# Evidence of spin-density-wave to spin-glass transformation in *Y*Nd alloys

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Low-temperature ac and dc magnetic susceptibility and specific-heat measurements have been carried out on *Y*Nd dilute alloys, where the Nd concentrations are 1.9, 2.5, 4.5, 6.8, and 9.0 at. %. The most concentrated alloys (4.5, 6.8, and 9.0 at. %) present long-range antiferromagnetic spin-density-wave (SDW) order below the critical temperature, but some residual frustration is likely to produce a reentrant behavior at low temperatures, where the magnetic data show strong irreversibility effects and the specific heat indicates the occurrence of an anisotropy induced gap near zero frequency in the density of states of the magnetic excitations. For the less concentrated alloys (1.9 and 2.5 at. %), the interacting magnetic state shows typical features of spin-glass behavior. The evolution to this state is ascribed as being due to impurity disorder and frustration which induces a breaking up of the SDW coherence into small domains.

## I. INTRODUCTION

The detailed knowledge of the microscopic interactions of magnetic moments in spin glasses is still a debatable topic, despite the great progress made in the last decades in understanding the macroscopic physical behavior of these systems.[1] In contrast to the idealized picture of frozen-in disorder (where the spins are aligned at random directions) clustering or some other reminiscent short-range magnetic alignments have been assumed to have a significant influence in some properties observed experimentally in spin glasses.[2]

Within metallic systems, a new approach was proposed some years ago where a spin-glass (SG) type of behavior is considered to result from a short-range and "frustrated" spin-density-wave (SDW) state.[3] Such a state was suggested following results of magnetic neutron-scattering experiments performed on the archetypal SG system *Cu*Mn.[5] Instead of ordering in a real long-range and incommensurate SDW state, a system like *Cu*Mn would freeze into a glassy state where some of the SDW characteristics are still present, though the arrangement of spins breaks up into small domains. If this picture is indeed valid one might wonder about the possibility of existing materials presenting intermediate situations between the very "short-range" SDW spin-glass state and the "long-range" antiferromagnetic SDW state.[4] Furthermore, a related topic would be the possibility of a phase transition between these two extreme states.[6] Candidates for being described within this scheme are the *Y*-RE (rare-earth) dilute alloys. These have been known for a long time to stabilize in helical spin structures when the RE concentration is high enough. On the other hand, magnetic irreversibilities characteristic of SG are found in the dilute limit of systems such *Y*Tb, *Y*Dy, and *Y*Er.[7] This topic has been controversial since it has been clearly demonstrated from neutron scattering and other macroscopic experiments that *Y*Gd stabilizes in a long-range transverse helical polarized-SDW state, even in concentrations as low as 1.5 at. % Gd.[8,9] Moreover, evidence from neutron-diffraction experiments for modulated and long-range spin structures have been detected in *Y*Dy,[10] and *Y*Er,[11] even in the very dilute limit.

Within this pattern, the *Y*Nd is an interesting system to investigate because of its magnetic ground-state properties and its crystal-field levels scheme. A preliminary study of the whole *Y*-Nd magnetic phase-diagram was presented by Sharif and Coles.[12] Performing transport and magnetic measurements, these authors report on helical ordering above 12 at. % Nd and wonder about the possibility of SG-type behavior occurring below this concentration.

In the present work low-temperature specific-heat and magnetic-susceptibility (ac and dc) experiments were performed on a series of *Y*Nd alloys in order to better study the detailed ground-state (GS) properties of this system in the low concentration limit. Some preliminary results on *Y*Nd samples, with concentrations of 4.5, 6.8, and 9.0 at. %, are consistent with the long-range antiferromagnetic order SDW description, but some residual frustration is likely to produce a reentrant behavior at low temperatures.[13]

This paper is organized as follows. The experimental procedure and the bare results are presented in Sec. II. In Sec. III A, the magnetic contribution to the specific heat is extracted after a critical analysis of the crystal-field scheme reported in the literature.[14] In Sec. III B, the magnetic specific-heat and the susceptibility results are discussed in the light of the existing descriptions for the SDW and SG states. A magnetic phase diagram is proposed in Sec. III C, where not only the SDW and SG phases are present, but a reentrance from the SDW to a mixed SDW-SG state is also identified. The concluding remarks are given in Sec. IV.

