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[1]E. Abrahams, P. W. Anderson, D. C. Licciardello, and T. V. Ramakrishnan, Phys. Rev. Lett. 42, 673 (1979), and references therein.
[2]B. L. Altshuler, A. G. Aronov, and P. A. Lee, Phys. Rev. Lett. 44, 1288 (1980); B. L. Altshuler, D. Khmelnitzkii, A. I. Larkin, and P. A. Lee, Phys. Rev. B 22, 5142 (1980).
[3]See M. N. Alexander and D. F. Holcomb, Rev. Mod. Phys. 40, 815 (1968).
[4]N. F. Mott, *The Metal-Insulator Transition* (Taylor and Francis, London, 1974).
[5]W. Sasaki, J. Phys. Soc. Jpn. Suppl. 49, 31 (1980).
[6]W. Sasaki, Philos. Mag. B 42, 725 (1981).
[7]Y. Ootuka, S. Kobayashi, S. Ikehata, W. Sasaki, and J. Kondo, Solid State Commun. 30, 169 (1979).
[8]A. Kawabata, Solid State Commun. 34, 431 (1980), and J. Phys. Soc. Jpn. 49, Suppl. A, 375 (1980).
[9]Equation (1) corrects the factor equal to the number of valleys in Kawabata's calculation (Ref. 8), which has been shown by H. Fukuyama, J. Phys. Soc. Jpn. 49, 649 (1980), to be absent if intervalley scattering rates exceed the inelastic collision rates. Kawabata also calculates that the prefactor can increase by from 15% to 35% in Si because of mass anisotropy, dependent upon the orientation of the crystallographic axis relative to $\vec{H}$ and $\vec{E}$. Changes in the prefactor of a similar magnitude could be expected as well due to anisotropy in relaxation rates (see, e.g., R. N. Bhatt and T. V. Ramakrishnan, to be published, for the corresponding two-dimensional calculation).
[10]B. L. Altshuler and A. G. Aronov, Solid State Commun. 38, 11 (1981).
[11]P. A. Lee and T. V. Ramakrishnan, to be published.
[12]T. F. Rosenbaum, K. Andres, G. A. Thomas, and P. A. Lee, Phys. Rev. Lett. 46, 568 (1981). The theoretical coefficient, α, in this paper is too large by a factor of 4. This error has been corrected here in Eq. (2) and in Ref. 11. The function F, which is $(1/x) \times \ln(1+x)$ for free electrons in the Thomas-Fermi approximation with a single valley, where $x = (2k_F/K)^2$, should be modified for Si:P. Depending on the ratio of kT and the intervalley scattering rate, F may be increased or decreased by a factor (up to 6) that is unknown because the intervalley rate is not accurately known.

First-Order Magnetic Phase Transitions in Fe

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The first observation of a first-order phase transition in the magnetization of Fe in an applied field is reported. The samples were epitaxially grown (110) Fe films on GaAs. Mean-field theory, with the inclusion of uniaxial anisotropy and the constraint that $\vec{M}$ lie in the plane of the film, gives excellent agreement with the observed transition. The relationship of this transition to the predictions of the three-state Potts model for cubic ferromagnets is discussed.

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Cubic ferromagnetic iron is predicted by mean-field theory to exhibit a first-order transition in magnetization as a function of applied field along a [111] crystallographic direction in a single crystal.[1,2] There is no evidence of this transition in the original magnetization measurements of Honda and Kaya[3] or of Williams[4] on single crystals of 3.8% Si in Fe. There is observed instead a rapid increase of slope as $H \rightarrow H_A$, the anisotropy field, with a discontinuity in the first derivative at $H = H_A$. More recently an attempt was made to observe the predicted transition in single-crystal [111] iron whiskers.[5] That study also failed to reveal any discontinuity, yielding results very similar to the earlier work.

Renewed interest in observing this transition has been stimulated by the identification of cubic ferromagnets as realizations of the three-state

Potts model[6] in three dimensions, as discussed by Mukamel, Fisher, and Domany (MFD).[2] After several years of controversy regarding the nature of the phase transition in the Potts model, a recent renormalization-group calculation[7] has shown unambiguously that the transition of the three-state model in three dimensions is first order, the same order predicted by mean-field theory.[8] The mean-field phase diagram for cubic ferromagnets calculated by MFD is reproduced in Fig. 1. This phase diagram neglects the anisotropic effects of demagnetizing fields which can be present in real samples. In properly prepared samples, the demagnetizing field can be employed as an effective tool to alter the phase diagram, expanding selected regions to make them more accessible to investigation. In this paper we present the results of such an investigation which reveals for the first time first-order phase transitions in Fe single crystals.

