Wave-Vector–Dependent Susceptibility at $T > T_c$ in a Dipolar Ising Ferromagnet

J. Als-Nielsen
Atomic Energy Commission Research Establishment Risø, Roskilde, Denmark

and

L. M. Holmes*
Laboratorium für Festkörperphysik, Eidgenossische Technische Hochschule, Zürich, Switzerland

and

H. J. Guggenheim
Bell Laboratories, Murray Hill, New Jersey 07974
(Received 28 January 1974)

The singular behavior of the susceptibility $\chi_T(\vec{Q})$ near wave vector $\vec{Q}=0$, which is characteristic of long-range dipolar interactions, has been observed in a neutron-scattering study of $LiTbF_4$. Quantitative agreement with mean-field theory is obtained at temperatures $2 < T/T_c < 10$. In a rather broad critical region, $0.01 < T/T_c - 1 < 1$, the divergent part of $\chi_T(\vec{Q})$ follows a power law $\chi_T(Q_x \to 0)/\chi_T{}^0 = C(1 - T_c/T)^{-\gamma}$ with $C = 1.25 \pm 0.13$ and $\gamma = 1.13 \pm 0.06$.

During the past decade it has become evident that critical phenomena in a wide variety of systems are universal in the sense that they only depend on the dimensionality of the system, d, and of the order-parameter space, n; e.g., $n=1$ for the Ising model, $n=3$ for the Heisenberg model, etc. Details about the interaction between the constituents are irrelevant for the critical behavior; it is only necessary to distinguish between systems with short-range and long-range interactions. The mean-field behavior in the transition to superconductivity is thus ascribed to the extreme long range of the interaction. Another interaction of long range is the Coulomb interaction between electric or magnetic dipoles. It is emphasized that this long-range interaction has *not* to be defined by a large number of phenomenological interaction parameters, but is merely given by the size of the dipole moments and the lattice in which they are located.

Uniaxial dipolar systems have recently been treated theoretically by several authors.[1-3] but relatively little is known from the experimental point of view. The dipolar interaction is of course important in ferroelectrics, but ferroelectric transitions are driven by the coupling to the lattice, and it is difficult to separate out the effects of lattice coupling and of dipolar interactions in such materials.[4] The other possibility is provided by magnets with exclusive magnetic dipolar interaction. Although this interaction in general is weak compared to, e.g., superexchange, it can be the dominant interaction in some rare-earth compounds, and the large magnetic moments ($\sim 10\mu_B$) imply Curie-Weiss temperatures θ of several kelvins. By examining in great detail the high-temperature behavior of the susceptibility and of the specific heat, Wolf, Meissner, and Catanese[5] showed that, e.g., the rare-earth hydroxides are systems exhibiting superexchange and dipolar interactions of comparable magnitude.

Holmes, Johansson, and Guggenheim[6] recently found that $LiTbF_4$ is an Ising ferromagnet with strong dipolar interactions. The crystal structure is tetragonal $I4_1/a$ and the Curie temperature $T_C = 2.86$ K. The excited crystal-field levels of Tb^{3+} $^7F_6$ are more than 150 K above the ground-state doublet[7,8] $|J_z = \pm 6\rangle$, and the transverse susceptibility is less than 10^{-3} of the longitudinal susceptibility at $T/\theta \simeq 1$, so the magnetic moments of Tb ions, $8.9\mu_B$, may be considered as an Ising system with an effective spin of $\frac{1}{2}$.

In this Letter we describe a neutron-scattering study of the wave-vector– and temperature-dependent susceptibility in $LiTbF_4$. The results display the $Q=0$ singularity characteristic of the dipolar Coulomb interaction and a quantitative comparison with theory shows that $LiTbF_4$ can be considered as a model system for the uniaxial dipolar ferromagnet.