## II. EXPERIMENT AND RESULTS

Samples with Nd concentrations 1.9, 2.5, 4.5, 6.8, and 9.0 at. %, of about 1.5 g, were prepared by arc-melting the proper amounts of pure elements in a Zr-gettered Ar atmosphere. The starting *Y* and Nd were of 4 and 3*N* purity, respectively. They were remelted and rapid cooled several

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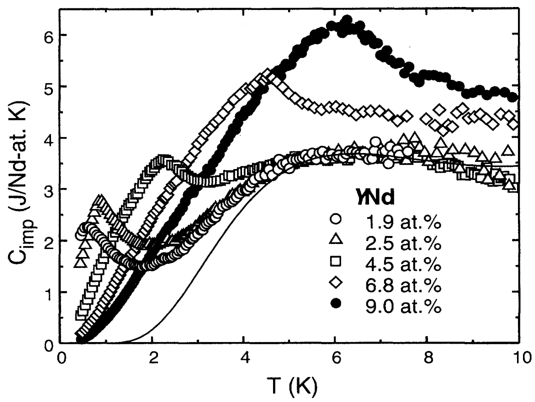


FIG. 1. Impurity specific heat per Nd atom in the $T<10$ K range. The solid line corresponds to a calculated five-doublet Schottky anomaly which is independent of concentration.

times in order to assure good homogeneity. After this procedure the mass losses were found to be negligible. The homogeneity and concentration of the samples were analyzed by means of the wavelength dispersive spectroscopy technique. The good homogeneity of the samples were confirmed and the concentration of the solute were observed to be within 2% of the nominal concentration. Due to the small deviations in concentration, the samples will be referred throughout this paper as for their nominal concentration of solute (i.e., 1.9, 2.5, 4.5, 6.8, and 9.0 at. %).

The specific-heat $C_P$ measurements were performed in a semiadiabatic $^3$He calorimeter using the standard heat-pulse method in the $0.4\text{ K} < T < 30$ K range. The zero-field-cooling (ZFC) and field-cooling (FC) dc magnetic-susceptibility $\chi_{dc}$ experiments were performed in a Foner magnetometer operating in a temperature interval that ranges from 2 to 20 K. The ac magnetic susceptibility $\chi_{ac}$ was measured using the mutual inductance method, with a mutual inductance bridge which operates at a fixed frequency of 128 Hz. The amplitude of the ac driving field of $\approx 10\ \mu$T assures linear response regime. The field dependence of $\chi_{ac}$ was measured under an external applied field of $\approx 10$ mT.

The $C_P(T)$ of a pure Y sample (of the same batch used for the preparation of the alloys) was measured in order to take into account the proper Nd-impurity contribution to the specific heat. Very good agreement was found between the values of the Sommerfeld coefficient and the Debye temperature extracted from the $C_P(T)$ measurement and those reported in the literature.[15]

The impurity contribution to the specific heat $C_{imp}$ was obtained after subtracting the contribution of the Y matrix and normalizing by the Nd concentration, as $C_{imp}(T) = [C_P(Y\text{Nd}) - C_P(\text{Y})]/\text{Nd at. }\%$. They are shown in Fig. 1 in a $C_{imp}$ vs $T$ plot. The transition temperatures, taken as the temperature of the inflexion point above the maximum of $C_{imp}(T)$, are shown in Table I. From the nature of the respective GS, these temperatures are represented as $T_g$ for the 1.9 and 2.5 at. % samples and $T_N$ for the 4.5, 6.8, and 9.0 at. % ones.

Two representative results of the magnetic susceptibility for high and low Nd concentration samples are shown in Fig.

TABLE I. Summary of the characteristic data of the studied $Y$Nd alloys. $T_g$: transition temperatures for the 1.9 and 2.5 at. % samples, and $T_N$: for the 4.5, 6.8, and 9.0 at. % Nd; $\Delta S_M$: magnetic entropy gain in $R\ln 2$ units evaluated at $T_g$ or $T_N^{\dagger}$; $a$ and $\Delta E/k_B$: fitting parameters of $C_M = aT\exp(-\Delta E/k_BT)$ to the magnetic contribution to the specific heat at low temperatures.

| at. % Nd | $T_g$ or $T_N^{\dagger}$ [K] | $\Delta S_M$ [$R$ ln2] | $a$ [mJ/mol K$^2$] | $\Delta E/k_B$ [K] |
|---|---|---|---|---|
| 1.9 | 0.85 | 0.36 | – | – |
| 2.5 | 1.2 | 0.40 | – | – |
| 4.5 | 2.6$^{\dagger}$ | 0.57 | 112 | 0.34 |
| 6.8 | 5.0$^{\dagger}$ | 0.59 | 117 | 0.71 |
| 9.0 | 6.6$^{\dagger}$ | 0.52 | 122 | 1.05 |

