6-2001

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Published in *Journal of Applied Physics*, Vol. 89 No. 11 (2001) at doi: 10.1063/1.1362651

Spin glass phase in spin-density-wave Cr–Co alloys

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A spin glass (SG) phase was observed in the spin-density wave (SDW) alloys $Cr_{1-x}Co_x$ at high Co concentrations ($x \geqslant 13\%$). After zero-field cooling, the temperature dependence of the magnetization $M(T)$ of $Cr_{0.87}Co_{0.13}$ and $Cr_{0.85}Co_{0.15}$, measured in an applied field $H=100$ Oe with a SQUID magnetometer, exhibits a low T maximum, characteristic of a SG. Cooling in the measuring field, however, gives quite a different behavior. The field dependence of the magnetization $M(H)$ is nonlinear, with a pronounced hysteresis. A strong time relaxation $M(t)$ is also an indication of a SG phase. SG in $Cr_{1-x}Co_x$ is formed at high impurity concentrations, possibly due to clustering of the magnetic impurities. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1362651]

I. INTRODUCTION

Spin-density wave (SDW) Cr alloys with $3d$ impurities Fe, Mn, and Co exhibit Curie–Weiss (CW) behavior in the paramagnetic phase, due to the local moments of the impurity atoms.[1] However, among these alloys only $Cr_{1-x}Fe_x$ shows CW paramagnetism below the Néel temperature T_N for all concentrations of Fe. The alloys $Cr_{1-x}Co_x$ and $Cr_{1-x}Mn_x$ with impurity concentrations below $x^* \sim 10\%$ do not follow a CW law below T_N, which is considered to be a result of a strong coupling between the local moment on an impurity and the SDW.[1]

A spin glass (SG) phase was observed in the alloys $Cr_{1-x}Fe_x$ at high Fe concentrations ($x > 10\%$).[1] At such concentrations, magnetic clusters become rather large and the magnetic behavior should be described in terms of a cluster glass, which can be considered to be a highly concentrated regime of a SG. This is probably the reason why in all articles devoted to the study of the magnetic behavior of highly concentrated $Cr_{1-x}Fe_x$ alloys [1] the term spin glass is used. Contrary to $Cr_{1-x}Fe_x$, SG in $Cr_{1-x}Mn_x$ was found at very small Mn concentrations ($x < 0.1\%$).[2,3] At low temperatures the behavior of $Cr_{1-x}Mn_x$ is that of a typical spin glass. However, a strong coupling between Mn moments and the SDW, prevents strong impurity–impurity interactions, a basic necessity for the spin glasses.[4] Besides, in $Cr_{1-x}Mn_x$ the freezing temperature is independent of Mn concentration, which also indicates that impurity–impurity Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction is blocked. Therefore in $Cr_{1-x}Mn_x$ the origin of SG is essentially different from that in canonical metallic spin glasses.

Although previous magnetic measurements have not revealed SG properties in the alloys $Cr_{1-x}Co_x$,[1] we report here the formation of a SG phase in this SDW system.

II. EXPERIMENT

Samples of the $Cr_{1-x}Co_x$ ($1\% \leqslant x \leqslant 15\%$) polycrystalline alloy system were prepared and characterized by methods described previously.[5] The temperature dependence of the magnetization $M(T)$ has been measured with a SQUID magnetometer in an applied magnetic field $H=100$ Oe, in the zero field cooled (ZFC) and field cooled (FC) states, over the temperature range $2 \leqslant T \leqslant 400$ K. In some cases $M(T)$ was measured in two FC regimes: on cooling from 400 K down to 2 K in the measuring field (FCC state) and on warming through the same temperature range in the same measuring field after a FCC procedure (FCW state). All alloys exhibit very little temperature hysteresis between the FCC and FCW states. Magnetization $M(T)$ of all alloys exhibit a CW behavior in the paramagnetic phase. However, below T_N the alloys with $x \leqslant 6.5\%$ in the ZFC state show a monotonic decrease of $M(T)$ as the temperature decreases. Alloys with high Co concentrations ($x \geqslant 13\%$) exhibit a CW law in both the paramagnetic and the SDW phases (inset of Fig. 1). However, below and above T_N, the fitting parameters to the CW law are essentially different. $M(T)$ was fitted by a CW expression of the form

$$M(T) = M_i^0 + \frac{C_i}{(T - T_i^0)}, \tag{1}$$

where i stands for the SDW(S) or the paramagnetic (P) phases, so that the fitting parameters are: M_S^0 and M_P^0, C_S and C_P, T_S^0, and T_P^0. The corresponding values of the effective moments of the Co atom in each phase, μ_S and μ_P, could be obtained from the expression

