Free Energy of Interacting Magnetic Dipoles*

ROBERT B. GRIFFITHS†
Physics Department, Carnegie-Mellon University, Pittsburgh, Pennsylvania 15213

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A system of spins on a lattice with magnetic dipole-dipole interactions, exchange, and anisotropy forces is shown to possess a well-defined bulk free energy, independent of sample shape, in the thermodynamic limit, provided there is no external magnetic field. The proof applies to classical or quantum spin systems with arbitrary g tensors, and is independent of any assumption of order or disorder in the magnetic phase, convergence of perturbation series, etc.

I. INTRODUCTION

ELEMENTARY magnetostatic considerations show that the free energy of a sample of magnetic material placed in an external magnetic field will depend on the sample's shape. The microscopic origin of this shape dependence is the long-range dipole-dipole interaction between the elementary magnetic moments. Such interactions are often neglected in theoretical calculations (typically, "exchange" energies are much larger than dipole-dipole terms), but they lead to important effects, such as domains, in real ferromagnets. In some materials,[1,2] the dipolar energy is comparable with or larger than the exchange terms and makes a non-negligible contribution in low-temperature phase transitions. In the absence of a magnetic field, one would expect the free energy to be shape-independent unless the sample has a net magnetization.

Calculations by Sauer[3] and by Luttinger and Tisza[4] indicated that the ground-state energy of a system of dipoles on a face- or body-centered cubic lattice is a minimum when the dipoles are ferromagnetically aligned along the axis of a thin, needle-shaped specimen. On the other hand, they concluded that the extra energy due to demagnetizing effects would favor an antiferromagnetic state in a spherical sample. Thus the ground-state energy should depend on the shape, even in the absence of an external field. However, Kittel[5] subsequently pointed out that the formation of domains (within which the dipoles are ferromagnetically aligned) would be expected in samples with finite demagnetizing factor, rather than a (local) antiferromagnetic arrangement. This explanation seems to agree with experiment for at least one dipolar ferromagnet.[2]

Hiley and Joyce,[6] Levy,[7] and Horner[8] have attacked the problem of systems with dipolar forces by means of perturbation series. They obtain the results expected on the basis of naive magnetostatics: The free energy is independent of shape in the absence of a magnetic field, and has the expected shape dependence in the presence of a field.

We think the arguments of Refs. 5–8 are in essence correct, though they stop somewhat short of rigorous proofs. Despite its numerous successes, domain theory still rests on rather insecure theoretical foundations.[9] And the difficulty with perturbation arguments is showing that the series converge. In fact, one would not expect the series to converge beyond a phase-transition point, and hence a different starting point for the perturbation series is needed, depending on the phase.

For these reasons, we wish to present a rigorous proof for the existence and shape independence of the bulk free energy for a lattice of interacting (permanent) dipoles in the absence of an external magnetic field. The argument works both at finite temperatures and at zero temperature for classical or quantum dipoles with arbitrary g tensors. It does not depend on whether the material is in an ordered or disordered phase. In addition, the Hamiltonian may include single-ion anisotropy and isotropic or anisotropic exchange (assumed to be of short range). Thus the argument applies to a majority of the lattice models used in discussions of ferromagnetism and antiferromagnetism. In particular, when applied to the systems considered by Sauer and Luttinger and Tisza, it shows that the energy they obtain for long, thin needles is an upper bound on the true energy for any sample shape.

An extensive (proportional to the number of particles) free energy for a macroscopic system presumably reflects the short-range character of the forces between the constituent atoms. Thus the free energy of a large system composed of smaller components should equal the sum of the free energies of the components, plus terms due to interactions between different components.

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† Alfred P. Sloan Research Fellow.

[1] W. P. Wolf, H. Meissner, and C. A. Catanese, J. Appl. Phys. **39**, 1134 (1968); M. Ball, M. J. Leask, W. P. Wolf, and A. F. G. Wyatt, *ibid.* **34**, 1104 (1963); W. P. Wolf, M. J. M. Leask, B. Mangum, and A. F. G. Wyatt, J. Phys. Soc. Japan **17**, Suppl. BI, 487 (1962).