2. The dc magnetic susceptibility for the 9.0 at. % alloy is shown in Fig. 2 (top). It shows paramagnetic behavior above the Néel temperature $T_N$, taken as $T_N = \partial(\chi T)/(\partial T)_{\max}$, which coincides with the temperature of the inflexion point of $C_{imp}$. This is an indication of a mean-field-type antiferromagnetic (AF) transition occurring in this sample. The ZFC and FC curves slightly split apart near the maximum of $\chi_{dc}$, and at temperatures below $\approx 0.4\ T_N$ the ZFC-FC separation becomes markedly pronounced, indicating the onset of strong irreversibility effects. The AF character of the transition at $T_N$ is confirmed by the temperature dependence of both components of $\chi_{ac}(T)$. The inductive component ($\chi'$) shows the same temperature dependence of $\chi_{dc}(T)$ and the dissipative component ($\chi''$) is independent of temperature at $T_N$ (as expected for an AF transition), but the onset of $\chi''$ at low frequency starts at 3 K. On the other hand, the dc sus-

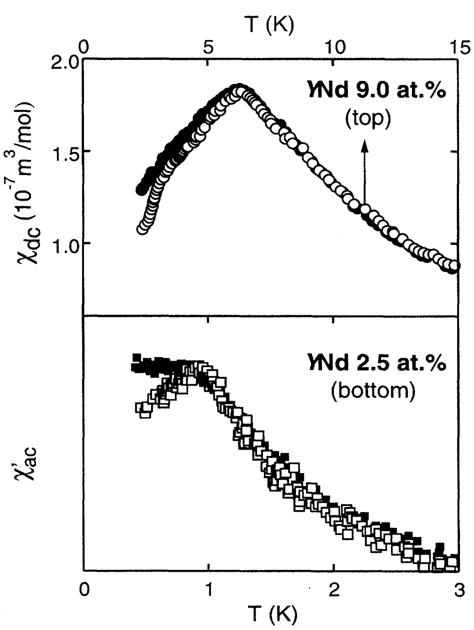


FIG. 2. (Top) Zero-field-cooled (○) and field-cooled (●) magnetic susceptibility for $Y$Nd 9 at. % under an external magnetic field of 10 mT. (Bottom) Inductive component of $\chi_{ac}$ for $Y$Nd 2.5 at. % under $B=0$ T (□) and $B=10$ mT (full □) in the $T<3$ K range.

ceptibility results for the 6.8 at. % sample (not shown) look quite similar to those of Fig. 2 (top), but the irreversibility effects start below $\cong 0.5\ T_N$.

On the dilute side, the inductive component of $\chi_{ac}$ for the 2.5 at. % sample for $B=0\ T$ (ZFC) and $B=10$ mT (FC) as a function of temperature is shown in Fig. 2 (bottom). The maximum of the ZFC curve coincides with $T_g$, that is also the temperature below which both curves begin to separate. On the contrary to the 6.8 and 9.0 at. % alloys, the ZFC-FC curves separate at the maximum of $\chi_{ac}(T)$, which is a signature of the presence of short-range interactions, therefore that temperature is labeled $T_g$ to differentiate it from the long-range magnetic order temperature ($T_N$) of the higher concentrated samples. For the 1.9 at. % sample (i.e., the more dilute alloy) the maximum of $\chi_{ac}$ (not shown) occurs at 0.53 K, which is about 35% below $T_g$. The observed results for the 1.9 and 2.5 at. % samples are a clear indication of SG behavior occurring in this system at these low concentrations.

## III. DISCUSSION

### A. Crystal field contribution to the specific heat

As can be seen from Fig. 1, the impurity contribution to the specific heat of the low-concentration samples (i.e., 1.9, 2.5, and 4.5 at. %) coincide for $T>5$ K. This is an indication of a concentration-independent low-lying excitation spectrum, characteristic of crystal-field (CF) effects. On the other hand, the transition temperatures of the five alloys is concentration dependent (i.e., increasing as concentration increases) and for the samples with higher concentration (i.e., 6.8 and 9.0 at. %), the magnetic contribution of the first-excited CF level is significant.

