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Spin-glass and double-transition behavior in Gd-La glasses

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We report studies of high-field magnetization, field-cooled magnetization, thermoremanent magnetization, and isothermal remanent magnetization as a function of temperature in the metallic glass system $Gd_xLa_{72-x}G_{28}$ where $G_{28} = Ga_{18}B_{10}$ for the compositions $x = 72, 68, 64$. The first two compositions show double-transition behavior and the last one shows only a paramagnetic to spin-glass transition. The approach to saturation at intermediate fields (up to 40 kOe) is of the form $1/H^{1/2}$ as predicted by a mean field model which incorporates a small random anisotropy. The field-cooled magnetization in these samples is independent of time and is reversible, suggesting it is the equilibrium magnetization. This field-cooled magnetization in the double transition samples (and spin-glass sample) goes smoothly to zero with applied field, indicating the absence of an equilibrium macroscopic moment in zero applied field. The thermoremanence shows a maximum which becomes progressively more pronounced as x decreases.

A range of systems including both crystalline[1] and amorphous alloys[2,3] are now known to show spin-glass and double-transition behavior, that is paramagnetic to ferromagnetic (FM) to spin-glass (SG) transitions. As pointed out by other authors, the wide range of systems showing SG behavior is not surprising since the only ingredients required to produce such behavior in computer simulations are frustration in the microscopic interactions, including a small anisotropic interaction in the case of Heisenberg spins.[4,5] It is not known what microscopic interactions are required to produce the double-transition behavior, though the presence of SG behavior suggests an anisotropic interaction is involved. We have recently reported evidence for a phase-transition description of the FM-SG transition in *a*-$Gd_xLa_{72-x}G_{28}$ where $G_{28} = Ga_{18}B_{10}$.[6] This system shows double-transition behavior[3] for $x \gtrsim 67$. In this work, we report on the SG behavior in this system for the $x = 72, 68, 64$ samples which span the experimental multicritical point. These alloys are prepared by splat cooling and are x rayed to ensure amorphousness. The samples are in the form of sandwiches of strips ~ 1 cm long (in the case of the $x = 72$ sample a 1.5-cm-long sandwich was used) aligned along the applied field direction to minimize demagnetization effects.

The field-cooled magnetization in an applied field of 20 Oe is shown in Fig. 1 for the $x = 72$, 68, and 64 samples. This magnetization is reversible and time independent and shows a decrease at low temperatures characteristic of SG systems. In the $x = 72$ sample, the magnetization shows a small decrease at 4.2 K and decreases rather rapidly below 4.2 K. ac susceptibility results[3] in an rms field of ~ 1 Oe for this sample indicate a FM-SG transition at ~ 44 K and the shift of this transition to below 4.2 K in applied fields of 20 Oe indicates the sensitivity of the SG transition in the $x = 72$ sample to applied fields. Similar behavior has been described by Manheimer and co-workers[7] in transition metal glasses at low fields. In the $x = 68$ and 64 samples, the transition temperatures are shifted by at most a few degrees in a 20-Oe field.

Figure 2 shows the high-field magnetization up to 40 kOe for the $x = 72$ sample. Chudnovsky and Serota[8] have discussed the effects of a small random magnetic anisotropy within a mean-field theory for Heisenberg spins and find what they call a "correlated SG" state. This state is characterized by a ferromagnetic correlation of spins over short distances and a smooth rotation of the magnetization direction at larger distances so that no macroscopic moment is present. In this model, the approach of magnetization to saturation at intermediate fields is of the form $1/H^{1/2}$ and at high fields it is $1/H^2$ as found by earlier work on random magnetic[9] anisotropy models. Also, the exchange fluctuation model of Larkin and Khmel'nitskii[10] predicts a $1/H$ approach to saturation. We have tried $1/H^n$ fits where $n = 2, 1, \frac{1}{2}$ to the high and intermediate field magnetization in our $x = 72$, 68, 64 samples and in all cases the $n = \frac{1}{2}$ fit is best below ~ 40 kOe. The fit for the $x = 72$ sample is shown in Fig. 2. In this sample, the $n = \frac{1}{2}$ fit is marginally better than the $n = 1$ fit above 40 kOe and below this field the $n = \frac{1}{2}$ fit remains good while the $n = 1$ fit shows some deviations. In summary then, the approach to saturation at fields below 40 kOe is of the form $1/H^{1/2}$. Computer simulations of Heisenberg spin systems indicate that an anisotropy is required to produce nonequilibrium SG behavior.[5,11] The source of anisotropy in our glasses is not known yet but two possibilities are a small random magnetic anisotropy as discussed in Ref. 3 or a Dzaloshinsky–Moriya interaction as discussed by Fert and Levy.[12] If the former is the dominant source of anisotropy, then this $1/H^{1/2}$ approach to saturation is evidence of a correlated SG picture of the low-temperature phase in finite fields. The possibility of a correlated SG state has been discussed in some related Gd alloys by Sellmyer and Nafis.[13]