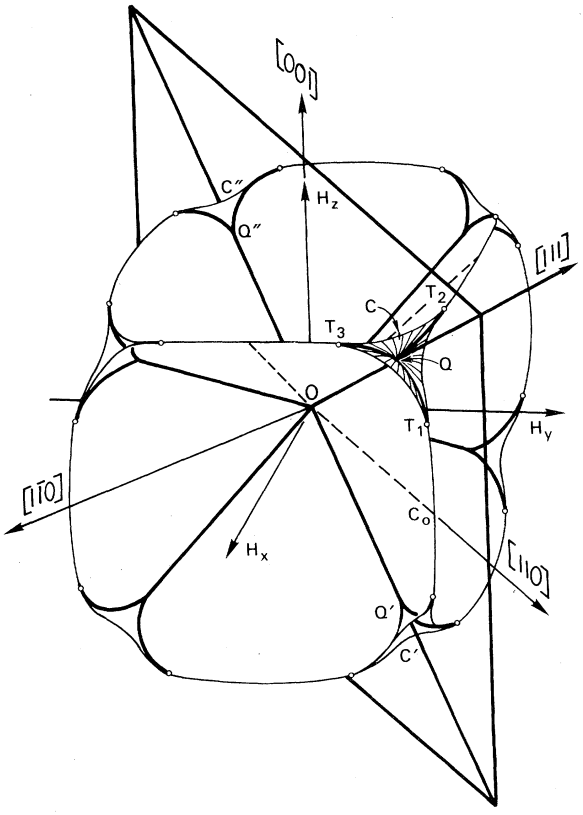

FIG. 1. Schematic mean-field phase diagram in H_x, H_y, H_z space of a cubic ferromagnet with three easy axes, from Mukamel, Fisher, and Domany, Ref. 2. Bold lines denote lines of magnetic triple points; thin curves represent critical lines of Ising character; C and C_0 are particular critical points; T_1, T_2, T_3 are tricritical points; and Q, Q', etc. are quadruple points. The (1$\bar{1}$0) plane of the film samples is superposed on the phase diagram.

The samples were prepared as thin single-crystal films by a procedure described elsewhere.[9] The film plane is a (1$\bar{1}$0) surface and therefore contains all of the principal magnetic axes of interest: [001], easy; [111], hard; and [110], intermediate. This is illustrated by Fig. 1 which shows a (1$\bar{1}$0) plane superimposed on MFD's phase diagram. The demagnetizing field (H_d) normal to this plane acts like a uniaxial anisotropy, distorting the phase diagram by stretching it out along the [1$\bar{1}$0] axis. The shaded surfaces near [111] are first-order phase boundaries where jumps in magnetization, M, occur as a function of field, H. These surfaces meet at tricritical points $T_{1,2,3}$ and the quadruple point Q. This region is small, with opening angles $COQ = 0.36°$ and $TOQ = 1.68°$. It is accessible if one applies a sufficient field nearly along [111] to overcome an offset from the origin of $1.466H_A$ created by the anisotropy which makes this a hard direction. The demagnetizing field opens up this phase space in such a way that the (T_2T_3Q) surface extends all the way to the [110] axis, intersecting it at a point P. Since the magnetization vector, $\vec{M}$, in this work was confined to the film plane, the information of interest is contained in the intersection of the phase diagrams with the (1$\bar{1}$0) plane, as shown in Fig. 2. In the undistorted phase diagram of MFD the first-order line O-Q along [111] branches out to C and T_1. The dashed line shows the results of the demagnetizing distortion: a first-order line which starts at a point P, $0.268H_A$ along the [110] axis, passes through Q on the [111] axis, and terminates at C of the original mean-field-theory diagram. For the demagnetizing fields appropriate for an iron film the distortion of the phase diagram becomes enormous. (Its original scale was determined by $H_A \approx 275$ Oe as compared with $H_d \approx 20\,000$ Oe.) It can be envisioned from Fig. 2 by simply constructing flat surfaces normal to the (1$\bar{1}$0) plane which intersect the plane to form the lines P-Q-C, P'-Q'-C', etc. One obtains in this way a flat first-order surface separating the $\pm H_z$ regions. This intersects two first-order dihedral surfaces in a line of triple points of which P is representative. The dihedral surfaces themselves terminate in a line of critical points. These dihedral surfaces are all that remain of the cubic anisotropy; they reflect the twofold symmetry of the (1$\bar{1}$0) plane as one rotates H around the direction H_d. (The cubic