The magnetic-scattering cross section when unpolarized neutrons are scattered from wave vector $\vec{k}_1$ to $\vec{k}_2$ depends on the wave-vector transfer $\vec{Q} = \vec{k}_1 - \vec{k}_2$:

$$\sigma(\vec{Q}) = \alpha[1 - (Q_z/Q)^2] f^2(Q) \chi_T(\vec{Q})/\chi_T{}^0. \quad (1)$$

Here Q_z is the component of $\vec{Q}$ along the spin direction, the z axis, and $f(Q)$ is the form factor of the Tb^{3+} ion.[9] $\chi_T(\vec{Q})$ is the *longitudinal* susceptibility and χ_T^0 is the susceptibility of the noninteracting system. The quasielastic approximation on which Eq. (1) is based was checked experimentally in $LiTbF_4$; no inelasticity in the scattering at different wave vectors and temperatures was observed within the resolution width of 0.1 meV. The data reported here were taken with a triple-axis spectrometer set for elastic scattering but with the resolution of the analyzer much coarser than 0.1 meV. With this setup transitions to the excited states of the Tb ions are not contributing to the recorded intensity, and the background count rate is low.

The susceptibility is at high temperatures evaluated using the mean-field approximation. In $LiTbF_4$ there are two inequivalent Tb^{3+} sites which we index as 1 and 2. Accordingly we distinguish between two generalized Curie-Weiss temperatures $\theta_{11}(\vec{Q})$ and $\theta_{21}(\vec{Q})$ that are the Fourier transforms of the intrasublattice and intersublattice interactions. We find

$$\chi_T^0/\chi_T(\vec{Q}) = 1 - \mathrm{Re}[\theta_{11}(\vec{Q}) + \theta_{21}(\vec{Q})]/T - (\mathrm{Im}[\theta_{21}(\vec{Q})]/T)^2\{1 + \mathrm{Re}[\theta_{21}(\vec{Q}) - \theta_{11}(\vec{Q})]/T\}^{-1}. \quad (2)$$

We define the deviation of $\vec{Q}$ from the reciprocal lattice vector $\vec{\tau}_{200}$ by $\vec{q} = \vec{Q} - \vec{\tau}_{200}$ and note that $\chi_T(\vec{q} + \vec{\tau}_{200}) = \chi_T(\vec{q})$. At long wavelengths ($q \simeq 0$) the susceptibility reduces to

$$\chi_T^0/\chi_T(\vec{q}) = 1 - \theta(\vec{q})/T, \quad (3)$$

with

$$\theta(\vec{q}) = -A_1(q_z/q)^2 + A_2 q_z^2 + A_3 + A_4 q^2. \quad (4)$$

The $q = 0$ singularity characteristic of the Coulomb interaction is noted. The dipolar contributions to A_i ($i = 1, \ldots, 4$) have been evaluated by Ewald's technique,[10] assuming $8.9\mu_B$ per Tb^{3+} ion,[6] with the results $A_1 = 8.403$ K, $A_2 = 11.811$ K Å^2, $A_3 = 3.925$ K, and $A_4 = 5.392$ K Å^2. The parameters $\theta_{11}(\vec{Q})$ and $\theta_{21}(\vec{Q})$ have also been calculated for general $\vec{Q}$ and will be reported in a subsequent publication.

We shall now compare Eqs. (1)–(4) with the experimental findings. In Fig. 1 we show the observed intensity along different symmetry directions at $T = 38.8$ K, i.e., a temperature that is high enough for Eq. (2) to be an accurate approximation. The scan along the (004) direction determines the background level $B = 350$ counts. The scan along (200) and from (200) towards (202) are thus compared with Eqs. (1) and (2) with one scale factor, $\alpha = 1864$ counts, as the only adjustable parameter. The variation of the intensity with $\vec{Q}$ is only partly due to the $\vec{Q}$ dependence of $\chi_T(\vec{Q})$, as illustrated by the dashed curves, where $\chi_T(\vec{Q})/\chi_T^0 = 1$ or $T \to \infty$. With these values of α and B we were able to determine the temperature dependence of $\chi_T(\vec{q})/\chi_T^0$ for $\vec{q}$ approaching zero. We found indeed that the extrapolated $q = 0$ value depends on whether the extrapolation is along the z axis or the x axis, in accordance with Eq. (4). Most strikingly, the temperature dependence of $\chi_T(q_z \to 0)$, as shown in Fig. 2, is completely different from that of $\chi_T(q_x \to 0)$. The latter exhibits normal ferromagnetic behavior, diverging at T_C and approaching the limiting Curie-Weiss law at high temperatures, $T_C/T < 0.5$. For $\chi_T(q_z \to 0)$ *no* divergence is observed at T_C, and the data may be fitted by a Curie-Weiss law with a *negative* intercept on the temperature axis. The two straight lines in Fig. 2 cross at unity, as expected, at infinite temperatures. For $\chi_T^0/\chi_T(q_x \to 0)$, the intercept with the abscissa, 0.77 ± 0.04, agrees

