Random magnetic fields and tricritical behavior of diamagnetically dilute antiferromagnets $(Ni_{1-x}Mg_x)O$

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The solid solutions $(Ni_{1-x}Mg_x)O$ are used as an example to show, for the first time, that a random substitution of magnesium ions in the nickel sublattice is manifested as a random-magnetic-field effect. This effect first causes a local magnetic disorder and then leads to a complete disruption of the long-range magnetic order in a topologically infinite antiferromagnetic cluster. A region of tricritical behavior and a cluster spin glass appears.

According to Imry and Ma,[1] a magnetically ordered state with continuous symmetry is unstable with respect to an arbitrarily weak random disordering field in a space of dimensionality $n \geqslant 2$. Serving as this random field might be various distortions of real systems, e.g., an impurity with an antiferromagnetic interaction in a ferromagnetic matrix or a uniform external magnetic field in a diamagnetically dilute antiferromagnet.[2] Proof that the long-range magnetic order is disrupted by a random field comes from the experimental observation of a broadening of coherent magnetic reflections[3] and a lowering of the Néel point[4] in diamagnetically dilute antiferromagnets in a uniform magnetic field.

In the present letter we offer evidence that the diamagnetic impurities can themselves serve as sources of random magnetic fields, by virtue of the formation of a local magnetic field at the diamagnetic ion. This field is directed opposite the local molecular field, which is in turn random by virtue of the nonuniform distribution of magnetic ions in the nearest neighborhood of the impurity. A disruption of the long-range antiferromagnetic order upon diamagnetic dilution has been established in many cases (see, for example, Refs. 5 and 6), but there has been no evidence in favor of the identification of a diamagnetic impurity as a random field. We present such evidence in the present letter, using as an example the effect of a diamagnetic Mg impurity on the antiferromagnetic order in NiO.

The $(Ni_{1-x}Mg_x)O$ system, which is a continuous series of solid solutions with a lattice of the NcCl type, was studied by neutron diffraction and SQUID magnetometry. The test samples were prepared by sintering powders of the constituent oxides. The test samples had the compositions $x = 0.1$, 0.3, 0.4, 0.5, 0.55, 0.6, 0.65, 0.7, 0.8, and 0.9. The neutron-diffraction measurements were carried out over the temperature range 4.2–600 K on a diffractometer ($\lambda = 1.81$ Å) installed in a horizontal channel of an IVV-2M reactor. The reversible and irreversible susceptibilities of the test samples were measured over the temperature range 1.5–300 K with a SQUID magnetometer in very weak magnetic fields (~ 1 mT).

For all the solid solutions with concentrations $x \leqslant 0.6$ we found identical neutron-diffraction patterns. These patterns contained a family of magnetic reflections with a

$(\frac{1}{2}\frac{1}{2}\frac{1}{2})$ parent reflection. From the temperature dependence of this reflection we found the Néel temperature T_N of the test samples, and from data on the intensities of the magnetic reflections at 4.2 K we found the average magnetic moment per atom, $\bar{\mu}$, under the assumption of a magnetic structure with a wave vector $\mathbf{k} = (2\pi/a)(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ (Ref. 7).

Working from the results of these measurements, we constructed a magnetic phase diagram of the $(Ni_{1-x}Mg_x)O$ solid solutions (Fig. 1). On this diagram there are regions of uniform and nonuniform antiferromagnetism and also a region of a spin-glass state. For the uniform antiferromagnetism ($0 \leqslant x \leqslant 0.37$), T_N and $\bar{\mu}$ typically are linear functions of the concentration. In addition, one can visually observe a domain structure on the single crystals of these compositions, because of the existence of a magnetic birefringence, which arises from a spontaneous deformation of the lattice.[9] The concentration dependence of the Néel temperature is described well in this composition region by the following expression, which was found from a calculation of the total energy of the exchange-interacting ion pairs:

$$T_N = \frac{1}{2k} Z_{\text{eff}} J_{\text{eff}} S_{\text{Ni}}^2 (1-x), \tag{1}$$

