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Magnetic properties of antiferromagnetic $Ni_{1-x}Mg_xF_2$ and $Ni_{1-x}Zn_xF_2$ single crystals

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Abstract

The magnetic properties of $Ni_{1-x}[Mg, Zn]_xF_2$ with randomly distributed of magnetic ions have been investigated with a vibrating sample magnetometer. At x less than $x_c = 0.75$, below $T_{N(x)}$, the crystals transfer into antiferromagnetic states with weak ferromagnetism $\sigma(T)$. The value of the Dzyaloshinsky-Moriya interaction responsible for $\sigma(T)$ is essentially independent of x.

The peculiarities of the magnetic properties of diluted antiferromagnets have been investigated in tetragonal symmetry $Ni_{1-x}(Mg, Zn)_xF_2$ with the random substitution of magnetic Ni^{2+}/ions for nonmagnetic Zn^{2+} or Mg^{2+} ions. At temperatures $T < T_N = 73$ K, in NiF_2 single crystals an antiferromagnetic state appears with a weak ferromagnetic moment, produced by the Dzyaloshinsky-Moriya interaction. The antiferromagnetic vector $\boldsymbol{L}$ is oriented along binary axis [100] or [010] and the weak ferromagnetic moment σ_D is oriented perpendicular to $\boldsymbol{L}$, [1]. Using a vibrating sample magnetometer, which can measure three perpendicular components of the magnetic moment of the sample, we have investigated the magnetic field and temperature dependences of the magnetic moments of $Ni_{1-x}(Zn,Mg)_xF_2$ single crystals with different nonmagnetic ion concentrations [2,3]. In Ref. [2] we described the classification of the distribution of interacting magnetic ions in these single crystals. Fig. 1 shows the concentration dependences of the antiferromagnetic phase transition temperature $T_N(x)$ of $Ni_{1-x}(Zn,Mg)_xF_2$ [2]. Values of the magnetic ordering temperatures were determined from the maxima of the temperature dependences of the magnetic susceptibility and by the disappearance of the weak ferromagnetic moment at $T_N(x)$. The peculiarities of the magnetic states that appear in these samples increase when x approaches the percolation limit $x_c = 0.75$. No antiferromagnetic order is observed at $x > x_c$, although the magnetic state that appears with decreasing temperature differs from the usual paramagnetic states. At nonmagnetic ion concentrations $x < x_c$ (Fig. 1), the $M(H)$ dependences of the $Ni_{1-x}(Zn,Mg)_xF_2$, when the applied magnetic field is oriented along the binary axis, can be described by the expression $M(H, T) = \sigma_D(T) + \chi_\perp H$, where $\sigma_D(0) = (169 \pm 10)$ CGSM/mol. The ferromagnetic moment $\sigma_D(0)$ does not depend on the nonmagnetic ion concentration x, [2], which is explained by the single-ion nature of the anisotropy interaction responsible for this moment, $-e\Sigma(S_i^xS_i^y - S_j^xS_j^y)$. With increasing nonmagnetic ion concentrations in weak magnetic fields there occurs a nonlinear magnetic field dependence $M(H)$ of the sample magnetic moment. At the same time, the perpendicular magnetic susceptibility grows with decreasing temperature [2]. The magnetic susceptibility in strong magnetic fields (Fig. 1) does not depend on x, which can be explained by the decrease in the exchange interaction between magnetic ions with increasing x and T_N changing from $T_N = 73$ to 8 K (Fig. 1). The nonlinear $M(H, T)$ dependences for

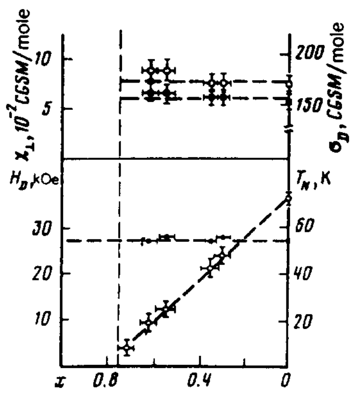

Fig. 1. Antiferromagnetic transition temperature T_N(○), weak ferromagnetic moment σ(□), magnetic susceptibility χ(■), and effective field $H = \sigma / \chi$ (●) as functions of the concentrations of Zn and Mg in $Ni_{1-x}(Zn,Mg)_xF_2$.

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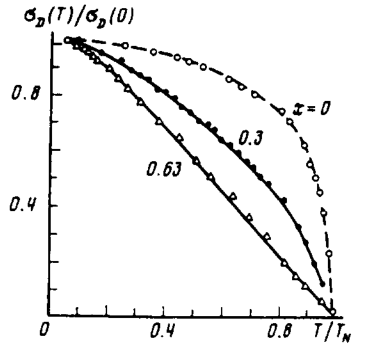

Fig. 2. Reduced ferromagnetic moments $\sigma(T)/\sigma(0)$ for $Ni_{1-x}(Zn,Mg)_xF_2$ single crystals ($x = 0$, 0.3 and 0.63) as a function of the reduced temperature T/T_N.