[2] A. H. Cooke, D. T. Edmonds, C. B. P. Finn, and W. P. Wolf, J. Phys. Soc. Japan **17**, Suppl. BI, 481 (1962).

[3] J. A. Sauer, Phys. Rev. **57**, 142 (1940).

[4] J. M. Luttinger and L. Tisza, Phys. Rev. **70**, 954 (1946); **72**, 257 (1947).

[5] C. Kittel, Phys. Rev. **82**, 965 (1951).

[6] B. J. Hiley and G. S. Joyce, Proc. Phys. Soc. **85**, 493 (1965).

[7] P. M. Levy and D. P. Landau, J. Appl. Phys. **39**, 1128 (1968); P. M. Levy, Phys. Rev. **170**, 595 (1968).

[8] H. Horner, Phys. Rev. **172**, 535 (1968).

[9] S. Shtrikman and D. Treves, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press Inc., New York, 1963), Vol. III, p. 395.

The latter (when forces are of short range) should represent a small fraction of the bulk free energy, roughly proportional to the ratio of surface to volume for the components.

The foregoing idea is the intuitive basis for a number of proofs[10–13] that the usual procedures of equilibrium-statistical mechanics (canonical ensembles, etc.) when applied to a variety of models yield extensive free energies in the limit of large systems. In essence, the problem is to find bounds for the ratio of "surface" to "volume" free energies, and show that this ratio goes to zero for an infinite system. The proof usually proceeds through three (almost) independent steps:

(1) One shows that the energy E (strictly speaking, the free energy, but it is usually not difficult to extend the bound to finite temperatures) for a system of N particles is bounded *from below* by

$$E \geq -CN, \tag{1.1}$$

where C is a constant depending on the interaction potentials but independent of the number of particles.

(2) One shows that the free energy F of a system composed of two components with free energies F_1 and F_2 is bounded *from above* by

$$F \leq F_1 + F_2 + \Delta_{12}, \tag{1.2}$$

where Δ_{12} (a "surface energy") is suitably small in comparison with F for a large system.

(3) The results (1.1) and (1.2) are combined with geometrical arguments to establish the existence of a bulk free energy for systems of increasing size that are sufficiently regular in shape.

To date, the proofs have been unable to cope with two important potentials: Coulomb and dipolar interactions. In both cases, difficulties arise at step 2 because of the long-range character of the interactions. With pure Coulomb interactions, even step 1 is a nontrivial problem whose solution has only recently appeared.[14]

Step 1 for magnetic dipoles on a lattice, discussed in Sec. II, is not difficult, but a bit more subtle than the arguments that suffice for forces of short range. In the absence of a magnetic field, step 2 may be handled by an appeal to time-reversal invariance of the Hamiltonian, as discussed in Sec. III. This argument for step 2 is the only really original feature of our proof; the remainder, including step 3 (also discussed in Sec. III) follows a well-marked path. (The analog of time reversal for Coulomb systems is charge conjugation, but this is only useful in the unphysical situation where positive and negative particles have equal mass.) Some comments on how our proof applies to model calculations and measurements on real magnetic systems are found in Sec. IV.

II. LOWER BOUND FOR THE ENERGY

Consider a system of N classical dipoles, where the jth dipole located at the position $\mathbf{R}_j$ has a moment $\boldsymbol{\mu}_j$. If for $j \neq k$,

$$\boldsymbol{\rho}_{jk} = \mathbf{R}_j - \mathbf{R}_k, \tag{2.1}$$

and

$$A_{jk}{}^{\alpha\beta} = |\,\rho_{jk}\,|^{-3}[\delta_{\alpha\beta} - 3\rho_{jk}{}^{\alpha}\rho_{jk}{}^{\beta}/|\,\rho_{jk}\,|^{2}] \tag{2.2}$$

(we use Greek superscripts for Cartesian components), then the dipole-dipole interaction energy $\mathcal{H}_d$ has the form

$$\mathcal{H}_d = \tfrac{1}{2} \sum_{jk} \sum_{\alpha\beta} A_{jk}{}^{\alpha\beta} \mu_j{}^{\alpha} \mu_k{}^{\beta} \tag{2.3}$$

(where we assume that A_{jk} vanishes if $j=k$).