The solid line shown in Fig. 1 corresponds to a calculated five-doublet Schottky anomaly which fits the data very well for $T>5$ K. Those doublets are distributed in energy as follows: a first excited level at $\delta=14$ K, two nearly degenerate doublets at 70 K, and the fifth doublet at 100 K. To this respect it is important to make some comments. The CF of RE metals and dilute alloys of RE solutes in nonmagnetic hosts like Y, Sc, and Lu, were thoroughly studied by Touborg *et al.*[14,16] The values of the CF parameters (i.e., the coefficients of the Steven's operators in the CF Hamiltonian) were extracted by fitting theoretical expressions for the paramagnetic susceptibility to experimental results at low fields, which agreement was confirmed by magnetic measurements, including neutron spectroscopy, performed at $T>4$ K.[16] In the particular case of the *Y*Nd system, the obtained CF levels scheme indicates that the lowest states are two doublets separated by 4 K, followed by other two closely spaced excited doublets at 67 and 68 K, and a fifth doublet at 130 K.[14,16] However, the values of those CF parameters are reported with uncertainties ranging between 11 and 47 %.[14] The values of the CF energy levels correspond to the mean values of those CF parameters, but no uncertainties in their respective energies were reported. Although there is good agreement for the energy of the highly excited CF levels evaluated from $C_{imp}(T)$ and $\chi(T)$, the difference between the energy of the first-excited CF level [14 K from $C_{imp}(T)$ and 4 K from $\chi(T)$] is quite important. To study this apparent contradiction with the results of the CF scheme reported by Touborg *et al.*, the CF calculations were reproduced and expressions for the energy levels as a function of the CF parameters were obtained. First, those expressions were evaluated using the mean values of the CF parameters reported by Touborg *et al.* and his CF-level scheme was reproduced. Second, the CF parameters were varied within the values of their standard deviations and checked that there is a low-lying CF-levels scheme that fits much better our specific-heat results.

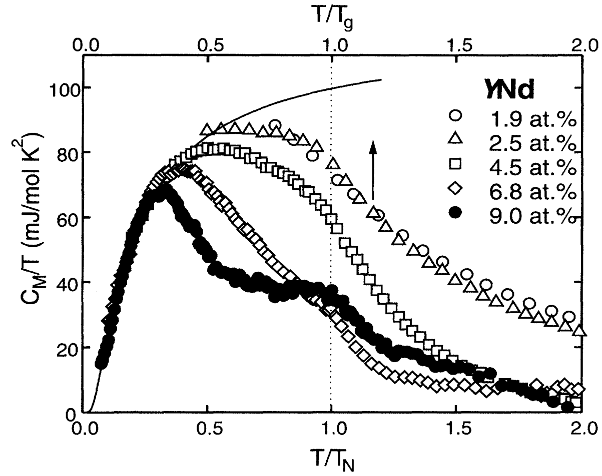


FIG. 3. Magnetic contribution to the specific heat in $C_M/T$ units as a function of the reduced temperature: $T/T_g$ for the *Y*Nd 1.9 and 2.5 at. % (top scale), and $T/T_N$ for the *Y*Nd 4.5, 6.8, and 9.0 at. % samples (bottom scale). The solid line is a fit to Eq. (1), see the text.

The main conclusions regarding this discussion can be summarized as follows. The Schottky contribution to the results in Fig. 1 can be adequately explained by a CF-level scheme slightly different from the one reported by Touborg *et al.*[14,16] This apparent discrepancy can be properly explained by varying the values of the CF parameters within the range of uncertainties reported in Ref. 14. For Nd ions dissolved in a Y host the energy splitting between the GS and the first excited doublet is of $\delta=(14\pm0.5)$ K. One has to take into account that the specific heat is a better experimental technique to study the low-lying CF effects compared to the magnetic susceptibility, because $C_P$ measures the derivative of the internal energy with temperature, whereas $\chi$ does it with respect to magnetic field (and only as a function of temperature). Moreover, the reported $C_P(T)$ was measured within the whole temperature range under discussion, starting from $T<0.1\ \delta$.

