Unusual Critical Behavior of the Diluted Uniaxial Dipolar Ferromagnet $\text{LiTb}_{0.3}\text{Y}_{0.7}\text{F}_4$

Seiden, P.; Seiden, J.; Laursen, Ib

Published in:
Physical Review Letters

Link to article, DOI:
[10.1103/PhysRevLett.45.1362](https://doi.org/10.1103/PhysRevLett.45.1362)

Publication date:
1980

Document Version
Publisher's PDF, also known as Version of record

[Link back to DTU Orbit](#)

Citation (APA):
Seiden, P., Seiden, J., & Laursen, I. (1980). Unusual Critical Behavior of the Diluted Uniaxial Dipolar Ferromagnet $\text{LiTb}_{0.3}\text{Y}_{0.7}\text{F}_4$. *Physical Review Letters*, 45(16), 1362-1365.
<https://doi.org/10.1103/PhysRevLett.45.1362>

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

- ¹J. M. Luttinger, J. Math. Phys. **4**, 1154 (1963).
²H. A. Bethe, Z. Phys. **71**, 205 (1931).
³A discussion of this property from a field-theory standpoint is given by A. B. Zamolodchikov and A. B. Zamolodchikov, Ann. Phys. (N. Y.) **120**, 253 (1979).
⁴A. Luther and I. Peschel, Phys. Rev. B **12**, 3908 (1975).
⁵A. Luther and I. Peschel, Phys. Rev. B **9**, 2911 (1974).
⁶See C. N. Yang and C. P. Yang, Phys. Rev. **150**, 321, 327 (1966), for a detailed discussion of this model, and earlier references.
⁷P. Jordan and E. Wigner, Z. Phys. **47**, 631 (1928); see also Ref. 4.
⁸E. H. Lieb and F. Y. Wu, Phys. Rev. Lett. **20**, 1445 (1968).
⁹D. C. Mattis and E. H. Lieb, J. Math. Phys. **6**, 304 (1965).
¹⁰R. Heidenreich *et al.*, Phys. Lett. **54A**, 119 (1975).
¹¹F. D. M. Haldane, J. Phys. C **12**, 4791 (1979).
¹²D. C. Mattis, J. Math. Phys. **15**, 609 (1974).
¹³This ensures that the ground state of (1) has finite overlap with the original Fermi-sea state of the free system.
¹⁴F. D. M. Haldane, to be published.
¹⁵M. Fowler and M. W. Puga, Phys. Rev. B **18**, 421 (1978).
¹⁶J. D. Johnson *et al.*, Phys. Rev. A **8**, 2526 (1973).
¹⁷The spin model has a modified selection rule on J : $(-1)^J = +1$.
¹⁸Limiting behavior found numerically was (i) $\frac{1}{4}\pi[4 - \exp(-2\varphi)]/v_J \rightarrow 1$, independent of Δ , for $n \rightarrow \frac{1}{2}$, $\Delta < -1$; (ii) $\frac{1}{4}\pi v_N/(1-\Delta) \rightarrow 1$, independent of n , as $\Delta \rightarrow 1^-$.
¹⁹V. J. Emery *et al.*, Phys. Rev. B **13**, 1262 (1976).
²⁰R. B. Griffiths, Phys. Rev. **133**, A768 (1964).

Unusual Critical Behavior of the Diluted Uniaxial Dipolar Ferromagnet $\text{LiTb}_{0.3}\text{Y}_{0.7}\text{F}_4$

P. Beauvillain and J. Seiden

Institut d'Electronique Fondamentale, Laboratoire associé au Centre National de la Recherche Scientifique, Université Paris-Sud, F-91405 Orsay, France

and

I. Laursen

Department of Electrophysics, The Technical University of Denmark, DK-2800 Lyngby, Denmark

(Received 2 April 1980)

The experimental critical behavior of the susceptibility of $\text{LiTb}_{0.3}\text{Y}_{0.7}\text{F}_4$ 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.

PACS numbers: 75.50.Dd, 75.40.-s

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 predicted¹ 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 field¹ 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 $\text{LiTb}_{0.3}\text{Y}_{0.7}\text{F}_4$ which for the first time clearly shows departure from the behavior expected from a system at marginal dimensionality. The parallel susceptibility of $\text{LiTb}_{0.3}\text{Y}_{0.7}\text{F}_4$ 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 $\chi_{||}$ (emu g^{-1}) of $\text{LiTb}_{0.3}\text{Y}_{0.7}\text{F}_4$ 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 published⁹ measurements on $\text{LiTb}_{0.5}\text{Y}_{0.5}$ where a clear departure from marginal dimensionality was not found.

The sample of optical quality was cut from a single crystal of $\text{LiTb}_p\text{Y}_{1-p}$ 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

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 $\chi_{\text{theor}}^{-1} = (1/\Gamma)t^\gamma + \chi_{\text{max}}^{-1}$ and the best fit values of the parameters Γ , T_c , and γ .

crystal axis was measured in the temperature range 0.3–4.2 K.