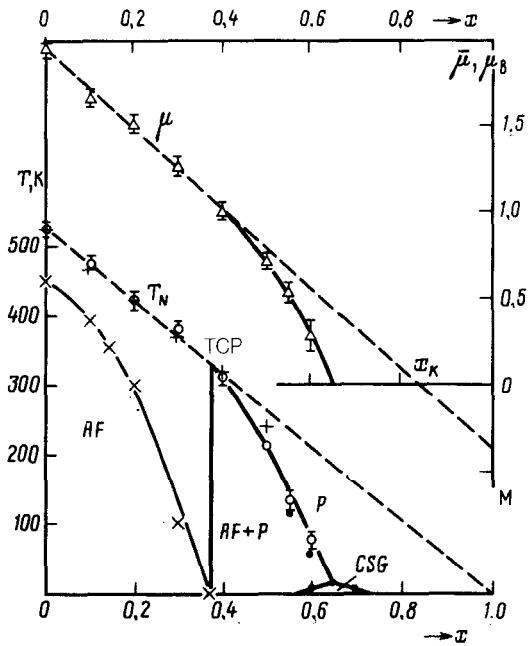

FIG. 1. Bottom: Magnetic phase diagram. Top: Concentration dependence of the average magnetic moment for $(Ni_{1-x}Mg_x)O$ solid solutions. ○, Δ—Data of present study, found by neutron diffraction; ●—data found; by SQUID magnetometry. + —Temperature at which the magnetic susceptibility reaches a maximum according to the measurements of Ref. 8. x—the temperatures up to which a good antiferromagnetic domain structure is observed.[9] Solid lines) Lines of first-order phase transitions; dashed lines) lines of second-order phase transitions; TCP) tricritical point.

where Z_{eff} is the effective number of neighbors, J_{eff} is the effective exchange integral, and S_{Ni} is the spin of the nickel ion. A similar expression can be derived from the mean-field theory.

The agreement between expression (1) and the experimental data at $x \leqslant 0.37$ means that here we have $J_{\text{eff}}^2 S_{\text{Ni}}^2 = \text{const}$. Furthermore, it can be concluded from the weak dependence of the lattice constant on the concentration ($a_{\text{MgO}} = 4.215$ Å, $a_{\text{NiO}} = 4.175$ Å) that the effective exchange integral and the total magnetic moment of the nickel ions, μ_{Ni}, are also constants. Nevertheless, we see from Fig. 1 that the average magnetic moment does not satisfy the relation $\bar{\mu}(x) = \mu_{\text{Ni}}(1-x)$. There is a reason for this circumstance. In order to describe the $\bar{\mu}(x)$ observed experimentally, we need to use a different expression:

$$\bar{\mu}(x) = \mu_{\text{Ni}}(1-x) - Mx, \tag{2}$$

where M is a magnetic moment, antiparallel to the direction of μ_{Ni}, which is induced at the magnesium ions. Its value, $M = 0.35\mu_B$, is found through a linear extrapolation of the experimental results to $x = 1$. Its physical meaning follows directly from the idea of a random magnetic field, as we will now show.

A magnesium ion which replaces a nickel ion cannot be regarded as simply a nonmagnetic atom. Since the electron orbits of the Mg atom rotate around the local mean field, a magnetic field in the opposite direction is produced at the position of this ion. This oppositely directed magnetic field is random with respect to the magnetic order of the matrix because the nickel ions can have more than one type of nearest neighborhood. For magnesium concentrations $x \leqslant 0.37$ the random fields disrupt the magnetic order of the spins in the nearest neighborhood of the diamagnetic ion, which leaving the topologically infinite antiferromagnetic cluster unchanged. The concentration $x_T = 0.37$, however, is critical with respect to the breakup of this infinite cluster into clusters of finite size. At this concentration $T_N(x)$ and $\bar{\mu}(x)$ begin to deviate from linearity; these deviations correspondingly signify the appearance of a certain number of atomic spins which are not involved in either the formation of exchange-coupled pairs or the coherent scattering of neutrons.

On the basis of this behavior of the magnetic properties, we can identify the point $T_N = 325$ K, $x = 0.37$ on the phase diagram as a tricritical point. According to the theory of Ref. 10, the line of second-order phase transitions in the coordinates T–x converts into two lines of first-order phase transitions at this point. Between these two lines, high-symmetry and low-symmetry phases coexist (in the case at hand, these are paramagnetic and antiferromagnetic phases). From the thermodynamic standpoint, the reason for the appearance of this point lies in the interaction of vector and scalar order parameters. Serving as the scalar order parameter here is an elastic distortion of the lattice, which is accompanied by a rhombohedral deformation of the cubic lattice with a compressional axis along the 11 direction in the antiferromagnetic NiO. At a concentration $x > 0.37$, the scalar order parameter disappears, since a domain structure is not observed (because there is no magnetic birefringence).

That a topologically infinite antiferromagnetic cluster is broken up into regions of finite dimensions, between which paramagnetic spins exist, is confirmed by the very