If $R>0$ is one-half the minimum distance between any pair of dipoles, we shall show that

$$\mathcal{H}_d \geq -\frac{1}{2R^3} \sum_j \sum_k (\mu_j{}^{\alpha})^2. \tag{2.4}$$

The inequality is established using the result that the energy of a configuration of magnetic "charges" (monopoles)[15] may be expressed as an integral of B^2 over all space, if $\mathbf{B}$ is the magnetic field arising from the "charges". Unfortunately, the integral diverges for point dipoles, but the interaction energy between dipoles is unchanged if each dipole is replaced by a sphere of radius R with an appropriate surface "charge" distribution. Of course, two such spheres must not overlap; this determines the choice for R.

The field $\mathbf{B}_j(\mathbf{r})$ due to the jth dipole may be written as

$$\mathbf{B}_j = -\nabla \psi_j, \tag{2.5}$$

$$\begin{aligned} \psi_j(\mathbf{r}) &= \boldsymbol{\mu}_j \cdot \mathbf{r}_j/(r_j)^3, && \text{for } r_j \geq R \\ &= \boldsymbol{\mu}_j \cdot \mathbf{r}_j/R^3, && \text{for } r_j \leq R \end{aligned} \tag{2.6}$$

where

$$\mathbf{r}_j = \mathbf{r} - \mathbf{R}_j. \tag{2.7}$$

The self-energy of the dipole is

$$E_j = \frac{1}{8\pi} \int d^3r\, (B_j)^2 = \frac{(\mu_j)^2}{2R^3}, \tag{2.8}$$

and the total energy of the magnetic charges is

$$\mathcal{H}_d + \sum_j E_j = \frac{1}{8\pi} \int d^3r\, \Big(\sum_j \mathbf{B}_j\Big)^2 \geq 0, \tag{2.9}$$

from which we obtain (2.4).

One can, of course, verify (2.9) directly without any appeal to magnetostatics by showing that

$$\sum_{\alpha\beta} A_{jk}{}^{\alpha\beta} \mu_j{}^{\alpha} \mu_k{}^{\beta} = \frac{1}{4\pi} \int d^3r\, (\mathbf{B}_j \cdot \mathbf{B}_k), \tag{2.10}$$

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[11] D. Ruelle, (a) Helv. Phys. Acta **36**, 183 (1963); (b) **36**, 789 (1963).
[12] R. B. Griffiths, J. Math. Phys. **5**, 1215 (1964).
[13] M. E. Fisher, Arch. Ration. Mech. Anal. **17**, 377 (1964).
[14] F. J. Dyson and A. Lenard, J. Math. Phys. **8**, 423 (1967); **9**, 698 (1968).

[15] Needless to say, we introduce magnetic monopoles for conceptual purposes only. See M. E. Fisher and D. Ruelle [J. Math. Phys. **7**, 260 (1966)] for an analogous argument.

where the integral is most easily evaluated by noting that ψ_j [as defined in (2.6)] satisfies Laplace's equation except at $|\mathbf{r}_j|=R$.

With m an abbreviation for the pair (j, α), and n for (k, β), we write (2.4) as

$$\tfrac{1}{2}\sum_m\sum_n A_{mn}\mu_m\mu_n \geq -\frac{1}{2R^3}\sum_m(\mu_m)^2. \quad (2.11)$$

