valuable help with the experiment and to Professor W. A. Little for suggesting the problem and for stimulation and guidance.

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EXISTENCE OF A CRITICAL LINE IN FERROMAGNETIC TO PARAMAGNETIC TRANSITIONS

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We show a possible connection between a theorem due to Griffiths and a conclusion of van der Hoeven, Teaney, and Moruzzi about the critical magnetic properties of EuS.

van der Hoeven, Teaney, and Moruzzi[1] have concluded that the critical point of the ferromagnetic material EuS is not a point, but a line on an H-T diagram. Such a result would follow if the ferromagnetic state were not one of uniform magnetization. Griffiths[2] has proved a theorem which has as one of its immediate consequences that in the absence of an externally applied magnetic field, the lowest energy state of a magnetic system is not a state of uniform magnetization. It is the purpose of this note to show a possible connection between Griffiths' theorem and the conclusion of van der Hoeven, Teaney, and Moruzzi.

In a ferromagnetic system, there will be a competition between dipole-dipole interactions and exchange interactions which tends to keep adjacent moments closely aligned while at the same time eliminating to a great extent divergences of the magnetization. When anisotropy is present, this leads to domains and domain walls. When anisotropy is not present, the minimum energy state is not known (except for a few special finite-geometry situations). But it can be argued that it is not one of uniform magnetization (except for certain finite dimensions).

Micromagnetics has shown that ellipsoidal ferromagnetic specimens above a critical size, while uniformly magnetized for applied field B_a greater than $4\pi DM$, where D is the demagnetizing factor and M is the spontaneous magnetization, cease to be uniformly magnetized as B_a becomes less than $4\pi DM$.[3] Thus, if the ellipsoidal dimensions are scaled beyond a critical size (which depends upon the dimension ratios), uniform magnetization will not occur for $B_a = 0$. This is in the absence of crystalline anisotropy.

The theorem of Griffiths includes the case of a system of spins interacting only with exchange interaction (isotropic and anisotropic) and dipole-dipole interaction. Let one choose a shape of the specimen, find the minimum free energy for that shape, and then compare the free energies for various shapes. In the limit as the dimensions go to infinity, the minimum free energies become equal. A corollary of this theorem is that the minimum-free-energy state cannot be one of uniform magnetization; for in that case, the dipole-dipole terms would be shape dependent. (This author does not wish to comment on what happens if the ellipsoid has an infinite dimensions ratio before passing to the limit of all dimensions going to infinity at constant dimensions ratio.) We assume from this that the minimum-energy configuration is not one of uniform magnetization and that it should depend upon sample shape and sample size. What this configuration would be even for a simple shape like a sphere is an unsolved problem in micromagnetics. Let us call this configuration the "spherical mode."

As one lowers the temperature of a spherical sample, one will reach a critical temperature at which this mode spontaneously appears. This temperature may depend upon the size of the sphere, but, according to Griffiths' theorem, in the limit of an infinite sphere, it will not depend upon the fact that it is a sphere or that the minimum-energy configuration is the "spherical mode."

If a sufficiently small field is applied, something very close to the "spherical mode" will appear spontaneously at a slightly lower temperature. That is, despite the fact that the small applied field creates a small uniform magnetization at all temperatures, there will still be a temperature at which the spherical mode appears. If the applied field is sufficiently large, the "spherical mode" may be suppressed and no critical temperature is observed; i.e., the uniform-magnetization mode continues to increase with decreasing temperature without the onset of any other configuration.

The situation is quite similar to the behavior of an antiferromagnetic material (wave vector $q \neq 0$) in a uniform magnetic field ($q=0$), or, for that matter, to a ferromagnetic material (wave vector $q=0$, ignoring dipole-dipole effects) in a spatially varying magnetic field ($q \neq 0$). As long as there appears a spontaneous mode with a spatial variation different from that of the field, there will be a critical temperature in that field.

As a specific example, we can calculate the Curie temperature of a thin toroid in the molecular-field approximation. In this case, the spontaneous mode is the curling pattern around the hole. If a field is applied perpendicular to the toroid (through the hole), the curling pattern appears at a temperature which decreases as the square of the applied field for small fields:

$$T_c = T_c(0)\left[1 - \frac{3}{5}\frac{H_A^{\,2}}{(2\pi M_0)^2}\right].$$

As long as $H_A < 2\pi M_0$, the curling mode will eventually appear below some temperature. The observation of any anomaly associated with this critical temperature should become more difficult as the field becomes larger. If the field were applied by passing current in a coil wrapped around the toroid (threaded through the hole), there would not be a critical point for any finite current.