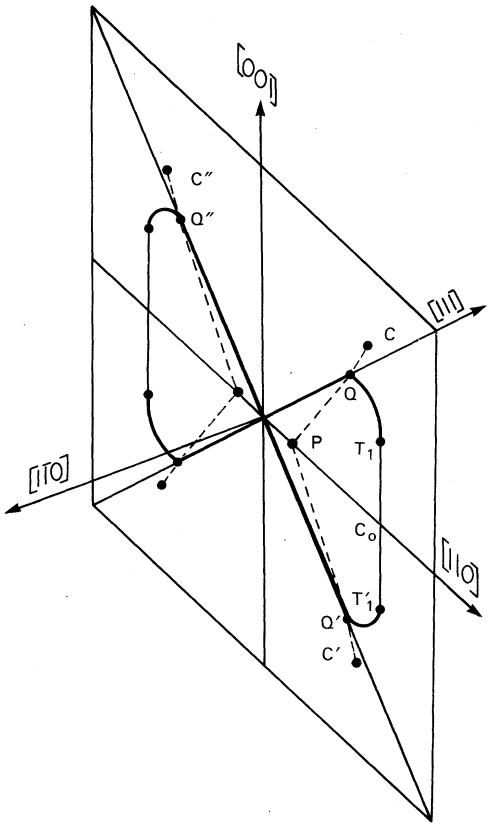

FIG. 2. Intersection of mean-field phase diagram and (1$\bar{1}$0) plane. Bold lines indicate first-order phase boundaries and thin lines indicate critical lines, for no demagnetizing field. The dashed line is the first-order phase boundary for the phase diagram modified by a demagnetizing field H_d normal to (1$\bar{1}$0) plane. P is a particular triple point where surface intersects [110] axis. The displacement of the C points away from the trigonal axes has been purposely exaggerated to correspond to the schematic distortion of Fig. 1. See line P-Q-C in Fig. 4(a) for an accurate representation.

anisotropy terms in the direction of H_d are completely suppressed by the large H_d.)

To calculate the phase line P-Q-C we consider the classic mean-field energy with fourth-order anisotropy constant only:

$$E = -\vec{H}\cdot\vec{M} - \tfrac{1}{2}K_4(M_x^4 + M_y^4 + M_z^4)/M_0^4.$$

The energy contribution of the demagnetizing field described above is assumed to be of the form $4\pi(M_x - M_y)^2$. If $K_4 \ll 4\pi M_0^2$ and H is to be applied only in the (1$\bar{1}$0) film plane, energy minimization will require that $M_x - M_y \approx 0$. Thus we can treat the effect of the demagnetizing field as a constraint which confines the moment to rotations in the plane of the film. The magnetic energy is

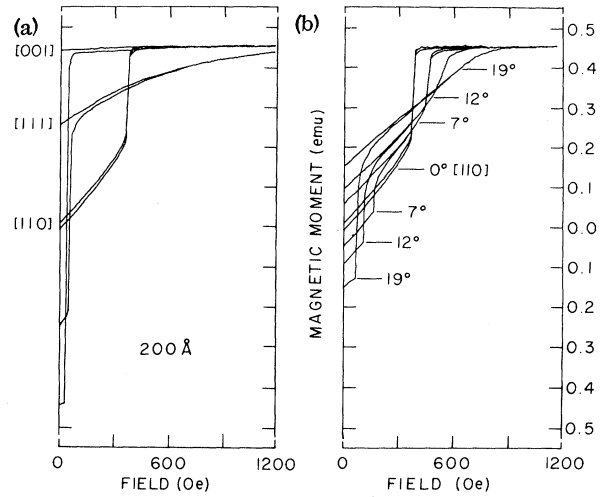

FIG. 3. (a) Magnetization of single-crystal Fe(1$\bar{1}$0) film for $\vec{H} \parallel [001]$, [111], and [110]. First-order transition occurs for $\vec{H} \parallel [110]$ at $H_c = 385 \pm 10$ Oe. (b) Magnetization of film for various angles of applied field from [110] in (1$\bar{1}$0) plane.

then

$$E = -\vec{H}\cdot\vec{M} - \tfrac{1}{2}K_4[M_z^4 + \tfrac{1}{8}(M_x + M_y)^4]/M_0^4.$$

Minimizing this energy gives the phase line P-Q-C of Fig. 2.

