

Lattice sums for dipolar systems

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A general method for the evaluation of lattice sums determining the effective parameters in the Hamiltonian of a dipolar magnetic system is described. The anisotropy of the Hamiltonian is examined for crystal structures of tetragonal and hexagonal type. The results are of particular relevance in systems where exchange and any other nondipolar interactions are isotropic. Applications to gadolinium are considered.

I. INTRODUCTION

Dipolar interactions in magnetic materials may lead to critical behavior significantly different from that found in materials with short-range interactions only.^{1,2} This difference is particularly marked in three-dimensional systems with a uniaxial order parameter where dipolar interactions lead to a critical behavior predicted by classical (mean-field) theory with logarithmic corrections.³

For a lattice with cubic symmetry, the quadratic term which appears in the magnetic Hamiltonian

$$H = \int dq \sum_{\alpha, \beta}^n A^{\alpha, \beta}(q) \phi^\alpha(q) \phi^\beta(-q) + \text{quartic terms} \quad (1)$$

is expected to be of the form¹

$$A^{\alpha\beta} = -(\alpha_3 + \alpha_4 q^2 - \alpha_5 q_\alpha^2) \delta_{\alpha\beta} + \alpha_1 \frac{q_\alpha q_\beta}{q^2} - \alpha_2 q_\alpha q_\beta. \quad (2)$$

Evaluation of the effective coefficients appearing in (2) requires the performance of a lattice sum (Sec. II). These sums have been performed for lattices with cubic symmetry^{1,4} and provide a useful check in the calculations described below.

Here we shall be concerned with anisotropies in the effective coefficients which arises when deviations from the high-symmetry situations of cubic symmetry in tetragonal lattices and ideal hard-sphere packing in hexagonal-closed-packed (hcp) lattices occur.

Our interest in this problem for hcp systems was, to some extent, motivated by experimental studies of gadolinium. Gadolinium is well known to order ferromagnetically with its net magnetic moment aligned with the *c* axis; however, its universality class has not been conclusively established. For example, measurements of the static magnetization exponent β by Chowdhury *et al.*⁵ led them to conclude that Gd is in the universality class of Heisenberg model with short-range interactions for reduced temperatures $t > 10^{-3}$. (However, measurements of the autocorrelation function dynamical exponent by the same authors were not consistent with either the Ising or Heisenberg short-range interaction universality classes.⁶) On the other hand, analysis of resistivity data from a single run with $|t| < 10^{-3}$ found that the data could not be fitted by a power law of the

type expected if only short-range interactions determined the critical behavior, but was consistent with logarithmic corrections to the regular term of the sort expected for a uniaxial dipolar system.⁷

In Sec. II a general method is described for the efficient evaluation of lattice sums such as those required to evaluate the dipolar contribution to the effective magnetic Hamiltonian. Although we shall describe the method with special emphasis on the type of sum required for dipolar magnetic materials, it is quite general and readily adapted to other problems. As with previous computations for the lattices with cubic symmetry,^{1,4,8} the method employs the properties of the Θ functions under the Jacobi imaginary transformation⁹ to convert the lattice sum to a rapidly convergent form. Section III is a detailed account of the calculation of the anisotropies in tetragonal and hcp lattices, and Sec. IV summarizes these results and the implications for magnetic systems, such as Gd, which are considered to be described by isotropic exchange interactions plus dipole-dipole interactions.

II. GENERAL LATTICE SUMS

Consider the sum over the orthorhombic lattice vectors \mathbf{L}

$$B(\mathbf{q}, \mathbf{x}) = \sum_{\mathbf{L}}' \frac{e^{i\mathbf{q} \cdot \mathbf{L}}}{|\mathbf{x} - \mathbf{L}|^s}. \quad (3)$$

Such a sum arises in the study of dipolar magnetic systems, as the dipole-dipole contribution to the magnetic Hamiltonian of a *d*-dimensional system is (cf. Ref. 1)

$$E_{\text{dd}} = \frac{(g\mu_B)^2}{2} \sum_{l, l'}' \frac{\partial^2}{\partial \mathbf{R}_l^\alpha \partial \mathbf{R}_{l'}^\beta} \frac{1}{|\mathbf{R}_l - \mathbf{R}_{l'}|^{d-2}} S_l^\alpha S_{l'}^\beta, \quad (4)$$

where *l* and *l'* label lattice sites, α and β denote components of the *d*-dimensional system, S_l is the spin at site *l*, and the prime on the sum excludes the terms with $l = l'$. Taking the Fourier lattice transform