### B. Magnetic specific heat

The magnetic contribution to the specific heat $C_M$ was obtained after subtracting the contribution of the Schottky anomaly to $C_{imp}(T)$ as $C_M(T)=C_{imp}(T)-C_{Schottky}(T)$. Figure 3 shows the magnetic contribution in $C_M/T$ units versus reduced temperature, $t=T/T_g$ or $T/T_N$. Some common features are clearly observed: (i) A change in curvature is observed at $t=1$ and all the samples show a positive curvature for $t>1$, that can be attributed to short-range effects occurring above $T_N$ (or $T_g$). (ii) For $t<1$ a well-defined maximum

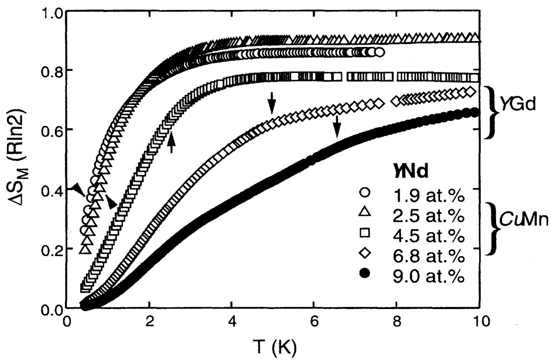


FIG. 4. Magnetic entropy gain as a function of temperature in a $\Delta S_M(T)$ vs $T$ plot for the studied $Y$Nd alloys. The arrows indicate the values of $T_g$ for the 1.9 and 2.5 at. % samples, and $T_N$ for the 4.5, 6.8, and 9.0 at. % ones. The expected values for the archetypes $Y$Gd and $Cu$Mn at the ordering temperature are indicated for comparison.

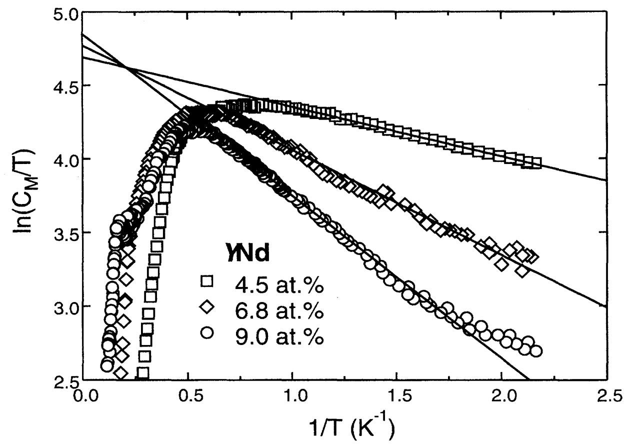


FIG. 5. Plots of $\ln(C_M/T)$, where $C_M$ is the magnetic contribution to the specific heat, as a function of $1/T$. Solid lines are fits of $C_M = aT\exp(-\Delta E/T)$ to the data. The deviation at low temperatures of the data of the 9.0 at. % Nd sample might be due to nuclear contributions.

appears for concentrations above 4.5 at. %, whereas an asymptotic value is observed for the two samples with lower concentrations (i.e., the 1.9 and 2.5 at. % alloys). (iii) For $T<0.3\ T_N$, the $C_M/T$ data for the higher concentration alloys scale as a function of $t=T/T_N$. Unfortunately the $T\rightarrow 0$ limit of the $C_M/T$ data for the 1.9 and 2.5 at. % samples are not well defined (in reduced units) because the values of $T_g$ for these alloys are close to the lowest attainable temperature.

The magnetic entropy gain as a function of temperature is calculated from $C_M(T)$ as $\Delta S_M(T)=\int_0^T C_M/T'dT'$, and it is shown in Fig. 4 in a $\Delta S_M(T)$ vs $T$ plot. The integral was evaluated after linearly extrapolating to zero the values of $C_M(T)$ as $T$ tends to zero. The arrows in Fig. 4 indicate the values of $T_g$ or $T_N$, and the values of $\Delta S_M$ evaluated at those temperatures are shown in Table I. The expected values of $\Delta S_M$ for the archetypes $Y$Gd and $Cu$Mn alloys[3] are also marked in Fig. 4. The values of $\Delta S_M$ at $T=10$ K approach $R$ ln2, that is the expected value for a doublet GS whose degeneracy has been removed. This is another experimental result confirming that the CF contribution was properly taken into account.

