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Study of Random Magnetic Alloys near Their Critical Concentrations under High Pressure

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(Received 3 September 1980)

The low-field ac magnetic susceptibility of the amorphous (FeMn)PBAl and (FeNi)-PBAl, and the crystalline (PdFe)Mn random magnetic alloys near their critical concentrations was measured under pressure up to 20 kbar between 1 and 300 K. The results support the proposed existence of reentrant ferromagnetism, and show the important role of magnetic clustering in the magnetic behavior of these alloys.

PACS numbers: 75.50.Kj, 75.20.Hr, 75.30.Kz

In a random magnetic metallic system, the delicate balance between various interactions dictates the magnetic structure at low temperature. The system[1] can be made of a Kondo system, a spin-glass, or a ferromagnet by continuously changing the magnetic-impurity concentration.

The study of the spin-glass state resulting from a random freezing has attracted great attention in the last few years. Recent scaling analysis[2] indicated that the spin-glass state was achieved through a phase transition. However, the nature of the different interactions involved and their delicate balance remain enigmatic. What is more unexpected is the predicted existence[3] of reentrant ferromagnetism in alloys near the critical concentration separating a ferromagnet from a spin-glass. In other words, ferromagnetism appears and then disappears with cooling when the alloy undergoes a paramagnetic to ferromagnetic and then a ferromagnetic to spin-glass transition. Subsequent low-field ac susceptibility measurements[4] did suggest the existence of reentrant ferromagnetism, evident from a double transition. The transition was exemplified by a sharp rise followed by a rapid drop in the ac susceptibility at lower temperature. More random magnetic systems were later discovered[5,6] or rediscovered to behave in a similar fashion. Nevertheless, the association of the rapid drop of ac susceptibility with a ferromagnetic to spin-glass transition has been questioned,[2,7] in spite of the similarities between this low-temperature state and a genuine spin-glass state on cooling. A systematic study on alloys near the critical concentration will therefore be most helpful in examining whether the double transition exists and, if it does, what the various interactions involved are and how they compete with each other giving rise to the unusual double and the spin-glass transitions. Since the magnetic properties of metallic alloys near the critical concentration are extremely sensitive to the sample and metallurgy conditions,[8] variation of pressure instead of small concentration was employed for the present investigation. This represents the first high-pressure study ever made on random magnetic systems exhibiting the above-mentioned anomalies. It should be noted that, in an alternative approach to minimize the metallurgical problems, the ionic compounds $Eu_{1-x}Sr_xS$ and the metallic alloys Fe_xCr_{1-x} near the critical concentration were recently examined.[9,10] Neutron-diffraction data[10] showed first the appearance and then the disappearance of long-ranged ferromagnetism, and also the decrease in spin-wave stiffness with decreasing temperature.

We have measured the temperature dependence of the low-field ac magnetic susceptibility χ of the amorphous (FeMn)PBAl and (FeNi)PBAl and the crystalline (PdFe)Mn alloys[11] near their critical concentrations x_c under hydrostatic pressure up to ~20 kbar between 1 and 300 K. The three alloy systems were specifically chosen because of the different composition effects on the magnetic properties of the different systems, as will be evident later in Fig. 2. The materials examined are the well-characterized amorphous[6,12,13] $(Fe_{1-x}Mn_x)_{75}P_{16}B_6Al_3$ (with $x = 0.35$, 0.40, 0.45, and 0.55) and $(Fe_{1-x}Ni_x)_{75}P_{16}B_6Al_3$ (with $x = 0.80$) and crystalline[11] $(Pd_{0.9965}Fe_{0.0035})_{1-x}Mn_x$ (with $x = 0.01$, 0.05, 0.06, and 0.065). The amorphous alloys were prepared by centrifugal spin quenching and the crystalline ones by arc melting followed by vacuum annealing at 1000 C for 2 h. The χ was measured in a field of ~1.5 Oe (rms) at 25 Hz with a standard inductance bridge. The hydrostatic pressure was generated by a self-clamp technique[14] with the 1:1 fluid mixture of n-pentane and isoamyl alcohol as pressure medium. The pressure at low temperature was determined by a lead manometer situated adjacent to the sample, and the temperature by a Ge thermometer below 20 K and by a Chromel-Alumel thermocouple above 20 K.