The isothermal remanent magnetization (IRM) and thermoremanent magnetization (TRM) are shown in Figs. 3–5 along with their associated finite field magnetizations.

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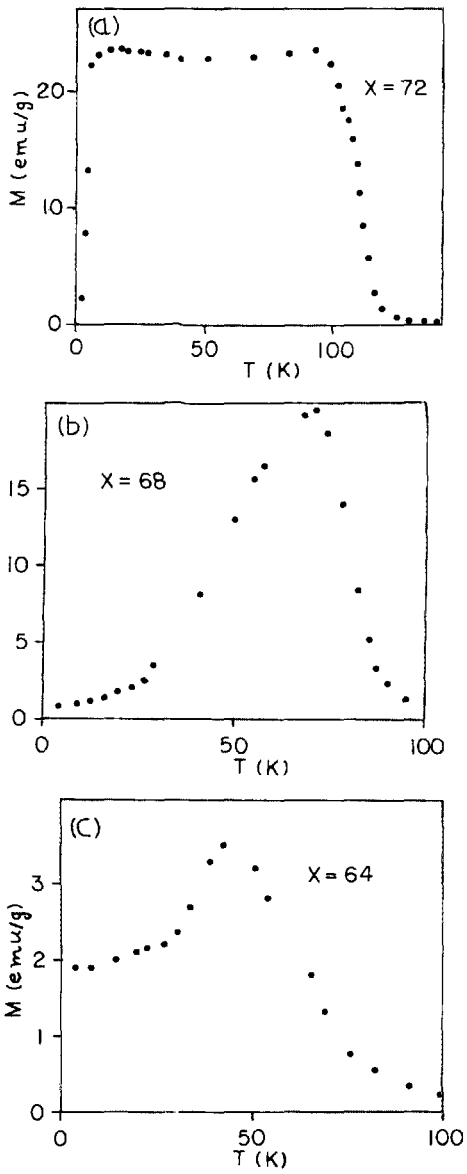

FIG. 1. Field-cooled magnetization for the (a) $x = 72$ sample, (b) $x = 68$ sample, and (c) $x = 64$ sample in an applied field of 20 Oe.

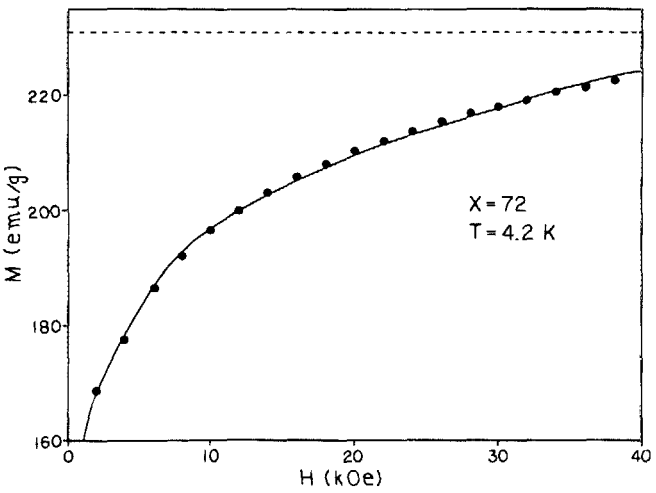

FIG. 2. Fit of a $1/H^{1/2}$ approach to saturation of magnetization between 0.1 and 4 kOe for the $x = 72$ sample at 4.2 K. The dashed line indicates the magnetization at 80 kOe.