Comparing the curves of Fig. 3 to those corresponding to the long-range SDW system $Y$Gd,[9] and the SG $Cu$Mn,[17] some significant analogies are observed. The magnetic specific heat of samples with high Nd concentration shows qualitative as well as quantitative agreement with experimental data and theoretical calculations of long-range helical SDW systems.[9] For instance, the rounded maximum predicted to occur at $T/T_N \cong 0.26$ in SDW magnets and observed in $Y$Gd alloys[9] is found at $T/T_N \cong 0.31$ in the 9 at. % Nd sample and at $T/T_N \cong 0.39$ in the 6.8 at. % Nd alloy. On the other hand, the $C_M$ curves for the samples with 1.9 and 2.5 at. % Nd resemble those of the SG system $Cu$Mn,[17] with a constant value of $C_M/T$ as $T\rightarrow 0$. The shape of $C_M$ for the 4.5 at. % Nd sample looks intermediate between those for the long-range SDW and the SG cases.[2,17] The values of the magnetic entropy gain evaluated at $T_N$ for the most Nd-concentrated samples (see Fig. 4) are close to the value of $\Delta S_M(T_N) \cong (0.6$ to $0.7)R\ \ln 2$ as observed for $Y$Gd,[9] whereas those for the 1.9 and 2.5 at. % Nd alloys approach $\Delta S_M(T_g) \cong 0.3R\ln 2$, typical of spin glasses.[3] This indicates that most of the ordering (or freezing) develops gradually far above $T_N$ (or $T_g$) as short-range magnetic interactions. All this experimental evidence, combined with the magnetic measurements shown in Fig. 2, show that these $Y$Nd alloys behave as long-range SDW systems, at least for Nd concentrations above 4.5 at. %. Decreasing the temperature and solute concentration, the magnetic GS of these alloys evolves to that of a short-range and frustrated SDW system which displays characteristic SG features.

As an attempt to study the nature of the low-lying excitations in the $Y$Nd system, the low-temperature dependence of the magnetic contribution to the specific heat $C_M$ was analyzed. Albeit there is qualitative and quantitative agreement between the archetypes $Y$Gd and $Cu$Mn and the $Y$Nd alloys, a notorious difference arises from the temperature dependence of $C_M$ at low temperatures. Whereas in the $Y$Gd system a concave temperature dependence of $C_M$ was reported at low temperatures,[3] the $C_M$ data for the $Y$Nd samples with higher Nd concentration (i.e., 4.5, 6.8, and 9.0 at. %) scale when plotted as $C_M/T$ vs $T/T_N$ for $T/T_N<0.3$. It is found that the $C_M(T)$ data at low temperatures can be described by

$$C_M = a\ T\exp(-\Delta E/k_B T). \tag{1}$$

This expression was first proposed by Schröder, Löhneysen, and Bauhofer[18] to describe the magnetic specific heat of the anisotropic spin-glass (ASG) system $Eu_xSr_{1-x}As_3$. This type of behavior, which supposes the existence of an anisotropy-induced gap near zero energy in the density of states of magnetic excitations, has also been identified in the rare-earth SG systems $Sc$Er and $Sc$Dy.[19] The $Y$Nd data were analyzed by plotting in Fig. 5 the low-temperature data of the three higher Nd concentration samples in a $\ln(C_M/T)$ vs $1/T$ plot. A linear behavior over an extended $1/T$ range is observed, leading to the values of the fitting parameters $a$ and $\Delta E/k_B$ shown in Table I. As previously found in

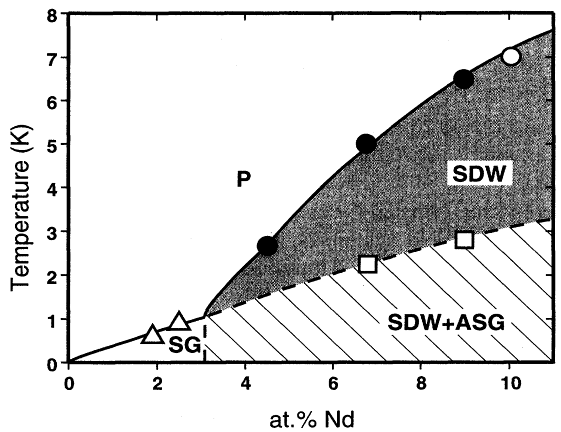


FIG. 6. Magnetic phase diagram of temperature vs concentration of the $Y$Nd system (below 10.0 at. % Nd) constructed with the experimental data: $T_g$ (△) and $T_N$ (●) are extracted from the magnetic susceptibility and specific-heat results. The open squares (□) indicate the onset of strong irreversibilities in the susceptibility data. Four regions are observed: paramagnetic (P) for high temperatures, spin glass (SG) for concentrations below ≈ 3 at. %, a spin-density-wave (SDW) region; and above ≈ 3 at. % Nd and at low temperatures, a region with coexistence of SDW and anisotropic spin glass (ASG) behavior. The phase boundary at ≈ 3 at. % should be vertical according to the mean-field theory (Ref. 26). The point at 10 at. % (○) is after Sharif and Coles (Ref. 12).