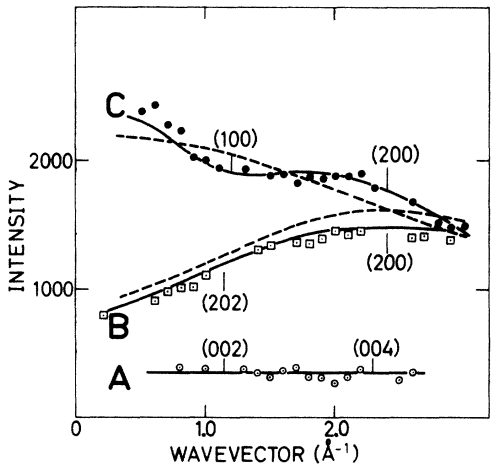

FIG. 1. Intensity along symmetry directions at $T/T_c = 13.5$. Note that the scattering due to nuclear Bragg peaks at (000), (200), etc. has been omitted. Scan *A* along (004) gives the background level. Scan *C* along (200) measures the product of the squared form factor and the susceptibility $\chi_T(\vec{Q})/\chi_T^0$. Scan *B* from (200) to (204) is, in addition, affected by the longitudinal susceptibility weight factor $1 - (Q_z/Q)^2$. The solid lines in *B* and *C* contain only an over-all scale factor as adjustable parameter, assuming $LiTbF_4$ to be a dipolar, Ising ferromagnet. The dashed lines correspond to infinite temperature or $\chi_T(\vec{Q})/\chi_T^0 = 1$ in Eq. (1).

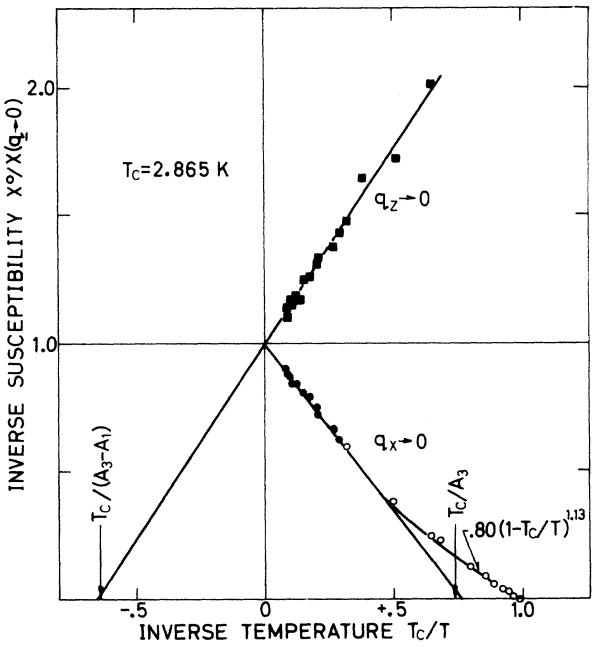

FIG. 2. Inverse susceptibility $\chi_T^0/\chi_T(\vec{q})$ versus the inverse temperature T_c/T when $\vec{q}$ approaches 0 along the z axis (the spin direction) or the x axis. The solid lines are a least-squares fit to the data points with filled signatures. The arrows mark the intercepts assuming $LiTbF_4$ to be a purely dipolar-coupled Ising ferromagnet.

with the value 0.73 calculated from Eq. (4) and also with bulk susceptibility data.[6] Also the intercept of $\chi_T^0/\chi_T(q_z \to 0)$, -0.65 ± 0.04, is in excellent agreement with the value -0.64 which we calculated by assuming dipolar interactions only.