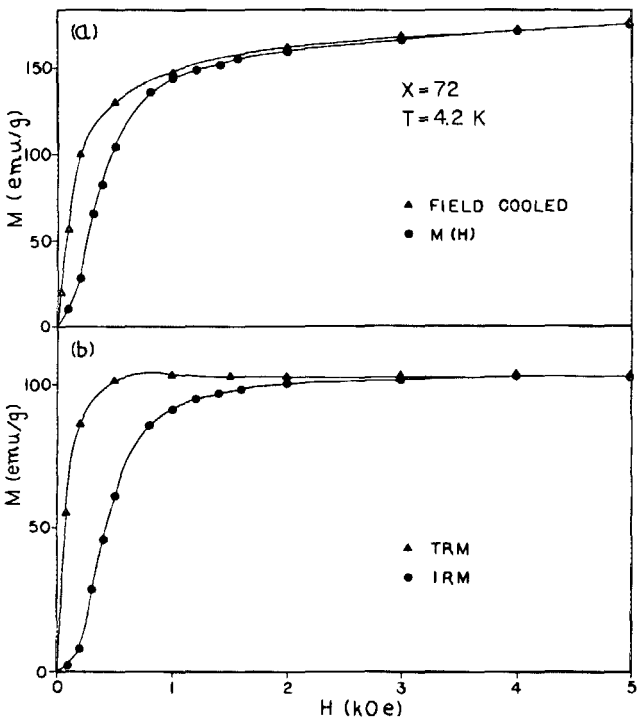

FIG. 3. (a) Field-cooled magnetization and initial magnetization $M(H)$ and (b) IRM and TRM at 4.2 K in the $x = 72$ sample after 15 min.

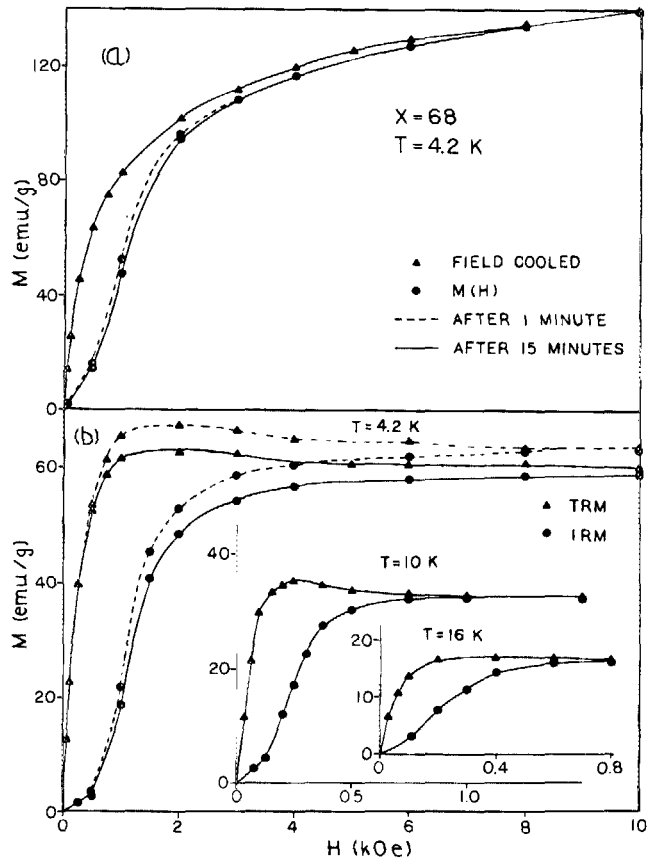

FIG. 4. (a) Field-cooled magnetization and initial magnetization $M(H)$ and (b) IRM and TRM as a function of time at 4.2 K in the $x = 68$ sample. The insert in (b) shows the IRM and TRM as a function of temperature.