$$S_l^\alpha = \sum_{\mathbf{q}} \frac{e^{i\mathbf{q} \cdot \mathbf{R}_l}}{\sqrt{V}} \sigma_{\mathbf{q}}^\alpha \quad (5)$$

and writing $\mathbf{L} = \mathbf{R}_j - \mathbf{R}_{j'}$, we obtain

$$E_{dd} = \frac{(g\mu_B)^2}{2} \frac{N}{V} \sum_{\mathbf{q}} \sigma_{-\mathbf{q}}^\alpha \sigma_{\mathbf{q}}^\beta A^{\alpha\beta}(\mathbf{q}), \quad (6)$$

where

$$B(\mathbf{q}, \mathbf{x}) = \sum_{\mathbf{L}}' \frac{e^{i\mathbf{q}\cdot\mathbf{L}}}{\Gamma(s/2)} \int_0^\infty dy y^{-1+s/2} e^{-(\mathbf{x}-\mathbf{L})^2 y} \quad (8)$$

$$= \frac{1}{\Gamma(s/2)} \int_0^\infty dy y^{-1+s/2} e^{-x^2 y} \left[-1 + \prod_{j=1}^d \Theta \left[\frac{iq_j + 2x_j y}{2\pi/a_j}; \frac{a_j^2 y}{\pi} \right] \right], \quad (9)$$

where

$$\Theta(z; X) = \sum_{l=-\infty}^{\infty} e^{2\pi l z} e^{-\pi l^2 X} \quad (10)$$

is the Jacobi Θ function of the third kind.⁹ a_j in (9) denotes the lattice spacing along the j th axis.

The integral in (9) may be divided at a point y_0 , which is to be chosen to give good convergence of the sums in both parts of the integral. We write

$$B(\mathbf{q}, \mathbf{x}) = B_1(\mathbf{q}, \mathbf{x}) + B_2(\mathbf{q}, \mathbf{x}) + B_3(\mathbf{q}, \mathbf{x}),$$

with

$$\Gamma(s/2)B_1(\mathbf{q}, \mathbf{x}) = \int_0^{y_0} dy y^{-1+s/2} e^{-x^2 y} \left[-1 + \frac{\pi^{d/2} y^{-d/2}}{\Omega_a} e^{-(\mathbf{q}/2 - i\mathbf{x}y)^2/y} \right], \quad (11)$$

where Ω_a is the atomic volume. We note that $B_1(\mathbf{q}, \mathbf{x})$ contains all of the singular small- \mathbf{q} behavior and in the limit $\mathbf{x} \rightarrow \mathbf{0}$ can be written

$$B_1(\mathbf{q}, \mathbf{x}=\mathbf{0}) = -\frac{y_0^{s/2}}{\Gamma(1+s/2)} + \frac{Cd}{q^2 \Omega_a} - \frac{Cd}{4\Omega_a} \int_0^{1/y_0} d\omega e^{-\omega q^2/4}, \quad (12)$$

$$\Gamma(s/2)B_2(\mathbf{q}, \mathbf{x}) = \int_0^{y_0} dy y^{-1+s/2} e^{-x^2 y} \frac{\pi^{d/2} y^{-d/2}}{\Omega_a} e^{-(\mathbf{q}/2 - i\mathbf{x}y)^2/y} \left[\prod_{j=1}^d \Theta \left[\frac{q_j}{2a_j y} - \frac{i x_j}{a_j}; \frac{\pi}{a_j^2 y} \right] - 1 \right]. \quad (13)$$

Here the Jacobi imaginary transform⁹

$$\Theta(z; X) = \frac{e^{\pi z^2/X}}{\sqrt{X}} \Theta \left[\frac{z}{iX}; \frac{1}{X} \right] \quad (14)$$

has been used to obtain a form for $B_2(\mathbf{q}, \mathbf{x})$ which will be a rapidly convergent sum when the Θ function is evaluated term by term.

Finally, the remaining contribution, from the upper range of the integration over y , is

$$\Gamma(s/2)B_3(\mathbf{q}, \mathbf{x}) = \int_{y_0}^\infty dy y^{-1+s/2} e^{-x^2 y} \left[-1 + \prod_j \Theta \left[\frac{iq_j a_j}{2\pi} + \frac{x_j a_j y}{\pi}; \frac{a_j^2 y}{\pi} \right] \right]. \quad (15)$$

$A^{\alpha\beta}(\mathbf{q})$ can then be evaluated by differentiating (11), (13), and (15) and taking the limit $\mathbf{x} \rightarrow \mathbf{0}$. The required sums, arising from those in (13) and (15), are rapidly convergent if an appropriate choice of y_0 is made and may readily be evaluated numerically.