The temperature dependence of χ at ambient pressure similar to that previously observed was detected for all our samples. For instance, $\chi(T)$'s are shown in Fig. 1 for (FeMn)PBAl at two different pressures. For simplicity, T_C, T_f', and T_f are defined as the Curie and spin-glass temperatures in the same figure. Such a definition

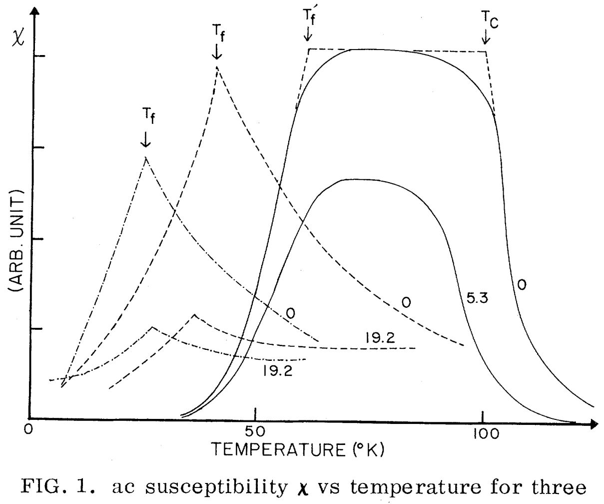

FIG. 1. ac susceptibility χ vs temperature for three alloys of $(Fe_{1-x}Mn_x)P_{16}B_6Al_3$ under two different pressures. T_C, T_f', and T_f are defined by the arrows. The number denotes the pressure in kilobars. Solid lines, $x = 0.35$; dashed lines, $x = 0.40$; dot-dashed lines, $x = 0.45$.

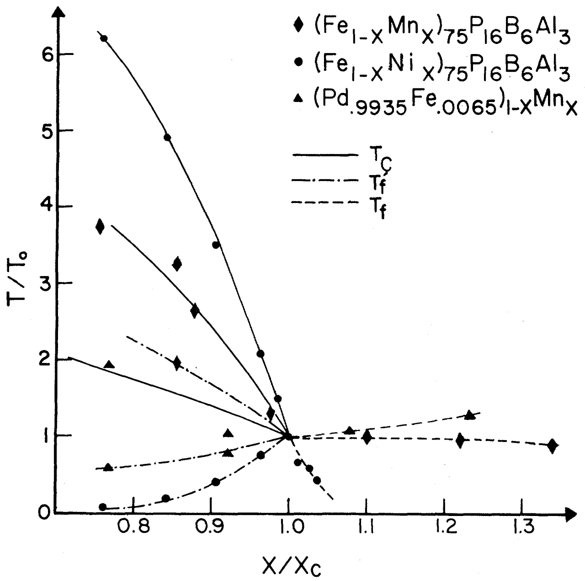

FIG. 2. The magnetic phase diagrams of the three random magnetic systems studied. (See also Moorjani *et al.*, Ref. 15.)

of transition temperatures is by no means unique. The reduced magnetic phase diagrams for the three systems were reproduced and schematically represented in Fig. 2. The critical concentrations x_c, above which only a single spin-glass state occurs, are ~0.40, ~0.82, and ~0.06 for (FeMn)PBAl, (FeNi)PBAl, and (PdFe)Mn, respectively. T_C and T_f (T_f will be T_f' when T_C exists) are listed in Table I together with their pressure coefficients for all samples investigated. As shown in Fig. 2, although T_C always decreases with an x increase, the x dependences of T_f' and T_f are quite different for the three alloy systems. We have thus found that (1) for $x < x_c$, pressure always suppresses T_C while it enhances T_f'; (2) for $x \sim x_c$, where an asymmetrical peak in χ appears, pressure moves the high-temperature shoulder downward in temperature whereas it moves the low-temperature shoulder upward, with a net downward shift in the peak temperature or T_f; (3) for $x > x_c$, pressure enhances T_f for all samples studied except for (FeMn)PBAl with $x - 0.55$ where $\partial T_f/\partial P$ changes from positive to negative with pressure at ~3 kbar; (4) pressure always suppresses $\chi(T_f)$, the χ anomaly signaling the spin-glass transition, at different rates for the different alloy systems; and (5) pressure always suppresses the χ_0, the maximum χ plateau below T_C and above T_f'.