We have therefore obtained agreement with both the temperature and the wave-vector dependence of $\chi_T(\vec{Q})$ by including only dipolar interactions. The temperature intercepts in Fig. 2 put an upper limit of 0.2 ± 0.2 K on the magnitude of the algebraic sum of the exchange interactions in the material. It might be argued that the individual exchange contributions could still be large, but accidentally cancel out of the sum, and in fact we cannot entirely rule out this possibility. It is possible to obtain a coarse upper bound of 1 K on the individual exchange integrals by including exchange in the fit to the data on Fig. 1. Indeed, it should be possible, in principle, to determine the exchange coupling with high precision from such large-Q scans and improved statistics. However, for the present we may conclude that there is *no* experimental evidence to suggest the presence of exchange coupling of magnitude $\gtrsim 0.2$ K.

The data for $\chi_T^0/\chi_T(q_x \to 0)$ at $T_C/T > 0.5$, shown as open circles in Fig. 2, give the critical behavior of the susceptibility. The data have been corrected for the instrumental $\vec{q}$ resolution by assuming the mean-field form of $\chi_T(\vec{q})$,

$$\chi_T^{-1}(\vec{q}) \propto \kappa_1^2(T) + q^2 + B_1(q_z/q)^2 - B_2 q_z^2,$$

using best-fit values of κ_1 at each temperature and values of B_1 and B_2 as determined from data at $T = T_C$. The resolution-corrected data have been fitted by a power law

$$\chi_T^0/\chi_T(q_x \to 0) = C^{-1}(1 - T_c/T)^{\gamma}$$

with the results $C = 1.25$ and $\gamma = 1.13$. At present we estimate that the systematic errors do not exceed 10% on C and 5% on γ. Mean-field theory predicts $\gamma = 1$ and $T_C = \theta(q_x \to 0)$, whereas recent theoretical work[1-3] suggests logarithmic corrections to mean-field behavior in dipolar-coupled systems. Our value of $T_C/\theta(q_x \to 0) = 0.73 \pm 0.02$ in $LiTbF_4$ may provide a crucial test of possible future calculations of this ratio. A detailed report on the spatial and temperature-dependent correlations in the critical region will be published later.

We are pleased to acknowledge discussions and correspondence with Professor R. A. Cowley and Professor M. E. Fisher.

*Work partly performed at Bell Laboratories and as a guest scientist at the Technical University and the Atomic Energy Commission Research Establishment Risø, Roskilde, Denmark.

[1]A. I. Larkin and D. E. Khmel'nitskii, Zh. Eksp. Teor. Fiz. 56, 2087 (1969) [Sov. Phys. JETP 29, 1123 (1969)].

[2]M. E. Lines, Phys. Rev. B 5, 3690 (1972).

[3]A. Aharony and M. E. Fisher, Phys. Rev. B 8, 3323 (1973); A. Aharony, *ibid.*, p. 3363.

[4]W. Cochran, Advan. Phys. 18, 157 (1969).

[5]W. P. Wolf, H. Meissner, and C. A. Catanese, J. Appl. Phys. 39, 1134 (1968); C. A. Catanese, A. J. Skjeltorp, H. E. Meissner, and W. P. Wolf, Phys. Rev. B 8, 4223 (1973).

[6]L. M. Holmes, T. Johansson, and H. J. Guggenheim, Solid State Commun. 12, 993 (1973).

[7]L. M. Holmes, H. J. Guggenheim, and J. Als-Nielsen, in Proceedings of the International Conference on Magnetism, Moscow, U.S.S.R., 1973 (to be published).

[8]The doublet is actually split into two singlets, but the splitting δ is negligibly small for our purposes. The crystal-field analysis in Ref. 7 gave $\delta < 2$ K, while recent ESR studies of 1% Tb^{3+} in $LiYF_4$ (I. Laursen, V. Frank, and L. M. Holmes, unpublished) gave $\delta = 1.34$

K, whence $2\theta/\delta \gtrsim 4$, taking $\theta = 3.6$ K from Ref. 6.
[9]G. H. Lander, T. O. Brun, J. P. Desclaux, and A. J. Freeman, Phys. Rev. B 8, 3237 (1973).
[10]See, e.g., M. Born and K. Huang, *Dynamical Theory of Crystal Lattices* (Oxford Univ. Press, Oxford, England, 1954), Sec. 30.