Since A_{mn} [see (2.2)] is a real, symmetric matrix, its eigenvalues λ_p are real, and A may be diagonalized with the help of a real, orthogonal matrix V:

$$A_{mn}=\sum_p \lambda_p V_{mp}V_{np}. \quad (2.12)$$

Define

$$\nu_p=\sum_m \mu_m V_{mp} \quad (2.13)$$

and insert (2.12) in (2.11) to obtain

$$\sum_p\left(\lambda_p+\frac{1}{2R^3}\right)\nu_p^2\geq 0. \quad (2.14)$$

Since (2.11) holds when the μ_m are any set of real numbers, the same holds for ν_p in (2.14), from which it follows that for all p,

$$\lambda_p\geq -1/(2R^3). \quad (2.15)$$

With the help of (2.15), (2.4) may be extended to the case of quantum dipoles, for which the μ_j^α appearing in (2.3) must be replaced by appropriate Hermitian operators. A Hermitian operator $\mathfrak{A}$ is *positive*,[16] $\mathfrak{A}\geq 0$, if all its eigenvalues are non-negative or, equivalently, if the inner product $(\phi, \mathfrak{A}\phi)$ is non-negative for any ϕ; $\mathfrak{A}\geq\mathfrak{B}$ means that $\mathfrak{A}-\mathfrak{B}$ is positive. It is in this sense that we shall establish (2.4) as an operator inequality. Let the Hermitian operators ν_p be defined by (2.13). The square of such an operator is positive, and (2.15) implies that each summand on the left side of (2.14) is positive. Thus (2.14) holds as an operator inequality, and the same is true for (2.11) and (2.4).

For free electrons, the magnetic moment $\boldsymbol{\mu}$ (in units of the Bohr magneton) and spin $\mathbf{S}$ (in units of $\hbar$) operators are parallel and related by a g factor ($\simeq 2$), but in paramagnetic crystals $\mathbf{S}$ will in general be some "effective" spin operator, not in general parallel to $\boldsymbol{\mu}$. We shall assume that

$$\mu_j^\alpha=\sum_\beta g_j^{\alpha\beta}S_j^\beta, \quad (2.16)$$

where g_j is a real, symmetric matrix, and $|g_j|$ is the largest of the absolute values of its eigenvalues. By diagonalizing g and using the relation

$$\sum_\beta (S_j^\beta)^2=\hat{S}_j(\hat{S}_j+1) \quad (2.17)$$

(with $\hat{S}_j$ the total spin quantum number), one obtains

$$\sum_\alpha(\mu_j^\alpha)^2\leq|g_j|^2\hat{S}_j(\hat{S}_j+1) \quad (2.18)$$

as an operator inequality (where the right side is a multiple of the identity) in the quantum case or an ordinary inequality for a classical spin: a point on the surface of a sphere of radius $[\hat{S}(\hat{S}+1)]^{1/2}$. In either case,

$$\mathfrak{IC}_d\geq -\frac{1}{2R^3}\sum_j|g_j|^2\hat{S}_j(\hat{S}_j+1) \quad (2.19)$$

results from combining (2.18) and (2.4).

III. EXTENSIVE FREE ENERGY

A. Preliminaries

Consider an infinite crystal consisting of an aggregate of identical unit cells arranged on a regular lattice. Each unit cell contains a finite number of dipoles at arbitrary locations and with arbitrary choices of $\hat{S}_j$ and g_j. (Of course, the choice is only arbitrary for one unit cell, since the cells are all identical.) The "prototype" Hamiltonian for the infinite crystal is

$$\mathfrak{IC}=\mathfrak{IC}_c+\mathfrak{IC}_d \quad (3.1)$$

with $\mathfrak{IC}_d$ defined in (2.3) and

$$\mathfrak{IC}_c=\sum_i\sum_j J_{ij}^{\alpha\beta}S_i^\alpha S_j^\beta, \quad (3.2)$$

where the finite constants J_{ij} are assumed to vanish when spins i and j are further apart than some specified distance, and are chosen so that $\mathfrak{IC}_c$ has the translational symmetry of the lattice. Note that $i=j$ is permitted in (3.2), so that single-spin anisotropy is allowed as well as isotropic or anisotropic exchange.

A finite crystal Ω is a finite set of unit cells, $N(\Omega)$ in number, chosen from the infinite crystal. Its Hamiltonian $\mathfrak{IC}(\Omega)$ is defined by deleting from (3.1) all terms that involve one or more spin operators for spins not in Ω. The partition function Z and free energy $F(\Omega)$ are defined as usual:

$$Z(\Omega)=\exp[-F(\Omega)/kT]$$
$$=\mathrm{Tr}\{\exp[-\mathfrak{IC}(\Omega)/kT]\}, \quad (3.3)$$

where Tr stands for trace, a sum over the quantum states of the crystal Ω or the corresponding multiple integral for a classical system, and the temperature T is positive.

