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Unusual Critical Behavior of the Diluted Uniaxial Dipolar Ferromagnet $LiTb_{0.3}Y_{0.7}F_4$

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The experimental critical behavior of the susceptibility of $LiTb_{0.3}Y_{0.7}F_4$ is described by the power law $\chi = \Gamma[(T-T_c)/T_c]^{-\gamma}$ with $T_c = 0.520 \pm 0.003$ K and $\gamma = 1.80 \pm 0.04$. This behavior is dramatically different from that previously observed in $LiTbF_4$ and is evidence of a departure from marginal dimensionality when magnetic ions have been randomly replaced by nonmagnetic ions. Series expansion of $(\chi T)^{-1}$ in powers of T^{-1} for a diluted Ising dipolar ferromagnet gives a good description of experimental results described in this Letter.

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The critical behavior of pure uniaxial dipolar ferromagnets is one of the best explained critical phenomena. In this case the marginal dimensionality is $d^* = 3$. In the close vicinity of the critical temperature T_c, the magnetic susceptibility is predicted[1] to have logarithmic corrections to the classical law so that it diverges as $t^{-1}|\ln t|^{1/3}$, where t is the reduced temperature $t = (T-T_c)/T_c$. The first higher-order term of the logarithmic corrections has been calculated for all the thermodynamic quantities in zero magnetic field[1] and in the whole critical region for a finite field.[2]

$LiTbF_4$ is a quasiuniaxial dipolar ferromagnet.[3,4] The experimental critical behavior of these crystalline pure compounds[5,6,7] is well described by the theoretically predicted classical behavior with logarithmic corrections.

In diluted uniaxial dipolar ferromagnets where some magnetic ions are randomly replaced by nonmagnetic ones, quite a different behavior was predicted by Aharony.[8] In this Letter we report measurement on the dilute ferromagnet $LiTb_{0.3}Y_{0.7}F_4$ which for the first time clearly shows departure from the behavior expected from a system at marginal dimensionality. The parallel susceptibility of $LiTb_{0.3}Y_{0.7}F_4$ corrected from demagnetizing effects cannot be described by the Aharony law $\chi = \Gamma t^{-1}\exp[D\ln(1/t)]^{1/2}$ with the universal parameter $D \sim 0.11795$ in the temperature range $10^{-3} < t < 10^{-1}$ even by substituting $1/t$ by t_0/t, taking approximately into account high-order terms. As the crossover between "pure" behavior to asymptotic "random" behavior is not well known, we have described our experimental results by a power law with the unusual large effective exponent $\gamma = 1.80$.

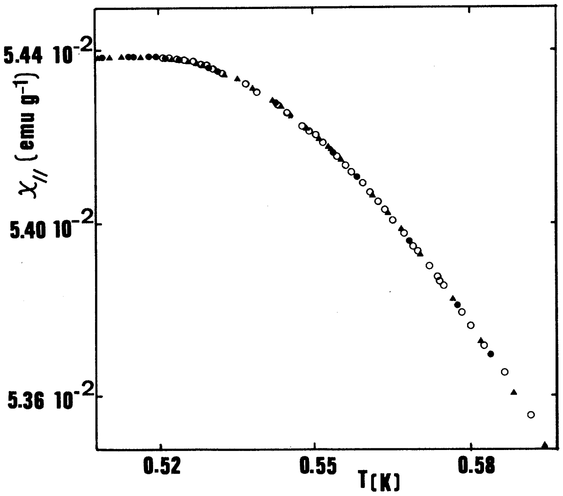

FIG. 1. Parallel susceptibility $\chi_{\parallel}$ (emu g^{-1}) of $LiTb_{0.3}Y_{0.7}F_4$ vs temperature in the critical range 0.52–0.57 K.

A theoretical expansion of the inverse susceptibility in power of $\beta = (kT)^{-1}$ for a diluted uniaxial dipolar ferromagnet gives a good description of these experimental results as well as similar published[9] measurements on $LiTb_{0.5}Y_{0.5}$ where a clear departure from marginal dimensionality was not found.

