temperature.$^5$–$^7$ A magnetic phase diagram of Fe$_x$Pd$_{82-x}$Si$_{18}$, 2 \( \leq x \leq 25 \), was constructed$^5$ 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 Fe$_2$Pd$_{90}$Si$_{18}$ and Fe$_3$Pd$_{70}$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.$^5$

The purpose of the present report is to establish and examine the FM-SG critical line in the magnetic phase diagram of Fe$_x$Pd$_{82-x}$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 $a$-(Fe$_{1-x}$Pd$_x$)$_{73}$P$_{16}$B$_6$Al$_3$ 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$_x$Pd$_{82-x}$Si$_{18}$ alloys differ from the a-($M_1-xM'_x$)$_{73}$P$_{16}$B$_6$Al$_3$ 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$_x$Pd$_{82-x}$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$_x$Pd$_{82-x}$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 $\alpha$-Fe$_{12}$Pd$_{70}$Si$_{18}$ at several fields. Demagnetization effects which become significant only at fields below $\approx 100$ 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 $\sim 2000$ Oe for $\alpha$-Fe$_{12}$Pd$_{70}$Si$_{18}$ (Fig. 1). Similar behavior was observed in other $\alpha$-Fe$_x$Pd$_{82-x}$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$_x$Pd$_{82-x}$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 exchange 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 $\alpha$-Fe$_x$Pd$_{82-x}$Si$_{18}$.$^{5,6}$ Alternatively, as was proposed elsewhere,$^7$ 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 $x$.\$
In the following we apply scaling analysis to both FM-PM and FM-SG transitions in α-Fe$_x$Pd$_{82-x}$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 $y = \text{sgn}(t)(H/t)^\delta$. 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 $\lim_{y \to \infty} m^*_+ = y^{1/8}$. A completely analogous equation of state is applicable along the FM-SG phase boundary with $t = 1 - T/T_{fg}$ and the critical exponents $\beta$ and $\delta$ replacing $t$, $\beta$, and $\delta$, 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 $M/t^\beta$ vs $y$ plot. This is illustrated in Figs. 3 and 4 for α-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 α-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 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 α-Fe$_{11}$Pd$_{69}$Si$_{18}$ we estimate $T_{fg} \approx 5$ K, and for α-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 α-Fe$_{10}$Pd$_{72}$Si$_{18}$ (Fig. 3) with the same critical exponents. The $T_C$ values thus obtained increase linearly with $x \geq 9$, reaching 170 K in ferromagnetic α-Fe$_{30}$Pd$_{60}$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 α-Fe$_x$Pd$_{82-x}$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 $9 \leq x \leq 15$ (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,$^5$ or by ac susceptibility for α-Fe$_{15}$Pd$_{67}$Si$_{18}$ and α-Fe$_{20}$Pd$_{60}$Si$_{18}$.$^1$ 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 α-Fe$_x$Pd$_{82-x}$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 α-Fe$_x$Pd$_{82-x}$Si$_{18}$ are each described by a single set of critical exponents regardless of composition, $9 \leq x \leq 20$. Along the FM-PM line we find $\delta = 5.0$

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**FIG. 3.** Scaled magnetization vs scaled field of α-Fe$_{10}$Pd$_{72}$Si$_{18}$ in the vicinity of $T_C$.

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**FIG. 4.** Scaled magnetization vs scaled field of α-Fe$_{10}$Pd$_{72}$Si$_{18}$ in the vicinity of $T_{fg}$. 
The existence of a FM-SG critical line in the SK phase diagram has been questioned recently\(^1\) 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\(_2\)Pd\(_{32-x}\)Si\(_{18}\) here and in other cases.\(^8-10\)

In summary, it was shown that the magnetization of the Fe\(_2\)Pd\(_{32-x}\)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 — \(\delta = 3.5 \pm 0.3\), \(\beta = 0.40 \pm 0.03\), 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\(^8\) was modified and is in good qualitative agreement with the SK diagram.

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