An upper bound for Z is obtained by replacing $\mathfrak{IC}(\Omega)$ by its lowest eigenvalue, or a lower bound for this quantity. A lower bound for $\mathfrak{IC}_c$ proportional to $N(\Omega)$ (and not otherwise dependent on Ω) is easily constructed using the methods of Ref. 12, while (2.19) gives a corresponding estimate for $\mathfrak{IC}_d$. The sum of these lower bounds is a lower bound for $\mathfrak{IC}(\Omega)$.

Since, in addition

$$\mathrm{Tr}[2]=\exp[CN(\Omega)], \quad (3.4)$$

[16] A discussion for finite-dimensional matrices (adequate for our purposes) will be found in P. R. Halmos, *Finite-Dimensional Vector Spaces* (D. Van Nostrand Co., Inc., Princeton, N.J., 1958), p. 139.

where C is a constant determined by the contents of a single unit cell, we conclude that $F(\Omega)$ has a lower bound

$$F(\Omega)\geq -DN(\Omega) \tag{3.5}$$

with D a function of temperature, etc., but independent of Ω.

B. Basic Inequality

Theorem: Let Ω be the union of two nonoverlapping (disjoint) "subcrystals" Ω_1 and Ω_2. Then

$$F(\Omega)\leq F(\Omega_1)+F(\Omega_2). \tag{3.6}$$

Proof: We may write

$$\mathcal{H}(\Omega)=\mathcal{H}(\Omega_1)+\mathcal{H}(\Omega_2)+\mathcal{H}_{12}, \tag{3.7}$$

where $\mathcal{H}_{12}$ includes all the terms in (3.2) and (2.3) that involve the product of two spin operators with one spin in Ω_1 and one in Ω_2. Let $\{\phi_j\}$ be a set of orthonormal eigenfunctions for $\mathcal{H}(\Omega_1)$ and $\{\epsilon_j\}$ the corresponding eigenvalues; for $\mathcal{H}(\Omega_2)$ let $\{\psi_k\}$ and $\{\eta_k\}$ be eigenfunctions and eigenvalues, respectively. The product states $\{\phi_j\psi_k\}$ form a complete orthonormal set for Ω, and may be used in evaluating the trace in (3.3):

$$\begin{aligned} Z(\Omega)&=\sum_{jk}(\phi_j\psi_k,\exp[-\mathcal{H}(\Omega)/kT]\phi_j\psi_k)\\ &\geq\sum_{jk}\exp[-(\phi_j\psi_k,\mathcal{H}(\Omega)\phi_j\psi_k)/kT], \end{aligned} \tag{3.8}$$

where the inequality is a consequence of "Peierls's theorem."[17] Using (3.7), we see that

$$(\phi_j\psi_k,\mathcal{H}(\Omega)\phi_j\psi_k)=\epsilon_j+\eta_k+\zeta_{jk}, \tag{3.9}$$

$$\zeta_{jk}=(\phi_j\psi_k,\mathcal{H}_{12}\phi_j\psi_k). \tag{3.10}$$

Let θ_1 be the time-reversal operator[18] for the crystal Ω_1; it is an antiunitary operator with the property that

$$\theta_1 S_i^{\alpha}\theta_1^{-1}=-S_i^{\alpha}, \tag{3.11}$$

if i denotes a spin in crystal Ω_1. If, in particular, we choose basis states so that the matrix elements of S_i^y are imaginary, an explicit form for θ_1 is

$$\theta_1=[\prod_{j\in\Omega_1}\exp(i\pi S_j^y)]\mathcal{C}, \tag{3.12}$$

where $\mathcal{C}$ is the (antilinear) operator that takes the complex conjugate of numbers appearing to its right.