The sample of optical quality was cut from a single crystal of $LiTb_pY_{1-p}$ grown by the Stockbarger method at the Lyngby Technical University. This was ground into a sphere of 3.995 ± 0.003 mm diameter weighing 149.0 ± 0.2 mg whose measured density yields $p = 0.32 \pm 0.02$.

The magnetic susceptibility parallel to the c crystal axis was measured in the temperature range 0.3–4.2 K.

As shown in Fig. 1, $\chi_{\parallel}$ displays a maximum at $T = 0.520 \pm 0.003$ K. The value of the maximum $\chi_{max} = 5.43 \times 10^{-2}$ emu g^{-1} is in reasonable agreement with the expected value determined by the demagnetizing factor for a ferromagnetic transition $3/4\pi\rho = 5.35 \times 10^{-2}$ emu g^{-1}. Instead of a constant value below T_c, we have a drop of $\chi_{\parallel}$ usually observed in ac susceptibility measurements. This drop is probably due to frictions in the motion of Bloch walls.

In the critical range 0.51–0.65 K, we have performed with a good reproducibility (Fig. 1) three different experimental runs. The statistical treatment of these data was performed with use of the same procedure as for $LiHoF_4$ and $LiTbF_4$ (Ref. 7) which have been studied within the same range of t. The critical susceptibility $\chi_c(t)$ is found to obey the power law $\chi_c(t) = \Gamma t^{-\gamma}$. The experimental data were compared to the theoretical susceptibility $\chi_{theor}(t)$ taking into account the de-

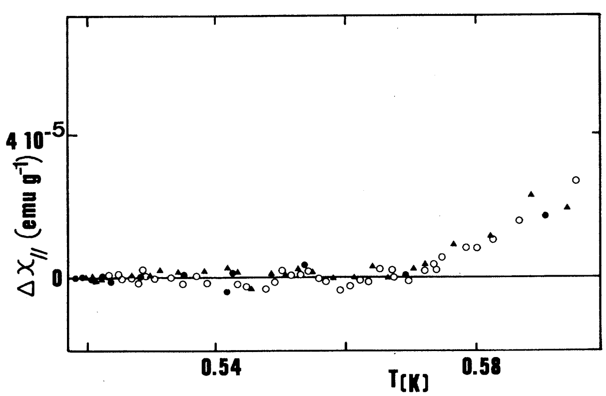

FIG. 2. Discrepancy $\Delta\chi$ between the experimental susceptibility per gram and the theoretical value χ_{theor} vs temperature T. χ_{theor} is calculated with the relation $\chi_{theor}^{-1} = (1/\Gamma)t^{\gamma} + \chi_{max}^{-1}$ and the best fit values of the parameters Γ, T_c, and γ.