The method is easily generalized to other lattices by decomposing the required lattice under consideration into a set of orthorhombic sublattices¹⁰ such that each site is in exactly one sublattice. For each such sublattice γ , there is then a sum

$$B_\gamma(\mathbf{q}, \mathbf{x}') = e^{i\mathbf{q}\cdot\rho_\gamma} \sum_{\mathbf{L}}' \frac{e^{i\mathbf{q}\cdot\mathbf{L}}}{|\mathbf{x}' - \mathbf{L}|^s}. \quad (16)$$

$$A^{\alpha\beta}(\mathbf{q}) = -\lim_{\mathbf{x} \rightarrow \mathbf{0}} \frac{\partial^2}{\partial x^\alpha \partial x^\beta} \sum_{\mathbf{L}}' \frac{e^{i\mathbf{q}\cdot\mathbf{L}}}{|\mathbf{x} - \mathbf{L}|^{d-2}}, \quad (7)$$

and all sums over \mathbf{q} are restricted to the first Brillouin zone.

Returning to the general form of the sum (3), we write

Here ρ_γ is a vector from the origin (chosen to be a site in one sublattice) to a neighboring atom in the sublattice ($\rho_{\gamma=0} = \mathbf{0}$). The contribution of each sublattice to $A^{\alpha\beta}(\mathbf{q})$ is then obtained by taking the appropriate derivatives of (16) with respect to \mathbf{x}' and evaluating these in the limit $\mathbf{x}' \rightarrow -\rho_\gamma$. The treatment of (16) to yield sums which are rapidly convergent follows that described above for the case $\rho = \mathbf{0}$. However, here, the prime on the summation excludes the term with $\mathbf{L} = \mathbf{0}$ only if $\rho_\gamma = \mathbf{0}$.

III. RESULTS FOR DIPOLAR SYSTEMS

In order to consider systems in which the dipolar interactions give rise to a spin anisotropy, we generalize (2) to

$$A^{\alpha\beta} = -[\alpha_3^\alpha + \alpha_4^\alpha q^2 - \alpha_z q_z^2 (1 - \delta_{\alpha,z}) - \alpha_5^\alpha q_\alpha^2] \delta_{\alpha\beta} + \alpha_1^{\alpha\beta} \frac{q_\alpha q_\beta}{q^2} - \alpha_2^{\alpha\beta} q_\alpha q_\beta. \quad (17)$$

The coefficients in (17) may then be evaluated from the small- q expansion of (16) using the technique of Sec. II with $s=d-2$, by inserting the appropriate lattice parameters a_i and summing over the required vectors ρ_γ .

Of principal interest here is the anisotropic situation $\alpha_3^c > \alpha_3^a$ ($\alpha_3^c < \alpha_3^a$), in which the dipolar interactions favor either the c axis or basal plane as the easy direction of magnetization.³ Figures 1–4 illustrate the behavior of α_3^a and α_3^c as functions of the ratio c/a of the unit-cell axes in a region where the crossover from the c axis to the basal plane as the easy direction of magnetization occurs.

As an illustrative example, we consider the tetragonal

$$A_1^i(\mathbf{q}=\mathbf{0}) = -\frac{2}{\Gamma(s/2)} \int_0^{y_0} dy y^{s/2}, \quad (18)$$

$$A_2^i(\mathbf{q}=\mathbf{0}) = \frac{\pi^{d/2}}{\Omega_a \Gamma(s/2)} \int_{\omega_0}^{\infty} d\omega \left[\frac{2\pi a}{a_i} \right]^2 \left[\sum_{l=-\infty}^{\infty} l^2 e^{-\pi^2 l^2 \omega (a/a_i)^2} \right] \prod_{j \neq i}^3 \Theta \left[0; \pi \omega \left[\frac{a}{a_j} \right]^2 \right], \quad (19)$$

$$A_3^i(\mathbf{q}=\mathbf{0}) = \frac{2}{\Gamma(s/2) a^{2+s}} \int_0^{\omega_0} d\omega \frac{1}{\omega^{2+s/2}} \left[-1 + \prod_{j=1}^3 \Theta \left[0; \frac{(a_j/a)^2}{\pi \omega} \right] \right] - \frac{4}{\Gamma(s/2)} \frac{(a_i/a)^2}{a^{2+s}} \int_0^{\omega_0} d\omega \frac{1}{\omega^{3+s/2}} \left[\sum_{l=-\infty}^{\infty} l^2 e^{-(a_i/a)^2 l^2 / \omega} \right] \prod_{j \neq i}^3 \Theta \left[0; \frac{(a_j/a)^2}{\omega \pi} \right]. \quad (20)$$