Since $\mathcal{H}(\Omega_1)$ contains only terms quadratic in the spin operators of Ω_1, it is invariant under, that is it commutes with, θ_1. Hence

$$\hat{\phi}_j=\theta_1\phi_j \tag{3.13}$$

is an eigenfunction of $\mathcal{H}(\Omega_1)$ with eigenvalue ϵ_j, and since θ_1 is antiunitary, the $\{\hat{\phi}_j\}$ form a complete orthonormal set for Ω_1. Indeed, the inequality (3.8) still holds if ϕ_j is everywhere replaced by $\hat{\phi}_j$. However, $\hat{\zeta}_{jk}$, defined using (3.10) and replacing ϕ_j by $\hat{\phi}_j$, is equal to $-\zeta_{jk}$, because $\mathcal{H}_{12}$, unlike $\mathcal{H}(\Omega_1)$, contains only terms linear in the spin operators of Ω_1, and these change sign under θ_1. Thus if we add to (3.8) the inequality obtained by replacing ϕ_j by $\hat{\phi}_j$ and divide by two, the result is

$$\begin{aligned} Z(\Omega)&\geq\sum_{jk}\{\exp[-(\epsilon_j+\eta_k)/kT]\}\cosh(\zeta_{jk}/kT)\\ &\geq\sum_{jk}\exp[-(\epsilon_j+\eta_k)/kT]=Z(\Omega_1)Z(\Omega_2), \end{aligned} \tag{3.14}$$

and (3.6) is an immediate consequence.

The above proof was carried out for finite positive temperatures. However, it also applies in the limit $T\to 0$, since it is obvious that for any finite system the free energy goes continuously to the ground-state energy as $T\to 0$. The case of negative temperatures is best handled by reversing the sign of $\mathcal{H}$ in (3.1). We require in place of (2.4) an upper bound to $\mathcal{H}_d$, which can be obtained by replacing the magnetic "charge" on the spheres of radius R by a dipole (or current) distribution. (The reader may work out the details if he is interested.)

The analogous proof for classical spins should be obvious: θ_1 turns $\mathbf{S}_j$ into $-\mathbf{S}_j$ for every spin in Ω_1 and is thus a volume-preserving map of the phase space onto itself which reverses the sign of $\mathcal{H}_{12}$. The Peierls's theorem inequality is, of course, unnecessary.

C. Geometrical Arguments

Procedures for proving existence and shape independence of the bulk free energy given the basic inequality (3.6) have been worked out in great detail by Fisher,[13] so our discussion will be confined to a skeleton outline. For convenience of discussion, we assume a simple cubic lattice; the extension to other cases is obvious. Define the free energy per cell

$$f(\Omega)=F(\Omega)/N(\Omega), \tag{3.15}$$

and when Ω is a cube measuring 2^k unit cells on a side, denote $f(\Omega)$ by f_k. Since we can always put eight identical cubes together to form a larger cube, (3.6) implies in particular that the sequence $f_0, f_1, f_2, \cdots$ is monotone decreasing, and (3.5) that it has a lower bound. Therefore, the sequence possesses a limit f, which we call the bulk free energy.

While f was obtained as a limit for a special sequence of cubes, in fact it is also the limit for an arbitrary sequence of cubes of increasing volume. Suppose that Ω_m is a cube m unit cells on a side. If k is the largest integer such that $2^k\leq m$, we may think of Ω_m as composed of one cube with edge 2^k occupying one corner; either 0 or 19 cubes with edge 2^{k-1} placed next to the cube of edge 2^k, etc., so that Ω_m is composed of smaller cubes whose free energies belong to the sequence $\{f_k\}$. For large m, a negligible fraction of the volume is

[17] R. Peierls, Phys. Rev. **54**, 918 (1938); see also Refs. 11(b) and 12.

[18] E. P. Wigner, *Group Theory* (Academic Press Inc., New York, 1959), Chap. 26.

occupied by cubes with small edges, so that in the limit $m\to\infty$, (3.6) implies that the free energy $f(\Omega_m)$ is bounded from above by f. To put a lower bound on $f(\Omega_m)$, place Ω_m inside a cube of edge 2^{k+1} and fill up the remaining space in the large cube with cubes of edge 2^{k-1}, 2^{k-2}, etc., preference being given to the largest cubes. As $m\to\infty$, one finds that $f(\Omega_m)$ is bounded from below by f. Analogous arguments can be carried out for other sufficiently "regular" shapes—spheres,[19] ellipsoids, etc.—see Ref. 13.