While we have talked of the applied field, it is really the local field that matters. At any fixed temperature below $T_c(0)$ the magnetization in the curling mode decreases with the square of the applied field, while the component along the field increases linearly with field until it is equal to the spontaneous magnetization of the curling mode in zero applied field. The dipole-dipole interaction in the curling mode helps in the alignment of spins; in the uniform mode it hinders. The difference comes from the applied field increasing the "local" field. (In a uniformly magnetized sphere the "local" field is equal to the applied field. In the curling mode it is $+\frac{4}{3}\pi M$.) If the spontaneous mode itself produces a field which is not the same at all parts of a specimen, the most likely result would be a change of mode with temperature in the critical range and a "smearing out" of the critical-temperature anomalies. Thus, for anything except the very long rod or the toroid, it is not likely that the transition can be completely "sharp." In an applied field, any shape which departed significantly from an ellipsoid would necessarily have a smeared out transition.

To observe these effects, one would want a cubic material (anisotropy goes to zero as a high power of the magnetization) with a large dipole-dipole interaction and a low Curie temperature. Such a material is EuS. The Eu^{++} ion has a spin of $\frac{7}{2}$ and no orbital contribution. The Curie temperature is 16°K. To see the effects of this by bulk-magnetization measurements is quite difficult for one is then observing the $q=0$ mode and not the minimum-energy mode.

The measurement of specific heat may allow one to determine that a critical line exists. It has been the approach of van der Hoeven, Teaney, and Moruzzi to use as a criterion the appearance of a logarithmic singularity over several decades of $(T-T_c)/T_c$. They have interpreted their results as an indication that the critical point continues to exist in fields up to 1000 G and that the critical temperature decreases with the applied field. They find Eq. (1) with a power of 1.6 ± 0.4 rather than 2. For the model of the toroid discussed above, the anomalies should occur at the temperature that van der Hoeven, Teaney, and Moruzzi call T_M rather than at the T_H which comes from their curve fitting. Thus the connections between their result and the arguments given here are not quite clear, but in principle the effect noted here should exist.

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FAR-INFRARED ABSORPTION INDUCED BY ISOTOPES IN NaCl AND LiF†

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We have made measurements of the far-infrared absorption at 7°K in NaCl. The natural Cl^{35} and Cl^{37} isotopes acting as simple mass defects result in absorption in the acoustic band and yield information about phonon frequencies at critical points. The theory of this effect is derived and applied to NaCl. Similar measurements on LiF are also reported. We believe this to be the first observation of isotope-induced band-mode absorption in alkali halides.

In a large, perfect, harmonic, alkali-halide crystal infrared radiation is absorbed only by the transverse-optic phonon of essentially zero wave vector. This is the only mode of the crystal driven by the long-wavelength external radiation field. Since it is an eigenstate, there is a delta-function response at its frequency ω_0. When monovalent impurities are introduced substitutionally into the crystal, the radiation still drives only the $k \approx 0$, TO mode, but it is no longer an eigenstate. In addition to the delta function at ω_0, absorption appears in the band-mode region, and localized modes may sometimes appear.

The resulting in-band absorption in the acoustical region may be classified into two general types: (1) resonances due to strongly perturbing impurities[1-4] and (2) continuous absorption which reveals critical points in the phonon spectrum. We have given a preliminary report of such continuous spectra for NaCl doped with silver and other monovalent impurities, most of which were strongly coupled.[5] We were able to fit the data fairly well with theoretical calculations using shell-model phonons of Caldwell and Klein[6] after adjusting force-constant changes. However, the calculated peaks in the LA(X) and TA(L) critical-point region were about 6% too low; thus the shell-model parameters used probably gave the wrong phonon frequencies. Since the shape of absorption curves near critical points can be radically changed by strongly interacting impurities, a better test of the phonon model is obtained if the phonons are merely weakly perturbed. The best weak perturbation is provided by isotopes, since the perturbed dynamical matrix is then known exactly with no force-constant changes to guess or to fit.

For a pure harmonic crystal, the classical (and quantum mechanical) expression for the dielectric constant at angular frequency ω is[7]

$$\epsilon(\omega) = n_\infty^{\,2} + \frac{4\pi e_t^{*2}}{v\mu(\omega_0^{\,2} - \omega^2 - i\delta)}. \tag{1}$$

Here n_∞ is the high-frequency index of refraction; e_t^* is the macroscopic effective charge associated with the $q \approx 0$, TO mode; v the volume of a unit cell; and μ the reduced mass for the two atoms in the unit cell. An infinitesimal amount of damping is provided by $\delta = 0^+$.

With substitutional monovalent impurities we make the change

$$(\omega_0^{\,2} - \omega^2 - i\delta)^{-1} \to (0\,|\langle G(\omega^2 + i\delta)\rangle|\,0), \tag{2}$$

where $G(z) = (A - zI)^{-1}$ is the Green's function matrix for the entire impure crystal; A is the dynamic matrix; $|0)$ is a normalized eigenvector for the $k \approx 0$, TO phonon; and $\langle \cdots \rangle$ denotes a configurational average over impurity sites. G is given in terms of the Green's-function matrix for the perfect crystal:

$$G_0 = (A_0 - zI)^{-1},$$

and the defect matrix Γ, by the expression

$$G = G_0 - G_0 \Gamma G_0 + G_0 \Gamma G_0 \Gamma G_0 - \cdots,$$