Clearly, apart from a simple prefactor, $A_2(\mathbf{q}=\mathbf{0})$ and $A_3(\mathbf{q}=\mathbf{0})$ are dependent only on the ratio c/a and not on the particular value of a . Similarly, in the remaining lattices considered here, these quantities are dependent only on the ratios b/a and c/a of the orthorhombic sublattices apart from a prefactor. The b/a ratio is, in these cases, fixed by the symmetry of the basal plane. Thus the anisotropy $A^a(\mathbf{q}=\mathbf{0}) - A^c(\mathbf{q}=\mathbf{0})$ for any of these lattices can be written as a simple prefactor dependent on a and a "universal" function dependent on c/a . This feature, together with the use of the ρ_γ vectors as in (16), allows the computer program which evaluates the sum to be easily modified for any lattice structure and set of lattice parameters.

We have also examined the anisotropy in the ($\mathbf{q}=\mathbf{0}$) (nonsingular) part of A for the lattice relevant to the study of LiTbF₄ (Fig. 2). The Tb atoms in this structure are placed on two body-centered tetragonal sublattices.⁴ The appropriate vectors in (16) are, in this case,

$$\begin{aligned} \rho_0 &= (0, 0, 0), & \rho_1 &= (a/2, 0, c/4), \\ \rho_2 &= (a/2, a/2, c/2), & \rho_3 &= (a, a/2, 3a/4). \end{aligned}$$

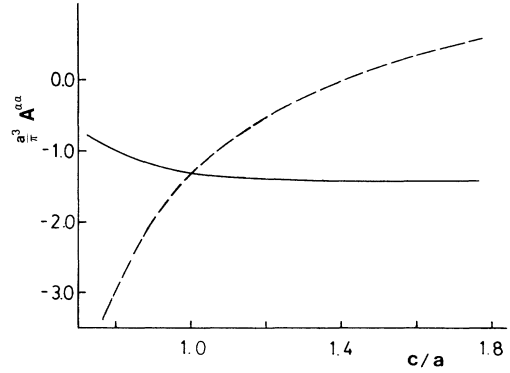


FIG. 1. Dipole interaction of a tetragonal lattice is plotted against the lattice constant ratio c/a . The solid line represents the basal plane component and the dashed line the c -axis component.

lattice as the simplest case in which this anisotropy occurs (Fig. 1). In this case, $\rho=\mathbf{0}$ and $a_1=a_2=a$, $a_3=c$. The contributions to the (nonsingular) q -independent term in A can be written

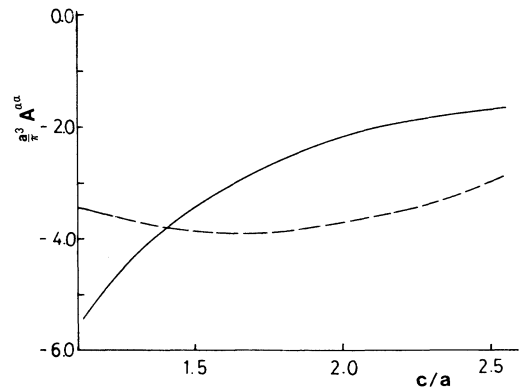


FIG. 2. Dipole interaction of LiTbF₄-type lattice (two body-centered tetragonals) vs the ratio c/a . The solid line represents the basal plane component and the dashed line the c -axis component (c/a ratio of LiTbF₄ is 2.099).