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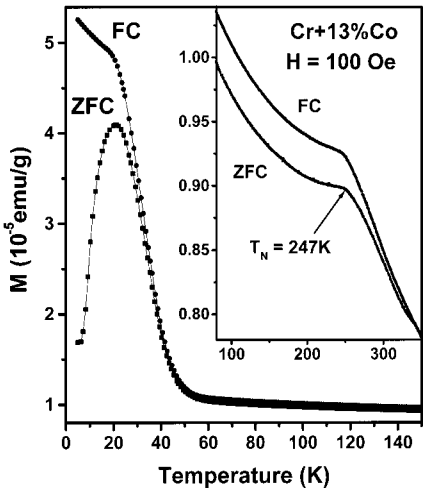

FIG. 1. Temperature dependence of the magnetization of Cr+13%Co, measured in $H=100$ Oe in the ZFC and FC states. The inset is an amplification of the same $M(T)$ curves in the high-temperature interval.

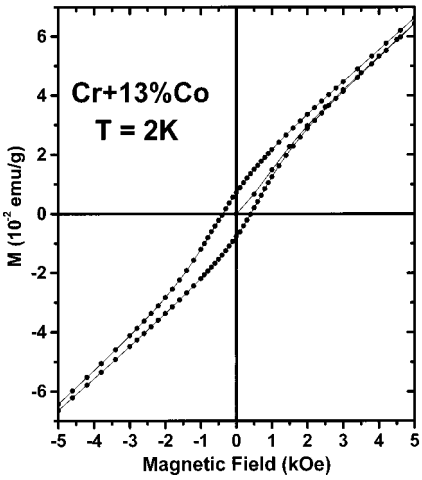

FIG. 2. The central part of the magnetic hysteresis loop of Cr+13%Co, measured over the magnetic field range, $-5\ \mathrm{T} \leqslant H \leqslant 5\ T$, at $T=2$ K in the ZFC state.

$$C_i = \frac{N\mu_i^2\mu_B^2}{3k_B}, \tag{2}$$

where N is the density of Co atoms. In the alloys $Cr_{1-x}Co_x$ ($x=13\%$ and 15%) C_S is about ten times smaller than C_P, indicating that the effective moment of Co below T_N is less than a third of its value in the disordered phase. Since the magnetic response is clearly different from one phase to the other, one can easily distinguish the Néel transition (inset of Fig. 1). Below a certain critical temperature T_i, all $Cr_{1-x}Co_x$ alloys with $x \leqslant 6.5\%$, exhibit a thermal hysteresis between the ZFC and FC curves.[6] For these alloys, T_i is close to T_N. At low temperatures no anomalies are observed. The alloys $Cr_{1-x}Co_x$ with $x=13\%$ and 15% exhibit a thermal hysteresis in $M(T)$ in ZFC and FC states in the vicinity of T_N as well (inset of Fig. 1) but, contrary to the alloys with $x \leqslant 6.5\%$, they exhibit a very strong magnetic irreversibility at low temperatures (Fig. 1). $M(T)$ in the ZFC state shows the characteristic low T maximum of a SG, while field cooling gives quite a different behavior. The field dependence of the magnetization $M(H)$ is nonlinear, with a pronounced hysteresis (Fig. 2). After field cooling the alloys $Cr_{1-x}Co_x$ ($x=13\%$ and 15%) in a strong magnetic field ($H=5$ T), the magnetization measured in $H=100$ Oe decays with time but, after ZFC, it increases with time (Fig. 3). These strong relaxation effects are also typical of spin glasses.

III. DISCUSSION

The lack of SG phase in the alloys $Cr_{1-x}Co_x$ with small Co concentrations could be explained by pinning of the Co moments in the SDW matrix, which blocks the impurity–impurity RKKY interaction. Formation of SG in the alloys with high Co concentration coincides with the advent of the CW paramagnetism below T_N and, therefore, may be attributed to the release of the Co moments. However, a more careful analysis of experimental data shows that the situation is more complicated.

Let us analyze the magnetic behavior of all three SDW Cr alloy systems with $3d$ impurities, Fe, Co and Mn, exhibiting SG properties. Such analysis gives a very contradictory picture. The behavior of $Cr_{1-x}Co_x$ and $Cr_{1-x}Mn_x$ is similar. In both cases, alloys with $x<x^*$ have the local moments on $3d$ impurities frozen in the SDW matrix and a CW paramagnetism appears below T_N approximately at the same impurity concentration: $x^* \sim 10\%$. On the other hand, the behavior of $Cr_{1-x}Co_x$ and $Cr_{1-x}Fe_x$ in the SDW phase is different. Contrary to $Cr_{1-x}Co_x$, $Cr_{1-x}Fe_x$ exhibits a CW law at all Fe concentrations. The concentration dependence of the Néel temperature $T_N(x)$ is also very different. In $Cr_{1-x}Fe_x$, $T_N(x)$ decreases monotonically with x and, for $x>10\%$, corresponding to the SG formation, T_N is below 100 K.[1] In the case of $Cr_{1-x}Co_x$, $T_N(x)$ is a nonmonotonic function of x[1] and, for Cr+13%Co, is as high as 247 K (Fig. 1). As in Cr alloys the magnitude of the moment on the Cr site is proportional to T_N,[1] one may expect that at concentrations corre-