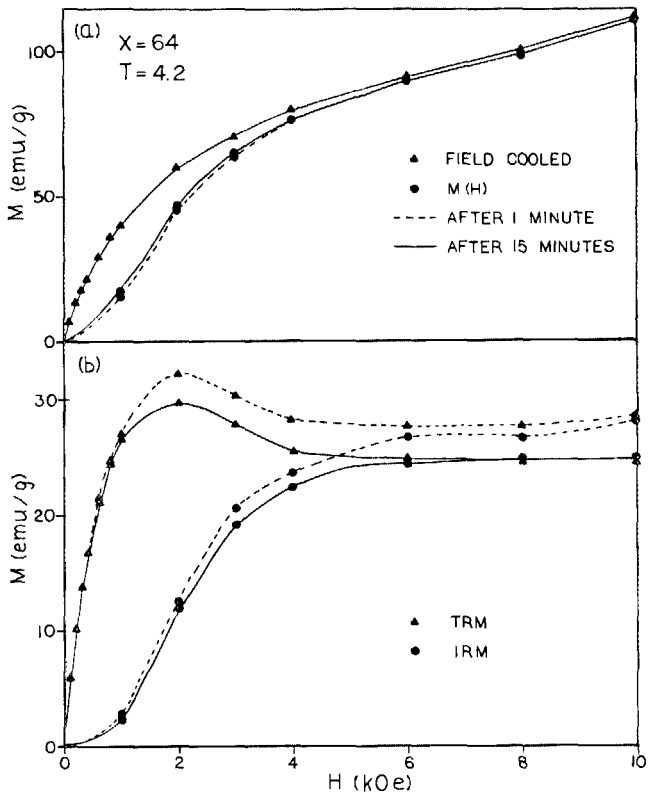

FIG. 5. (a) Field-cooled magnetization and initial magnetization $M(H)$ and (b) IRM and TRM as a function of time at 4.2 K in the $x = 64$ sample.

Only the field-cooled magnetization is time independent and the time dependence of $M(H)$, the magnetization taken after cooling in zero field to 4.2 K and increasing H, is shown after 1 and 15 min in Figs. 4(a) and 5(a). The TRM shows a maximum at intermediate fields similar to that seen in crystalline Cu-Mn (Ref. 14) and Au-Fe (Ref. 15) and saturates at the IRM value at large-enough fields. Both the TRM and IRM are time dependent and this is illustrated in Figs. 4(b) and 5(b) which show these magnetizations after 1 and 15 min.

In our experiments, the time taken to remove the field increases with the magnitude of field applied and is ~ 30 s at 10 kOe. This difference in times for reducing the applied field to zero may modify the shape of the TRM curve at short times ($\lesssim 1$ min) since the magnetization shows a decay with time. This decay is logarithmic in our samples up to the longest times studied so that at longer times one can expect these initial time differences to quickly become negligible in determining the shape of the TRM as a function of applied field. The TRM maximum at 4.2 K (after 15 min) becomes progressively larger compared to the saturation TRM as x decreases. Both the TRM and IRM become smaller with increasing temperature and the field below which hysteretic behavior is present decreases as shown in Fig. 4 for the $x = 68$ sample. This behavior is reminiscent of the de Almeida–Thouless line of transitions in the onset of irreversibility in finite fields, $H_c(T)$ predicted by replica symmetry breaking mean-field theory.[16]

Mean-field models incorporating exchange fluctuations[17] have predicted a mixed low-temperature phase in which transverse SG order coexists with longitudinal FM order. In our $x = 72$ and 68 samples, our field-cooled magnetization, which we have argued already is our equilibrium magnetization, extrapolates smoothly from low fields to the origin as shown in Figs. 3(a) and 4(a) indicating the absence of an equilibrium macroscopic magnetic moment in zero applied field. The absence of a significant macroscopic moment at low temperatures may also be seen from Fig. 1 where the field-cooled magnetization becomes small at low temperatures in a field of 20 Oe.

In summary, we have shown that the approach to saturation is of the form $1/H^{1/2}$ at intermediate fields in the alloys of this work. The TRM and IRM are time dependent and the TRM has a maximum which becomes more pronounced in the more dilute alloys. The time dependence is logarithmic up to longest times studied (~ 12 h). The field-cooled (equilibrium) magnetization extrapolates smoothly to zero indicating the absence of macroscopic longitudinal FM moment.

ACKNOWLEDGMENTS

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