$Ni_{1-x}(Zn,Mg)_xF_2$, when the magnetic field is oriented in the (001) plane, can be explained by the increasing role of the Dzyaloshinsky–Moriya interaction $H_{DM} = (27.2 \pm 0.5)$ kOe, which, like the weak ferromagnetic moment, is not dependent on the nonmagnetic ion concentration, (Fig. 1). In the sample with $T_N = (8 \pm 0.5)$K, the anisotropy interaction responsible for the ferromagnetic moment is comparable with the exchange interaction.

Besides the nonlinear $M(H)$ dependence and $\chi(T)$ dependence at $T < T_N(x)$ we observed that the low-temperature dependence of weak ferromagnetism $\sigma_D(T)$ changes with increasing of nonmagnetic ion concentrations. Fig. 2 shows the $\sigma_D(T)$ dependence for several $Ni_{1-x}(Zn,Mg)_xF_2$ single crystals, where $\sigma_D(T)/\sigma_D(0)$ is plotted as a function of T/T_N. For NiF_2 crystals at low temperature, the $\sigma_D(T)$ dependence is determined by the spin-wave law $\sigma_D(T) = \sigma_D(0)\ [1 - \alpha(T/T_N)^2]$. As x increases, $\sigma_D(T)$ departs from the corresponding curve for NiF_2. For $Ni_{0.37}Mg_{0.63}F_2$ ($T_N = (19.0 \pm 0.5)$ K) and 3.5 K $< T <$ 18 K, $\sigma_D(T)$ can be approximated by $\sigma_D^*(T) = \sigma_D(0)(1 - \eta T/T_N)$. For $T <$ 3.4 K or 18 K $< T < T_N$, the experimental $\sigma_D(T)$ curve departs slightly from a linear dependence. This temperature dependence of the weak ferromagnetic moment $\sigma_D(T)$ can be explained by the peculiarities of the spin wave propagation in $Ni_{1-x}(Zn,Mg)_xF_2$ single crystals. At antiferromagnetic order in $M_{1-x}(Zn,Mg)_xF_2$ there exists a linear dimension value ξ_c depending on the nonmagnetic ions concentration, which indicates the differences in the magnetic properties at scales lower or higher than ξ_c. Such a model for diluted antiferromagnets was discussed in Refs. [4,5], describing fractal systems. The change in the $\sigma_D(T)$ dependence may be explained by the typical linear dimension ξ_c in the magnetic system and by the typical spin wave frequency ω_c corresponding to ξ_c, which divides the spin wave spectrum into two parts. Spin waves with wavelengths $\lambda > \xi_c$ propagate in the magnetic system, while those with $\lambda < \xi_c$ are localized and attenuate rapidly at scales larger than ξ_c. As shown in Ref. [5], the temperature dependence of the magnetic moments of such a system can be written as:

$$\delta M/M = 1/2M_0^2 \int \frac{\omega\rho\ \inf(\omega)}{(e^{\omega}/T - 1)F(\omega)} d\omega,$$

where $\rho_{\inf}(\omega)$ is the spin wave density in the infinite cluster and $F(\omega)$ is the coefficient connecting the energy density and magnetic moment fluctuations. At sufficiently low temperatures, $kT \ll \hbar\omega_c$, $\delta M/M$ can be written in the usual spin wave form $\delta M/M \propto T^2$. As shown in Ref. [5], at temperatures $\hbar\omega_c \leq kT < kT_N$, the linear temperature dependences of $\delta M/M \propto T$ and of $\sigma_D(T) \propto M(T)$ can occur.

In $M_{1-x}(Zn,Mg)_xF_2$ crystals with nonmagnetic ion concentrations greater than x_c, there is no antiferromagnetic order, and magnetic states that arise with decreasing temperature are determined by the finite correlation length only. These magnetic states determine the $M(H, T)$ nonlinear magnetic field and temperature dependences and the $\chi(T, H)$ temperature dependences in weak magnetic fields [3]. As magnetic clusters arise with superparamagnetic properties, changes in the low-temperature magnetic susceptibility can be observed from $\chi^{-1}(T) \propto (T + \theta)$ to $\chi^{-1}(T) \propto T$ [3]. The magnetic susceptibility observed in strong magnetic fields is determined by the magnetic susceptibility inside the magnetic clusters.

Thus the diluted antiferromagnets $Ni_{1-x}(Zn,Mg)_xF_2$ single crystals are examples of magnetic systems with random distributions of the exchange interaction and of the interaction responsible for the weak ferromagnetism.

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