As shown in Fig. 1, $\chi_{||}$ displays a maximum at $T = 0.520 \pm 0.003$ K. The value of the maximum $\chi_{\text{max}} = 5.43 \times 10^{-2} \text{ emu g}^{-1}$ is in reasonable agreement with the expected value determined by the demagnetizing factor for a ferromagnetic transition $3/4\pi\rho = 5.35 \times 10^{-2} \text{ emu g}^{-1}$. Instead of a constant value below T_c , we have a drop of $\chi_{||}$ 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 $\chi_{\text{theor}}(t)$ taking into account the de-

FIG. 3. χt vs t on logarithmic scales for LiTbF_4 (solid circles), $\text{LiTb}_{0.5}\text{Y}_{0.5}\text{F}_4$ (open circles), and $\text{LiTb}_{0.3}\text{Y}_{0.7}\text{F}_4$ (solid triangles). χ is the molar parallel susceptibility corrected from demagnetizing effects in electromagnetic units.

magnetizing effect $[\chi_{\text{th cor}}(t)]^{-1} = \chi_c(t)^{-1} + \chi_{\text{max}}^{-1}$. 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 $t_{\text{max}} = 1.1 \times 10^{-1}$ and then increases rapidly. The critical region $t < 10^{-1}$ is then the same order of magnitude as for LiTbF_4 and $\text{LiTb}_{0.5}\text{Y}_{0.5}\text{F}_4$.^{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 $\text{LiTb}_p\text{-Y}_{1-p}\text{F}_4$ where $p = 0.51 \pm 0.01$ showed⁹ 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 $\text{LiTb}_{0.3}\text{Y}_{0.7}\text{F}_4$ is illustrated by Fig. 3 where $\ln \chi t$ has been plotted versus $\ln t$ for pure LiTbF_4 , $\text{LiTb}_{0.5}\text{Y}_{0.5}\text{F}_4$ ($T_c = 1.119$ K, $\gamma = 1.215$), and $\text{LiTb}_{0.3}\text{Y}_{0.7}\text{F}_4$. In order to describe our results in the experimental range $10^{-3} < t < 10^{-1}$ for $\text{LiTb}_{0.5}\text{Y}_{0.5}\text{F}_4$ and $\text{LiTb}_{0.3}\text{Y}_{0.7}\text{F}_4$ 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:

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

with

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

and

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

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

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

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

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

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

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

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

$\text{LiTb}_{0.5}\text{Y}_{0.5}\text{F}_4$ and $\text{LiTb}_{0.3}\text{Y}_{0.7}\text{F}_4$ 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 $\text{LiTb}_p\text{Y}_{1-p}\text{F}_4$.

p	$T_c(p)_{\text{theor}}$	$T_c(p)_{\text{expt}}$	$\gamma(p)_{\text{theor}}$	$\gamma(p)_{\text{expt}}$
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

$$\gamma(p) = pT_c(1)/T_c(p). \quad (6)$$

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 $\text{LiTb}_{0.5}\text{Y}_{0.5}\text{F}_4$. In $\text{LiTb}_{0.3}\text{Y}_{0.7}\text{F}_4$, 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 $\text{LiTb}_{0.5}\text{Y}_{0.5}\text{F}_4$] and in this compound the agreement with the experimental results is not so good.

With this experimental investigation of the critical behavior of $\text{LiTb}_{0.3}\text{Y}_{0.7}\text{F}_4$, 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.

We would like to thank Dr. J. P. Renard for many helpful discussions and we are grateful to Dr. S. Williamson for fruitful comments on the manuscript.

¹E. Brézin and J. Zinn-Justin, Phys. Rev. B **13**, 251 (1976).

²P. Beauvillain, thèse Doctorat d'Etat No. 2142A, Université Paris-Sud, Orsay, France, 1979 (unpublished).

³L. M. Holmes, T. Johansson, and H. J. Guggenheim, Solid State Commun. **12**, 993 (1973).

⁴L. M. Holmes, J. Als-Nielsen, and H. J. Guggenheim, Phys. Rev. B **12**, 180 (1975).

⁵G. Ahlers, A. Kornblit, and H. J. Guggenheim, Phys. Rev. Lett. **34**, 1227 (1975).

⁶J. Als-Nielsen, Phys. Rev. Lett. **37**, 1161 (1976).

⁷P. Beauvillain, C. Chappert, and I. Laursen, J. Phys. C **13**, 1481 (1980).

⁸A. Aharony, Phys. Rev. B **13**, 2092 (1976).

⁹P. Beauvillain, C. Chappert, J.-P. Renard, and J. A. Griffin, J. Phys. C **13**, 395 (1980).

¹⁰R. Liebmann, B. Schaub, and H. G. Schuster, to be published.