Magnetization measurements were carried out at room temperature on a vibrating-sample magnetometer in fields up to 1200 Oe, applied in the plane of the sample. Magnetization curves for $H \parallel [001]$, [111], and [110] are shown in Fig. 3(a). For $H \parallel [110]$ a discontinuity of 50% in M_s occurs at $H_C = 385 \pm 10$ Oe. Figure 3(b) shows M-H curves for the same sample as a function of the angle of H, θ_0, away from the [110]. The locations of first-order transitions were determined by numerically differentiating the M-H curves and locating peaks in the derivative functions. The first-order transition points are plotted as an experimental phase line, d, in Fig. 4(a). The phase line was terminated (critical point) at that field angle for which the derivative function exhibited only a finite step at the critical field. The experimental discontinuities in magnetization, ΔM, are shown in Fig. 4(b), line d. The theoretical magnitudes of the first-order discontinuities, ΔM, across the phase line are shown in polar coordinates in Fig. 4(b), line a, with use of the value $K_4 = 275$ Oe.[10] The first-order phase line and discontinuities for a second sample are shown in Figs. 4(a) and 4(b) as lines c.

Both samples exhibit the qualitative behavior predicted by mean-field theory—large first-

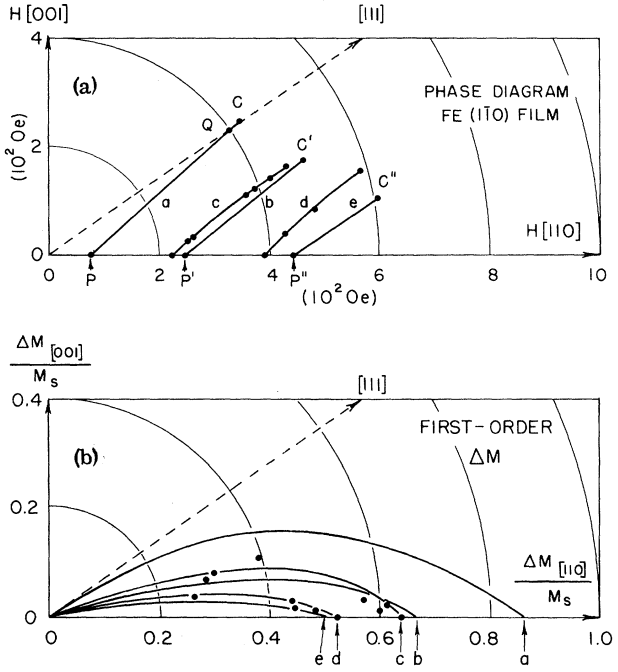

FIG. 4. (a) First-order phase boundaries in (1$\bar{1}$0) plane from mean-field theory with large demagnetizing field and *a*, no additional uniaxial anisotropy in plane; *b*, uniaxial anisotropy in plane, $K_2/K_4=0.5$; and *e*, $K_2/K_4=1.0$. C, C', C'' are critical points; P, P', P'' are triple points on [110] axis; Q denotes location of quadruple point from original mean-field-theory phase diagram. Experimental phase boundaries *c* and *d* for two samples with different growth-induced strains. (b) Relative discontinuities in magnetization at first-order transitions, as described in (a).

order jumps in magnetization when the field is oriented along [110], decreasing in magnitude and moving to larger field as the field is rotated away from the [110], the first-order transitions terminating at a critical point. However, neither sample is in quantitative agreement with the mean-field theory predictions, lines *a* in Figs. 4(a) and 4(b). The fact that both samples have higher critical fields along [110] than mean-field theory predicts indicates the presence of an additional anisotropy energy, which lowers the energy along [100]. The simplest possible anisotropy which can have this effect can be represented by a uniaxial term, $-K_2M_z^2/M_0^2$. Lines *b* in Figs. 4(a) and 4(b) show the predictions of mean-field theory with a uniaxial anisotropy $K_2/K_4=\frac{1}{2}$, and lines *e* for $K_2/K_4=1$.

These two samples were grown under slightly different conditions in order to determine the effect of crystal quality upon the magnetic properties. They were characterized by high-energy electron diffraction (HEED) and ferromagnetic resonance (FMR). The better crystal structure as determined by HEED gave the narrower FMR linewidth and required a smaller value for the uniaxial anisotropy to fit both the FMR and magnetization data. It is believed that the observed uniaxial anisotropy is the result of magnetostriction arising from growth-induced strain. There is a lattice mismatch of 1.35% between GaAs and the α-Fe lattice.