Various models of spin-glasses have been proposed and can be summarized into two basic ones, namely, the phase-transition model,[3,16] and the cluster-blocking model.[17] The former emphasizes the spin-glass transition while the latter the spin-glass behavior. The existence of reentrant ferromagnetism was successfully predicted by Sherrington and Kirkpatrick (SK)[3] on the basis of the phase-transition picture. They proposed a solvable model with N Ising spins interacting through infinite-ranged exchange interactions which are independently distributed with a Gaussian probability density. They found that the appearance of different magnetic phases is dictated by the ratio $\tilde{J}_0/\tilde{J}$, where $\tilde{J}_0 = J_0/N$ is the scaled most probable exchange interaction and $\tilde{J} = J/N^{1/2}$ the scaled width of the Gaussian distri-

TABLE I. Pressure and volume effects on T_C and T_f. T_f represents T_f' if there exists a T_C for that particular compound. The compressibilities used were 9.66×10^{-7}, 6.85×10^{-7}, and 5.42×10^{-7} bar^{-1}, respectively, for (FeMn)PBAl, (FeNi)PBAl, and (PdFe)Mn. An ellipsis indicates that the transition did not occur.

	T_C (K)	T_f (K)	$\partial T_C/\partial P$ (10^{-4} kbar^{-1})	$\partial T_f/\partial P$ (10^{-4} kbar^{-1})	$\partial \ln T_C/\partial \ln V$	$\partial \ln T_f/\partial \ln V$
			$(Fe_{1-x}Mn_x)_{75}P_{16}B_6Al_3$			
$x = 0.35$	98 ± 1	60.5 ± 0.5	-21.3 ± 0.5	$+2.7 \pm 0.2$	$+22.5 \pm 0.5$	-4.6 ± 0.4
$x_c = 0.40$	···	40.4 ± 0.5	···	-1.9 ± 0.2	···	$+4.9 \pm 0.2$
$x = 0.45$	···	24.8 ± 0.2	···	$+1.3 \pm 0.1$	···	-5.4 ± 0.1
			$(Fe_{1-x}Ni_x)P_{16}B_6Al_3$			
$x = 0.8$	89 ± 2	18.4 ± 0.5	-0.18 ± 0.05	$+0.20 \pm 0.05$	$+2.9 \pm 0.5$	-15.9 ± 0.4
			$(Pd_{0.9965}Fe_{0.0035})_{1-x}Mn_x$			
$x = 0.01$	14 ± 0.1	···	-0.31 ± 0.01	···	$+4.18 \pm 0.04$	···
$x = 0.05$	9 ± 0.1	3 ± 0.1	-0.95 ± 0.01	$+0.32 \pm 0.02$	$+19.46 \pm 0.05$	-19.67 ± 0.05
$x_c = 0.06$	···	4.9 ± 0.1	···	-0.45 ± 0.02	···	$+16.9 \pm 0.05$
$x = 0.065$	···	5.2 ± 0.1	···	$+0.27 \pm 0.02$	···	-9.75 ± 0.05

bution. The calculations showed that $kT_c = \tilde{J}_0$ for $\tilde{J}_0/\tilde{J} > 1$, $kT_f = \tilde{J}$ for $\tilde{J}_0/\tilde{J} < 1$, and kT_f' is a rapidly decreasing function of $\tilde{J}_0$ for $1 < \tilde{J}_0/\tilde{J} < 1.25$. Therefore $\tilde{J}_0$ and $\tilde{J}$ can be identified with the exchange interactions responsible, respectively, for the ferromagnetic ordering and spin-glass transition and can be varied by altering the composition[6, 13, 18, 19] and pressure P. The critical concentration x_c previously defined then occurs at $\tilde{J}_0 = \tilde{J}$. The effects of x and P on $\tilde{J}_0$ and $\tilde{J}$ are thus deduced from the x and P effects on T_C and T_f. SK[3] also found that $\chi(T_f)$ for $x > x_c$ decreases with decreasing $\tilde{J}_0/\tilde{J}$. A qualitative comparison of the predictions of the SK model with the experimental observations is given in Table II. The general agreement is quite remarkable, in view of the simplicity of the model. However, some disagreements do exist. It should also be noted that the "predicted" sign of $\partial\chi(T_f)/\partial P$ was obtained only self-consistently, based on the sign of $\partial T_f/\partial P$ observed. The comparison in Table II therefore suggests that $\tilde{J}$ may not be just a simple antiferromagnetic exchange interaction, e.g., arising from the nearest-neighbor Mn-Mn interaction.