All of the coefficients in (17) were evaluated for the ratio $c/a = 2.10$, appropriate to LiTbF_4 , and compared with the results of Ref. 4 as a further check on our program. Note that, although for a general vector ρ in (16) one might expect to find off-diagonal contributions to $A^{\alpha\beta}$ ($\mathbf{q}=\mathbf{0}$), these do not occur for the systems considered here, since it may readily be shown that the off-diagonal ($\mathbf{q}=\mathbf{0}$) terms vanish for any ρ such that at least two of the Cartesian components of ρ satisfy

$$\rho_i = 0, a_i, \text{ or } a_i/2, \quad i = x, y, z.$$

The simple hexagonal lattice (Fig. 3) is decomposed into two orthorhombic sublattices with $b = \sqrt{3}a$ and the close-packed hexagonal lattice (Fig. 4) can then be constructed from two such simple hexagonal lattices.¹⁰ In addition to the four systems referred to in the figures, we have also studied the anisotropy as a function of c/a in the body-centered tetragonal and face-centered tetragonal lattices.

In all of the cases studied, one may induce a change in the easy direction of magnetization from the c axis to the basal plane by varying the c/a ratio. In most cases, the point at which this crossover occurs can easily be identified with a higher symmetry of the lattice. For example, the crossover points for the tetragonal and hcp lattices (Figs. 1 and 4) occur when the lattices become simple-cubic and "ideal" (hard-sphere packing) hcp structures, respectively. At the crossover point ($c/a = \sqrt{2}$) for the LiTbF_4 structure shown in Fig. 2, each of the body-centered tetragonal sublattices acquires the face-centered cubic structure. The LiTbF_4 body-centered tetragonal and face-centered tetragonal lattices all have three points at which the easy direction of magnetization changes. For the body-centered tetragonal and face-centered tetragonal lattice, there are two points at which the easy direction of magnetization changes from the basal plane to the c axis as the c/a ratio decreases, corresponding to the lattice having either a bcc or fcc structure; however, this change is reversed as the c/a ratio is varied through an intermediate point.

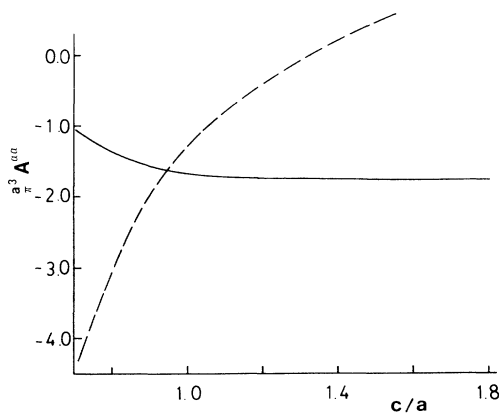


FIG. 3. Dipole interaction of a simple hexagonal vs c/a ratio. The solid line represents the basal plane component and the dashed line the c -axis component.

IV. SUMMARY

The method of Sec. II is of general utility for the performance of lattice sums and is capable of dealing with any lattice type which can be decomposed into orthorhombic sublattices. Notice that while we have dealt only with three-dimensional lattices, the method is suitable for general d -dimensional sums.⁹ For instance, one can extract general d formulas for the tetragonal lattice more easily, perhaps, than by other methods. (The case $d = 2$, of course, requires special care.)

Here we have used the method to investigate the anisotropy in a_3^c of Eq. (17) as a function of the ratio c/a for a variety of lattices, since it is the anisotropy in this parameter which will determine the effective number of components of the order parameter at the phase transition in a dipolar system.³ The dependence of this anisotropy on the c/a ratio is illustrated in the figures for four examples. In physical systems there may, of course, be other factors which affect the effective number of order-parameter components. For example, the crystal field is expected to be the dominant factor in LiTbF_4 .⁴

For reasons given in Sec. I gadolinium is of special interest as a possible example of a dipolar phase transition. The c/a ratio in gadolinium is $c/a = 1.59$, which may be compared with the hcp ideal value of $c/a = (8/3)^{1/2} \approx 1.63$. From our results (Fig. 4), we see that this is consistent with a model of Gd based on s -state ions in a largely isotropic environment. The dipole-dipole interaction is then sufficient to cause the observed uniaxial magnetic ordering at the Curie point (small electronic perturbations could also be present). The value of $\alpha_3^c = 0.13$, obtained for gadolinium here, may be interpreted as an effective change in the critical temperature due to the dipolar interactions from

$$\Delta T_c^{\text{dd}} = \frac{2}{3} S(S+1) \frac{(g_2 \mu_B)^2}{2k_B} \alpha_3^c \approx 1.7 \text{ K}$$

within the mean-field approximation. This indicates that the dipolar nature of the phase transition will be observed only within about 1 K of the critical temperature

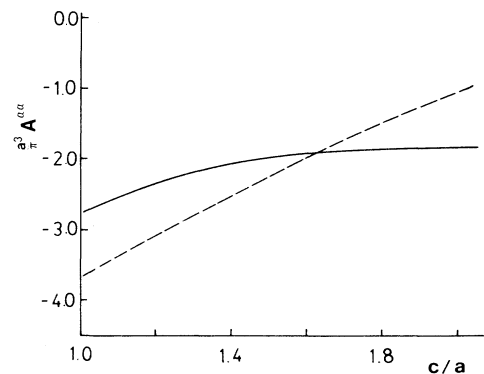


FIG. 4. Dipole interaction of a hexagonal closed-packed lattice vs c/a ratio. The solid line represents the basal plane component and the dashed line the c -axis component.