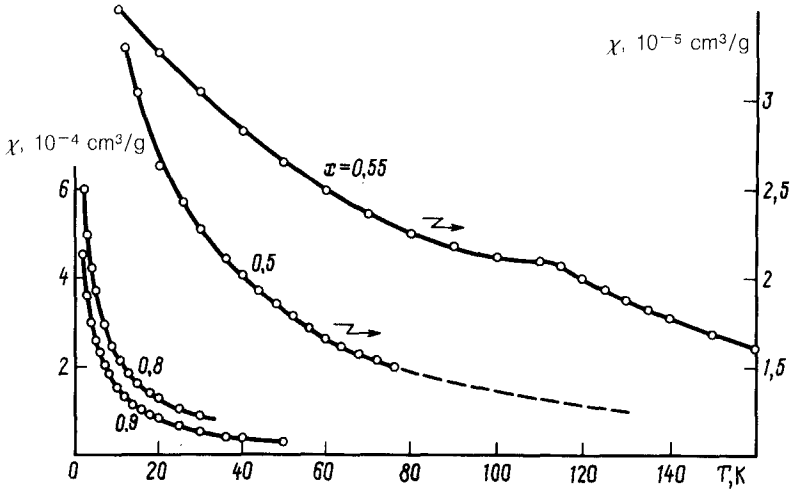

FIG. 2. Temperature dependence of the magnetic susceptibility for $(Ni_{1-x}Mg_x)O$ solid solutions with $x = 0.5$, 0.55, 0.8, and 0.9.

strong increase in the susceptibility below the Néel temperature in the samples with $x = 0.5$, 0.55, and 0.6 (Fig. 2). For uniform antiferromagnets, there is no such increase below T_N (see the inset in Fig. 3).

Near the tricritcal point the size of the finite regions of antiferromagnetic order is

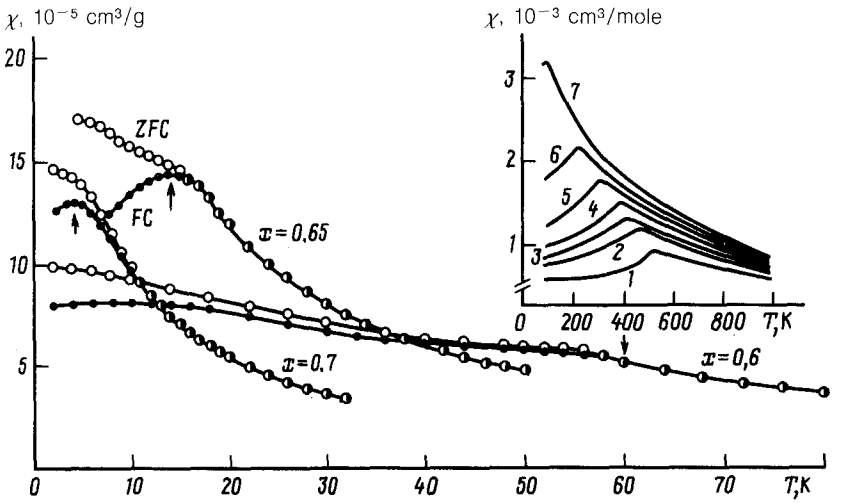

FIG. 3. Temperature dependence of the reversible (open circles) and irreversible (filled circles) magnetic susceptibility of $(Ni_{1-x}Mg_x)O$ solid solutions. ↑—Temperature of transition to spin-glass state, T_f; ↓—Néel temperature. The inset shows curves of $\chi(T)$ from Ref. 8. 1—$x = 0$; 2—0.1; 3—0.2; 4—0.3; 5—0.4; 6—0.5; 7—0.6.

still quite large, since there is no broadening of coherent reflections. At concentrations $x = 0.6$–0.7, however, this size becomes comparable to the region of coherent neutron scattering ($\sim$100–200 Å). Finally, at $x \geqslant 0.7$ the magnetic reflections disappear entirely. The irreversible features of the magnetic susceptibility become large here because of the appearance of a magnetization of clusters with an odd number of antiferromagnetically ordered planes (Fig. 3). The superantiferromagnetic state converts into a cluster-spin-glass state below T_f. At concentrations $x > 0.8$, however, we observe a paramagnetic behavior of the oxide down to liquid-helium temperature (Fig. 2).

[1] Y. Imry and S. Ma, Phys. Rev. Lett. **35**, 1399 (1975).
[2] S. Fishma and A. Aharony, J. Phys. **12**, 729 (1979).
[3] H. Yoshizawa *et al.*, Phys. Rev. Lett. **48**, 438 (1982).
[4] A. V. Drobshnin and Yu. M. Tsipinyuk, Pis'ma Zh. Eksp. Teor. Fiz. **37**, 500 (1983) [JETP Lett. **37**, 597 (1983)].
[5] A. S. Borovik-Romanov *et al.*, J. Magn. Magn. Mater. **31–34**, 1121 (1983).
[6] T. M. Giebultowicz *et al.*, J. Phys. (Paris) **49**, 8 (1988).
[7] W. L. Roth, Phys. Rev. **110**, 1333 (1958).
[8] A. A. Arkhipov, Izv. Akad. Nauk Latv. SSR, Ser. Fiz. Tekhn. Nauk, No. 3, 24 (1981).
[9] N. A. Mironova *et al.*, Ukr. Fiz. Zh. **26**, 848 (1981).
[10] L. D. Landau, Zh. Eksp. Teor. Fiz. **7**, 19 (1937).

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