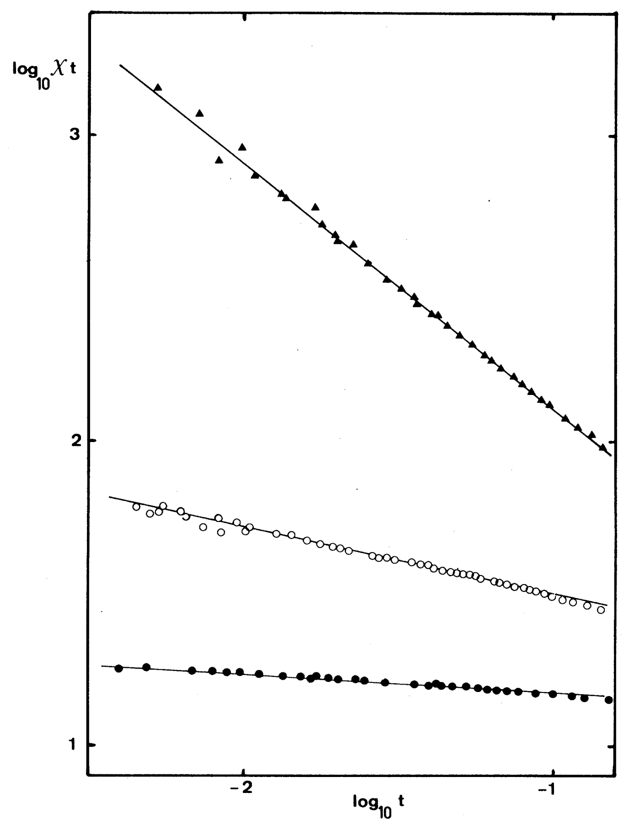

FIG. 3. χt vs t on logarithmic scales for $LiTbF_4$ (solid circles), $LiTb_{0.5}Y_{0.5}F_4$ (open circles), and $LiTb_{0.3}Y_{0.7}F_4$ (solid triangles). χ is the molar parallel susceptibility corrected from demagnetizing effects in electromagnetic units.

magnetizing effect $[\chi_{theor}(t)]^{-1} = \chi_c(t)^{-1} + \chi_{max}^{-1}$. The value of χ_{max} was given by the experimental value of the plateau just below T_c and the other parameters $\Gamma = (10.10 \pm 0.24)\times 10^{-2}$ emu cgs g^{-1}, $T_c = 0.5195 \pm 0.0004$ K, and $\gamma = 1.805 \pm 0.040$ were determined by a nonlinear least-squares fit (Fig. 2). We have studied how the variance of the fit and the parameters T_c and γ vary with the upper limit t_{max} of the critical region. The variance s^2 of the fit has approximately the same value $s^2 = 2.6\times 10^{-14}$ emu up to $t_{max} = 1.1\times 10^{-1}$ and then increases rapidly. The critical region $t < 10^{-1}$ is then the same order of magnitude as for $LiTbF_4$ and $LiTb_{0.5}Y_{0.5}F_4$.[7,8]

The most important result of this study is the unusually high value of the exponent $\gamma = 1.80 \pm 0.04$ for a three-dimensional compound. An earlier study on the more concentrated compound $LiTb_pY_{1-p}F_4$ where $p = 0.51 \pm 0.01$ showed[9] that the parallel susceptibility in the same critical range can be as well described by the classical law with logarithmic corrections, by the Aharony law for a random system, or by a power law with $\gamma = 1.215$. This clearly indicated that corrections to the classical law $\gamma = 1$ are important but did not establish a departure from marginal dimensionality. The striking difference between the critical behavior of pure and diluted $LiTbF_4$ and the unusual exponent γ observed in $LiTb_{0.3}Y_{0.7}F_4$ is illustrated by Fig. 3 where $\ln\chi t$ has been plotted versus $\ln t$ for pure $LiTbF_4$, $LiTb_{0.5}Y_{0.5}F_4$ ($T_c = 1.119$ K, $\gamma = 1.215$), and $LiTb_{0.3}Y_{0.7}F_4$. In order to describe our results in the experimental range $10^{-3} < t < 10^{-1}$ for $LiTb_{0.5}Y_{0.5}F_4$ and $LiTb_{0.3}Y_{0.7}F_4$ we have performed a series expansion of $(\chi T)^{-1}$ in powers of T^{-1} for diluted uniaxial dipolar ferromagnet. After partial summation, the series expansion for the susceptibility χ_c corrected from demagnetizing effect may be written:

$$\frac{\chi_0}{\chi_c} = 1 - p\sum_k \tanh\beta J_{ik} - \sum_{n=2}^{\infty} (p\beta)^n \sigma_n + \sum_{n=2}^{\infty} p^n \sum_{q=n+1}^{\infty} \omega_{nq}\beta^q, \qquad (1)$$

with

$$\beta = (k_B T)^{-1}$$

and

$$J_{ik} = (g^2\mu_B^2/4r_{ik}^3)[1 - 3\cos^2\theta_{ik}]; \qquad (2)$$

σ_n and ω_{nq} are constants which depend only on the crystal lattice and $\chi_0 = C/T$ is the Curie susceptibility for noninteracting moments. In this expansion we have neglected the short-range interactions. At the transition temperature $T_c(p)$ we have $\chi_0/\chi_c = 0$. In the calculation of $T_c(p)$, on account of the long-range interactions in (2), we can neglect the terms $\omega_{nq}\beta^q$ which are all with $q \geq 4$. The term $(p\beta)^2\sigma_2$ will be calculated exactly but the other, much smaller ones are evaluated with the approximation