TABLE I. Coefficients in the Taylor expansion of $a^3 A^{\alpha\beta}(\mathbf{q})$ for hcp lattice structure with $c=5.78 \text{ \AA}$, $a=3.63 \text{ \AA}$, and $c/a=1.59$, as appropriate for Gd near T_c . The wave vector \mathbf{q} in (17) is expressed in units of a^{-1} and becomes dimensionless.

	$A^{xx}(=A^{yy})$	A^{zz}	A^{xy}	$A^{xz}(=A^{yz})$
α_3^α	5.9859	6.2803		
α_4^α	-0.2347	-0.1638		
$\alpha_2^{\alpha\alpha}-\alpha_3^\alpha$	0.6332	0.4543		
$\alpha_1^{\alpha\beta}$	18.2521	18.2521	18.2521	18.2521
$\alpha_2^{\alpha\beta}$			0.6332	0.7749
α_z	-0.0894			

$T_c \approx 293 \text{ K}$. This is consistent with the experimental results referred to earlier (Refs. 5 and 7). We emphasize that this indication of the temperature range where dipole-dipole interactions become relevant and the fact that the order parameter becomes Ising-like are independent of the precise mechanism, whether the dipolar interaction itself or other perturbation is the primary cause of the uniaxiality. Finally, we have calculated the remaining parameters in (17) for gadolinium and these

are presented in Table I.

Our objective has been to develop an efficient method of performing dipole sums in anisotropic lattice systems. The method has been checked on a number of lattices including that for LiTbF_4 . (For LiTbF_4 there are extensive results by other methods and the different sources of anisotropy in LiTbF_4 have been extensively discussed—see Ref. 4.) New results have been given for body-centered and face-centered tetragonal lattices and for hexagonal close-packed structures. In the latter case, dipolar interactions may be important for the critical properties of Gd.⁷ Our understanding of the importance of the various contributions in the critical region to the anisotropy in Gd will be greatly assisted by further experiments.

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- ¹A. Aharony and M. E. Fisher, *Phys. Rev. B* **8**, 3323 (1973); corrections to some of the coefficients in (2) are given in Ref. 4.
²See, e.g., M. E. Fisher and A. Aharony, *Phys. Rev. Lett.* **30**, 559 (1973).
³A. Aharony, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, New York, 1976), Vol. 6, p. 357.
⁴L. M. Holmes, J. Als-Nielsen, and H. J. Guggenheim, *Phys. Rev. B* **12**, 180 (1975); these authors have performed the lattice sums for an anisotropic dipolar system appropriate to the study of LiTbF_4 . However, in this material the easy axis is believed to arise principally from the crystal field.
⁵A. R. Chowdhury, G. S. Collins, and C. Hohenemser, *Phys. Rev. B* **33**, 6321 (1986).

- ⁶G. S. Collins, A. R. Chowdhury, and C. Hohenemser, *Phys. Rev. B* **33**, 4747 (1986).
⁷D. J. W. Geldart, K. De'Bell, J. Cook, and M. J. Laubitz, *Phys. Rev. B* **35**, 8876 (1987).
⁸M. H. Cohen and F. Keffer, *Phys. Rev.* **99**, 1128 (1955); B. M. E. van der Hoff and G. C. Benson, *Can. J. Phys.* **31**, 1087 (1953); M. Born and M. Bradburn, *Proc. Cambridge Philos. Soc.* **38**, 104 (1942).
⁹*Higher Transcendental Functions—Bateman Manuscript Project* (McGraw-Hill, New York, 1953), Vol. 2, p. 294; we use the notation of J. Ambjørn and S. Wolfram, *Ann. Phys. (N.Y.)* **147**, 1 (1983).
¹⁰P. Brüesch and M. Lietz, *J. Phys. Chem. Solids* **31**, 1137 (1970).