$Eu_xSr_{1-x}As_3$,[18] and $Sc$Er and $Sc$Dy alloys,[19] the gap $\Delta E/k_B$ also follows a quasilinear concentration dependence, whereas the coefficient $a$ is found to be almost concentration independent. These results are strong evidence for an ASG-type behavior in the $Y$Nd system at low temperatures. The linearity of the gap with the solute concentration shows that $\Delta E/k_B$ is related to the average exchange energy, as verified in the $Sc$-RE spin glasses.[19] The gap should be originated from anisotropic exchange or from an exchange-induced splitting of the CF-doublet GS. It was observed that for the $Y$Nd studied alloys, the value of $\Delta E/k_B$ is also proportional to the ordering temperature (i.e., $\Delta E/k_B \approx 0.16\ T_N$), as expected from dilution effects, and extrapolates to zero for 2.3 at. % Nd. For this reason, and from the fact that the parameter $a$ is rather independent of concentration, the expression of Eq. (1) is concentration independent when expressed as a function of reduced temperature [i.e., $C_M = a\ T\exp(-\Delta E/k_B T_N T_N/T)$] as is shown in Fig. 3.

The results of Figs. 3 and 5 indicate that the description of a magnetic GS of the more concentrated $Y$Nd alloys includes some characteristics of SG behavior at the lowest temperatures. Further evidence in this sense is furnished by the ZFC-FC splitting of $\chi_{dc}$ [see Fig. 2 (top)] which becomes more pronounced at temperatures below $0.3-0.5\ T_N$, in coincidence with the onset of the dissipative signal of $\chi_{ac}$.

### C. Magnetic phase diagram

The proposed "temperature vs Nd concentration" phase diagram is depicted in Fig. 6, which is constructed from the specific-heat and magnetic-susceptibility results. For concentrations higher than 4.5 at. % the samples clearly undergo an AF transition at $T_N$ to a long-range ordered state, most probably of the SDW type. Some frustration should occur in this state since a weak ZFC-FC irreversibility in the susceptibility is observed below the ordering temperature. However, at temperatures 0.3 to 0.5 $T_N$ approximately, disorder and frustration effects are enhanced, as indicated by the strong ZFC-FC irreversibility observed in $\chi_{dc}$ and the onset of dissipation in $\chi_{ac}$. Below those temperatures a gap opens in the density of states of magnetic excitations and the magnetic contribution to the specific heat shows the same temperature dependence observed in ASG systems. In this concentration region, the behavior of the system as a function of temperature is representative of an AF-SG reentrant phenomenon. The $Y$Nd alloys seem to represent the first example where the reentrant phenomenon is observed in the dilute limit of a metallic binary solid solution. For the most dilute alloys (1.9 and 2.5 at. %), the SG is the only state that stabilizes below the paramagnetic phase. It seems natural to interpret this state as a realization of the short-range-SDW spin glass introduced by Mydosh.[3] As a consequence of dilution, the long-range coherence of the SDW cannot be stabilized and the system undergoes a transformation to a state characterized by small magnetic domains, where the SDW propagates only for relatively short distances. According to the mean-field theory,[20] the pure SG state should be distinct from the reentrant-SG phase. A vertical phase boundary separating these two phases was tentatively added, as shown by the vertical line at concentrations around 3 at. % in the diagram of Fig. 6.

Relative to its ferromagnetic analog, the AF-SG reentry behavior is less common. The best studied cases are the Ising insulating systems $Fe_{0.55}Mg_{0.45}Cl_2$,[21] and $Fe_xMn_{1-x}TiO_3$.[22] Antiferromagnetic-SG transitions have also been observed in concentrated metallic alloys as $Cr_{1-x}Fe_x$ alloys,[23] NiMn,[24] and $Fe_xNi_{80-x}M_{20}$ ($M$= Cr, Mn),[25] or in the pseudobinary compounds $R_xY_{1-x}$Ag ($R$= Tb, Gd).[26] It is noted, however, that the detailed description of the reentrant phase is still a matter of controversy. Most of the authors recognize it as a mixed state where the ferromagnetic or AF phase coexist with the SG. Nevertheless, at least in some cases[25] there are evidences that the long-range AF ordering is lost in the reentrant phase. Thus, for the moment it cannot be not definitely excluded that the reentry behavior in the $Y$Nd alloys would also correspond to a breaking up of the long-range coherence of the SDW into small domains, as much as in the description of the "pure" SG phase. Neutron-diffraction experiments would be the clue to clarify this point.