As one would expect intuitively, f is obtained only if all linear dimensions of the crystal increase to infinity. However, it does not depend on the order in which they become infinite. Thus, for example, let $f(L, L, P)$ be the free energy per cell for a rectangular block measuring $L\times L\times P$ unit cells. Then if $P=nL$, with n an integer, (3.6) implies that

$$f(P, P, P)\leq f(L, L, P)\leq f(L, L, L). \quad (3.16)$$

In particular, one can show that

$$f_L=\lim_{P\to\infty} f(L, L, P) \quad (3.17)$$

is well defined, and then (3.16) tells us that $f_L\geq f$ and

$$\lim_{L\to\infty} f_L=f. \quad (3.18)$$

IV. DISCUSSION

In Sec. III, we have established rigorously that for a large class of lattice models that include magnetic dipolar interaction, the bulk free energy exists and is independent of sample shape in the thermodynamic or infinite-volume limit, in the absence of an external magnetic field. These results have implications both for model calculations and (we believe) for real systems.

Consider first the work of Luttinger and Tisza.[4] The energy they calculated for a ferromagnetic configuration in a long, thin needle in the fcc (or bcc) lattice is an upper bound for the true ground-state energy for this sample shape. But from our results it must also be an upper bound for any other sample shape. This completely rules out the possibility, at $T=0°$, of antiferromagnetism of the type they considered.

Our arguments also seem to imply, though we have not constructed a rigorous proof, that a nonzero magnetization (per unit cell) for, say, a spherical sample always leads to an increase in the bulk free energy. This is in contrast to the simple Ising or Heisenberg ferromagnet without dipolar interactions in which the existence of "spontaneous magnetization" means that the magnetization may take on any value from zero up to its "spontaneous" value without a change in the bulk free energy.[20]

These two requirements of an energy corresponding to ferromagnetism while at the same time zero net magnetization suggest, of course, that there is a local ferromagnetic alignment, but the direction of this alignment is different in different parts of the crystal. A domain structure, as Kittel has proposed, would be appropriate given sufficient anisotropy. In systems where anisotropy is small—e.g., if the Hamiltonian is dominated by isotropic exchange (a situation to which our arguments are equally applicable)—some other pattern of nonuniform magnetization might be expected.

In considering the situation in real magnetic systems, one must keep in mind the limitations and simplifications involved in a model of spins located on a lattice. This is true even for insulators, in which a "localized electron" approximation should be better than in metals. In addition, we have considered the free energy corresponding to true thermodynamic equilibrium; and this is, of course, not a good approximation in materials where prominent hysteresis effects indicate the presence of metastable states. A valid criticism of our proof is that we give no estimate for the minimum crystal size for which the free energy will provide a good approximation to the bulk value. Hysteresis and size effects are, of course, subject to experimental investigation: It is possible to test the latter, for example, by measuring heat capacities on samples of various sizes and shapes. Since experiments are always evaluated in terms of (necessarily) oversimplified models, it is of value to know precisely what features of the experimental results are inherent in the model, and which are due to terms that have been left out. We hope our arguments may have made a modest contribution to this end.

One would also like a proof that the free energy for a system of dipoles in the presence of a magnetic field has the *expected* shape dependence predicted by magnetostatic arguments. We hope that arguments may soon be forthcoming, but it is clear that a more sophisticated approach is required than that of Sec. III.

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[19] One may define a spherical sample of radius R as the aggregate of unit cells whose centers lie within a distance R from the center of a particular cell, and analogous definitions work for ellipsoids, etc. The precise form of surface, if not too pathological, is unimportant in the infinite-volume limit. See Ref. 13.

[20] R. B. Griffiths, Phys. Rev. **152**, 240 (1966). In this reference the symbol a is used for the relevant "free energy."