$$\sum_{n=3}^{\infty} [p\beta_c(p)]^n \sigma_n \sim p\beta_c(p) \sum_{n=3}^{\infty} \sigma_n \equiv p\beta_c(p)\sigma, \qquad (3)$$

where $\beta_c(p) = [k_B T_c(p)]^{-1}$ and σ is a constant which depends only on the crystal lattice.

All the terms in (1) must be calculated in a cylindrical domain where the demagnetizing field is zero; the extension of the cylinder is supposed to be independent of p. We can write

$$\sum_k \tanh(\beta J_{ik}) \equiv \sum_k{}' \tanh(\beta J_{ik}) + \beta J, \qquad (4)$$

where the primed summation must be performed in the center of an infinite sphere. The equation which gives the transition temperature of a diluted dipolar ferromagnet with these approximations is given by

$$\chi_0/\chi_c = 0 = 1 - p\{\sum_k{}' \tanh[\beta_c(p)J_{ik}] + \beta_c(p)S + p\beta_c^2(p)\sigma_2\}, \qquad (5)$$

with $S = \sigma + J$ and $\sigma_2 = -\sum_k J_{ik}^2$. The dipolar summations have been performed on a UNIVAC-1110 computer for the crystal lattice of $LiTbF_4$; the value of S has been adjusted in order to obtain the observed value $T_c(1) = 2.90$ K. The values of $T_c(p)$ obtained from this series expansion for $LiTb_{0.5}Y_{0.5}F_4$ and $LiTb_{0.3}Y_{0.7}F_4$ are given in Table I. We can also observe the value of the critical exponent $\gamma(p)$ which results if one retains only the first terms in (5) and which is given by the relation $p\sum_k J_{ik} = \gamma(p)k_B T_c(p)$. So $\gamma(p)$ to this

TABLE I. Comparison between the theoretical series-expansion values and the experimental value of the transition temperature $T_c(p)$ and the critical susceptibility exponent $\gamma(p)$ for two diluted uniaxial dipolar ferromagnets $LiTb_pY_{1-p}F_4$.

p	$T_c(p)_{theor}$	$T_c(p)_{expt}$	$\gamma(p)_{theor}$	$\gamma(p)_{expt}$
0.51 ± 0.01	1.220 ± 0.025	1.119 ± 0.004	1.21	1.215 ± 0.01
0.32 ± 0.02	0.64 ± 0.04	0.520 ± 0.003	1.45	1.80 ± 0.04

order is given by

$$\gamma(p) = pT_c(1)/T_c(p). \tag{6}$$

The predicted values of $\gamma(p)$ calculated with the theoretical value of $T_c(p)$ are given in Table I. In this table we have a very good description of the experimental result in $LiTb_{0.5}Y_{0.5}F_4$. In $LiTb_{0.3}Y_{0.7}F_4$, the condition $T_c(p)/pT_c(1) = 0.56$ close to 1 is not satisfied $[T_c(p)/pT_c(1) = 0.75$ in $LiTb_{0.5}Y_{0.5}F_4]$ and in this compound the agreement with the experimental results is not so good.

With this experimental investigation of the critical behavior of $LiTb_{0.3}Y_{0.7}F_4$, we have clearly demonstrated the departure from marginal dimensionality when magnetic ions have been randomly replaced by nonmagnetic ions in a dipolar uniaxial ferromagnet. For observable temperature distances from the critical point, the effective critical exponent $\gamma(p)$ becomes nonuniversal and depends on the impurity concentration.[10] Series expansion of $(\chi T)^{-1}$ in powers of T^{-1} for a diluted Ising dipolar ferromagnet gives a good description of our experimental results.

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