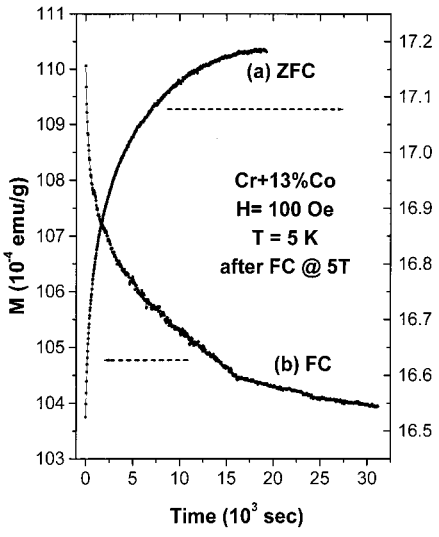

FIG. 3. Time dependence of magnetization of Cr+13%Co, measured in $H=100$ Oe: (a) after ZFC, (b) after FC in 5 T from 400 K.

sponding to SG formation the magnetic moment on Cr in $Cr_{1-x}Co_x$ could be much larger than in $Cr_{1-x}Fe_x$. Though both alloys, $Cr_{1-x}Co_x$ and $Cr_{1-x}Fe_x$, exhibit CW paramagnetism in the SDW phase at impurity concentrations corresponding to the SG formation ($x>10\%$), the magnetization in $Cr_{1-x}Fe_x$ at $x>10$ is about 2 orders of magnitude larger than that in $Cr_{87}Co_{13}$. This fact and our estimations of the effective moment on Co below and above $T_N(\mu_S<1/3\mu_P)$ show that in $Cr_{1-x}Co_x(x\geqslant 13\%)$ moments on Co are still strongly pinned by the SDW. Nevertheless, in spite of all these differences, the SG in $Cr_{1-x}Co_x$ is similar to that in $Cr_{1-x}Fe_x$, but not to $Cr_{1-x}Mn_x$. In $Cr_{1-x}Mn_x$, the SG phase is present even for very low Mn concentrations (below 0.1% Mn) and becomes negligible above x^*. On the other hand, in $Cr_{1-x}Co_x$ and $Cr_{1-x}Fe_x$ the SG phase is formed approximately at the same x (exceeding x^* for $Cr_{1-x}Co_x$; for $Cr_{1-x}Fe_x$ x^* does not exist).

Similarly to $Cr_{1-x}Fe_x$ (Ref. 1) we have considered the magnetic properties of $Cr_{1-x}Co_x$ ($x=13$, 15) employing the generic term spin glass for all concentrations of magnetic impurities. However, in both alloy systems SG properties are observed at high magnetic concentrations and in both cases the magnetic behavior should be considered in terms of a cluster glass. In fact in alloys with a concentration of magnetic impurities around 10%–15%, the magnetic behavior is dominated by the presence of magnetic clusters with effective moments on the order of $20-20\,000\mu_B$.[4] At sufficiently low temperatures, these clusters freeze with a random orientation in a manner analogous to the spin glass freezing. In the SDW alloys $Cr_{1-x}Co_x$ and $Cr_{1-x}Fe_x$, SG properties are not observed until a cluster glass is formed. In the alloys $Cr_{1-x}Co_x$ ($x=13\%$ and 15%), the Néel temperature is high and a magnetic moment on Cr is expected to be rather large, therefore the coupling between moments on Cr and Co seems to remain strong. This assumption is confirmed by estimations of the effective moment on Co that decreases more than three times as the paramagnetic–SDW transition takes place. Only rather large Co clusters seem not to be pinned by the SDW. Magnetic moments of such clusters give rise to the CW behavior and interact with each other, resulting in the formation of a cluster glass.

In spite of all differences in magnetic behavior of $Cr_{1-x}Co_x$ and $Cr_{1-x}Fe_x$, the common feature of both alloys is that Co and Fe are strong ferromagnetics. Therefore, one may expect that in both alloys with $x>10\%$ magnetic clusters carry rather large moments and that this is important for cluster glass formation. In $Cr_{1-x}Mn_x$ alloys with high Mn concentrations, the magnetic moments of individual atoms in Mn clusters seem to be coupled antiferromagnetically, resulting in small moments, thus preventing formation of a cluster glass.

ACKNOWLEDGMENTS

This work was supported in part by FAPESP, Brazil (V.Yu.G. and W.A.O.), CNPq, Brazil (W.A.O.), and NATO under Linkage Grant No. 973279 (V.Yu.G., N.A., and E.F.).

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