It is clear from these experimental results that mean-field theory provides a good description of magnetic phase transitions in a single-crystal (1$\bar{1}$0) Fe film. The most important result of this study, however, is the crucial role played by demagnetizing fields. In the (1$\bar{1}$0) film it enlarges the region over which the first-order transitions occur and increases the magnitude of discontinuities. It overcomes on one hand the difficulties of critical orientation which occur with spherical samples and, on the other hand, the difficulties of achieving small variations of the internal field in whiskers.

Shnidman and Domany[11] have proposed experiments on ferromagnetic films to study the crossover predictions of the Potts model—from first-order transitions in three dimensions to second-order transitions in two dimensions. We expect the choice of film geometry and resulting demagnetizing effects to be important in such experiments.

We are indebted to Amy Ryan for the care taken in the magnetization measurements, and we would like to thank Jim Cullen for several useful suggestions and comments.

[1]R. Gans and E. Czerlinski, Schriften Konigsberg. Gelehrten. Ges. Naturw. Kl. 9, S1 (1932).

[2]D. Mukamel, M. E. Fisher, and E. Domany, Phys. Rev. Lett. 37, 565 (1976).

[3]K. Honda and S. Kaya, Sci. Rep. Tohoku Univ. 15, 721 (1926).

[4]H. J. Williams, Phys. Rev. 52, 747 (1937).

[5]S. D. Hanham, B. Heinrich, and A. S. Arrott, J. Appl. Phys. 50, 2146 (1979).

[6]R. B. Potts, Proc. Cambridge Philos. Soc. 48, 106 (1952).

[7]B. Nienhuis, E. K. Riedel, and M. Schick, Phys. Rev. B 23, 6055 (1981).

[8]Experimental evidence for first-order transitions in three-state, three-dimensional Potts model systems is given by B. Barbara, M. F. Rossignol, and P. Bak, J. Phys. C 11, L183 (1978), for magnetic transitions in

$DyAl_2$, a cubic ferromagnet which differs from Fe in that it has very high fourth- and sixth-order single-ion anisotropies; and by A. Aharony, K. A. Müller, and W. Berlinger, Phys. Rev. Lett. 38, 33 (1977), for stressed $SrTiO_3$.

[9]G. A. Prinz and J. J. Krebs, Appl. Phys. Lett. 39, 397 (1981).

[10]H. Gengnagel and U. Hofmann, Phys. Status Solidi 29, 91 (1968).

[11]Y. Shnidman and E. Domany, to be published.

ERRATA

NEW INTERPRETATION OF THE SCALAR PRODUCT IN HILBERT SPACE. Y. Aharonov, D. Z. Albert, and C. K. Au [Phys. Rev. Lett. 47, 1029 (1981)].

The extreme right-hand side of the chain of equations in Eq. (4) should read $(2\pi)^{-1}|_{\psi}\langle\alpha,\beta|\varphi\rangle|^2$.

The right-hand side of Eq. (5) should read $\exp[i(\alpha\hat{x}+\beta\hat{p})]\,|\psi\rangle$.

In the paragraph where Eq. (7) is found, in the third line, the state $|x_1-x_2+\beta,\ p_1+p_2=\alpha\rangle$ should read $|x_1-x_2=\beta,\ p_1+p_2=\alpha\rangle$.

VELOCITY DEPENDENCE OF THE IONIZATION PROBABILITY OF SPUTTERED ATOMS. Ming L. Yu [Phys. Rev. Lett. 47, 1325 (1981)].

The sentence on page 1328, column 1, line 9 should read, "According to the authors of Ref. 6, it is conceivable that the extension of the theory to larger clusters may shift the region of strong velocity dependence toward lower escape velocities."

TIME-DEPENDENT VARIATIONAL PRINCIPLE FOR PREDICTING THE EXPECTATION VALUE OF AN OBSERVABLE. R. Balian and M. Vénéroni [Phys. Rev. Lett. 47, 1353 (1981)].

On page 1355, first column, the meaning of the sentence beginning at line 26 has been reversed by the insertion of a negation. Lines 27 and 28 should read, "*...the best choice for $\underline{D}(t)$ does depend in general on the observable A* to be measured at time t_1."

On page 1353, second column, line 16, $A(t)$ should read $\underline{A}(t)$.