## IV. CONCLUSIONS

Low-temperature specific heat and magnetic susceptibility (ac and dc) were measured on a series of dilute $Y$Nd alloys, where the concentrations are 1.9, 2.5, 4.5, 6.8, 9.0 at. %. The magnetic contribution to the specific heat has been extracted after a detailed analysis of the Schottky contribution which ended with a different set of CF low-lying energy levels than reported in the literature.

The magnetic specific heat for the most concentrated alloys (4.5, 6.8, and 9.0 at. %) at temperatures near $T_N$ are qualitative and quantitatively consistent with that of a helical

SDW system. However, the low-temperature data are quite well fitted by the exponential law of Eq. (1), which describes the specific heat of ASG systems. This type of behavior supposes the existence of a gap near zero energy in the density of states of the magnetic excitations. The marked enhancement of the ZFC-FC splitting of $\chi_{dc}$ and the onset of dissipation of $\chi_{ac}$ at low temperatures suggest the occurrence of the reentrant phenomenon in these SDW antiferromagnets.

For the less concentrated studied alloys (1.9 and 2.5 at. %), the interacting magnetic state shows typical features of the SG behavior. The evolution to this state is ascribed as being due to impurity disorder and frustration which induces a breaking up of the long-range SDW coherence into small domains. This picture was earlier suggested[3] as a possible description for the microscopic spin arrangement occurring in some metallic SG systems.

The magnetic phase diagram which results from our studies, shown in Fig. 6, appears improved in many respects when compared to that formerly proposed by Sharif and Coles.[12] In particular, a pure SG phase is attained at much lower concentration than the 10 at. % Nd previously suggested.[12] The recent discoveries of reentry behavior in antiferromagnets show that the reentrant phenomenon represents a compromise, not only between the SG and the ferromagnetic order, but is symmetric with respect to the nature of the long-range coherent magnetic state, from which the SG state emerges. Particularly, in our *Y*Nd alloys the magnetic phase diagram of Fig. 6 looks as a SDW counterpart to the one reported for the classical ferromagnetic-SG system *AuFe*.[27] The *Y*Nd system is, therefore one of the few cases where short-range (SG), long-range (SDW), and reentrance can be continuously driven by concentration and temperature variation.

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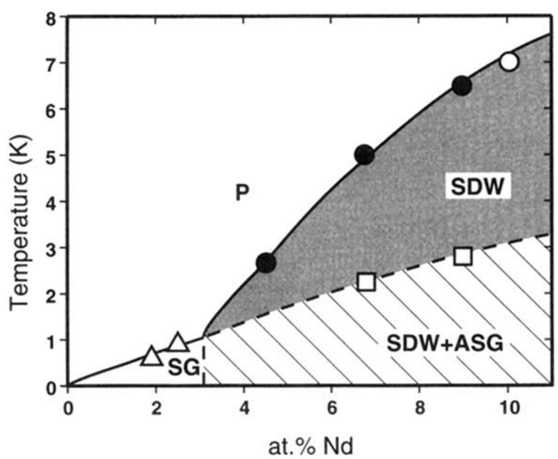


FIG. 6. Magnetic phase diagram of temperature vs concentration of the *Y*Nd system (below 10.0 at. % Nd) constructed with the experimental data: $T_g$ (△) and $T_N$ (●) are extracted from the magnetic susceptibility and specific-heat results. The open squares (□) indicate the onset of strong irreversibilities in the susceptibility data. Four regions are observed: paramagnetic (P) for high temperatures, spin glass (SG) for concentrations below ≈ 3 at. %, a spin-density-wave (SDW) region; and above ≈ 3 at. % Nd and at low temperatures, a region with coexistence of SDW and anisotropic spin glass (ASG) behavior. The phase boundary at ≈ 3 at. % should be vertical according to the mean-field theory (Ref. 26). The point at 10 at. % (○) is after Sharif and Coles (Ref. 12).