Effect of Stress on High-Field Magnetoresistance Anisotropy Due to Open Orbits in Iron*

M. A. Angadi,† E. Fawcett, and Mark Rasolt
Department of Physics, University of Toronto, Toronto, Ontario, Canada
(Received 26 December 1973)

The effect of uniaxial stress on the high-field magnetoresistance anisotropy of iron is measured and shown, by a combination of group theory and a band-structure interpolation scheme, to correspond to the origin of the ⟨100⟩ open orbits being magnetic breakdown between the hole octahedron and the electron jack on the minority-spin Fermi surface.

The magnetoresistance anisotropy curves for iron show deep minima due to open orbits in ⟨100⟩ and ⟨110⟩ directions.[1,2] Wakoh and Yamashita[3] proposed that the ⟨110⟩ open orbits occur along the ⟨110⟩ arms of a multiply connected hole sheet of the majority-spin Fermi surface which appears in their energy-band structure. Gold *et al.*[4] showed that a rigid exchange splitting of the energy bands of Wood[5] for paramagnetic iron results in the hole arms being pinched off as a result of hybridization with the electron surface centered on Γ. However, because of the magnetic breakdown the resultant surfaces (I and II in Fig. 1) are likely still to support open orbits along ⟨110⟩. Falicov and Ruvalds[6] showed that an accidental degeneracy, such as that at points i_1 and i_2, is allowed only when $\vec{B}$ is along a symmetry axis and the points are in a symmetry plane perpendicular to $\vec{B}$ as in Fig. 1. When $\vec{B}$ is tilted away from z in the (110) plane, the probability of breakdown at i_1 and therefore the number of open orbits should decrease as the degeneracy is progressively lifted by spin-orbit interaction. This is consistent with the observed angular dependence of the depth of the magnetoresistance minima associated with ⟨110⟩ open orbits.

In this Letter we are primarily concerned with the origin of the ⟨100⟩ open orbits. Wakoh and Yamashita[3] suggested that they result from breakdown between the minority-hole octahedron and electron jack. They further pointed out that their multiply connected majority-hole surface would support ⟨100⟩ (as well as ⟨110⟩) open orbits for $\vec{B}$ within 8° from a ⟨100⟩ axis, but these are unlikely to exist to any appreciable extent for the Fermi surface in Fig. 1.

Since uniaxial stress breaks the cubic symmetry, it will remove degeneracies in certain symmetry planes and directions, while leaving accidental degeneracies unresolved. We have therefore employed stress to determine whether

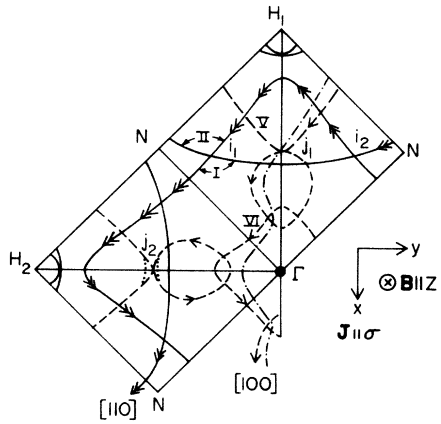

FIG. 1. Fermi surface of iron in (001) plane (after Gold *et al.*, Ref. 4) showing the splitting of levels at j_2 on the ΓH_2 line parallel to y resulting from uniaxial stress along x; solid line, majority surface; dashed line, minority surface; dash-dotted line, open orbit along ⟨100⟩ when $\vec{B}$ is rotated a few degrees away from z in the (100) plane; dashed arrows, open orbits along [100] resulting from the stress-induced splitting at j_2; solid arrows, open orbits along [110] resulting from breakdown between the majority large electron surface I and the hole arms II at i_1 and i_2. Spin-orbit splitting is not shown, though it is necessary to include spin-orbit interaction for breakdown to occur at j under zero stress, as explained in the text.