In the blocking picture,[17] the formation and growth of finite magnetic clusters play an important role in the behavior of spin-glass. The observed χ plateau immediately below T_C, χ_0, resulting from the real χ divergence at T_C is equal to the reciprocal demagnetization factor $1/D$. For a uniform sample, D depends only on the sample geometry and should change only negligibly by hydrostatic pressure. However, χ_0 was found to drop rapidly with pressure, as displayed in Fig. 1. This suggests that the sample may not be homogeneous magnetically and the effective D has changed through the pressure-induced reduction in the size and/or density of the magnetic clusters, consistent with the cluster-blocking model. In fact, we found that the pressure-suppressed χ_0 is common to all giant magnetic moment systems studied by us near x_c. With use of such a view, the pressure-suppressed $\chi(T_f)$ can also be understood, provided that a freezing mechanism increasing in strength with pressure is assumed. Unfortunately, the model cannot account for the increase of $\chi(T_f)$ with T_f by changing x, not to mention the occurrence of the various magnetic phases.

The above discussions show the incompleteness of either basic model. It is proposed that one including the main features of the two models will provide a more complete description of the aforementioned observations. In other words, in a random magnetic alloy, there exist not only the positive (ferromagnetic) and negative (antiferromagnetic) exchanges, but also an additional freezing mechanism. It is the delicate balance between the former two interactions responsible for the formation or destruction of the ferromagnetic long-range order, whereas the latter gives rise to the random freezing of the spins. Except for contributing to a positive pressure effect on T_f and T_f', the exact nature of the freezing mechanism remains unknown at present. Such a proposition will be consistent with not only the paramagnetic–ferromagnetic–paramagnetic–spin-glass transition[20] in *Fe*Al, but also the wide spread of $\partial \ln T_f/\partial \ln V$ both in magnitude and sign given in Table I. For example, T_f of (FeMn)PBAl with $x = 0.55$ peaks at 3 kbar. Recently, with the assumption of a constant pressure coefficient of the exchange interaction independent of x, it was suggested[21] that only the Ruderman-Kittel-Kasuya-Yosida interaction was important in a spin-glass transition in *Ag*Mn. The assumption made is clearly contrary to the present and previous[22] observations.

The ever decreasing $\chi(T_f)$ with P suggests the possible existence of a "critical" point in the T-P diagram separating a paramagnet from a spin-glass. For instance, $\chi(T_f)$ for (FeMn)PBAl with $x = 0.45$ becomes zero at 28 K and 25 kbar by lin-

TABLE II. Comparison of predictions by SK model with observations.

		$\partial T_C/\partial x$	$\partial T_f'/\partial x$	$\partial T_f/\partial x$	$\partial\chi(T_f)/\partial x$	$\partial T_f/\partial P$	$\partial\chi(T_f)/\partial P$
(FeMn)PBAl	SK	−	+	···	+	···	−
	Expt.	−	−	−	−	+	−
(FeNi)PBAl	SK	−	+	···	+	···	−
	Expt.	−	+	−	−	+	−
(PdFe)Mn	SK	−	+	···	−	···	−
	Expt.	−	+	+	−	+	−

ear extrapolation. Experiments are also under way to determine if the sequential order of the appearance of a paramagnet, ferromagnet, and spin-glass can be switched and randomized in the T field by pressure. This will provide us with further useful information concerning the various interactions proposed.

We would like to thank J. H. Lin for technical assistance. The work was supported in part by National Science Foundation Grant No. DMR-79-08486 and the Energy Laboratory, University of Houston.

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