for the magnetic moment[14] to estimate η and T_c for *single-crystal* Pr. A particularly simple approach is to neglect exchange between ions with different site symmetries. Then the problem reduces to two independent sublattices and the use of Bleaney's[13] crystal-field scheme allows qualitative agreement with the experimental data at 4.2 K for $\eta(\text{hex}) = 0.5$ to 0.7. On this basis, order-of-magnitude estimates for $T_c(\text{hex})$ and $T_c(\text{cubic})$ are 10 and 3 mK, respectively, for *single-crystal* Pr.

We consider these estimates of η and T_c uncertain enough to think it possible that the hyperfine heat capacity observed for *polycrystalline* Pr might be characteristic of the *single crystal* also. It should be pointed out that the susceptibility anomaly for ordering with ferromagnetic exchange has a large and strongly field-dependent magnitude (for $\eta \sim 0.6$ and ordering on the hexagonal sites only $\chi_{\parallel}$ is 1.3 and 0.4 emu/mole Pr in 100 and 1000 Oe, respectively). The field dependence of T_c, on the other hand, remains quite small.

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Spin-1/2 Decoupling Procedures in the First-Order Green's-Function Theory of Ferromagnetism*

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A decoupling procedure in first-order Green's-function theory of ferromagnetism is devised to give a better treatment for the spin kinematics. This improvement results in better high-temperature characteristics in the Heisenberg model (such as the Curie temperature and zero-field susceptibility) but reintroduces the spurious T^3 term in the low-temperature magnetization. Various first-order decoupling procedures are analyzed in view of their influence on the spin kinematics and the renormalization of quasiparticle energies. The proposed decoupling is also used to study the two-spin system. Over a wide temperature range, better results for various thermodynamics quantities are obtained when compared with previous first-order decouplings.

I. INTRODUCTION

Over the past several years, attempts to describe the thermodynamical properties of the Heisenberg model for all ranges of temperature have made considerable use of the double-time temperature-dependent Green's functions. To solve the problem of the infinite chain of equations that appear in the Green's-function theory, several first-order decoupling procedures have been proposed. In the spin-operator formalism, Bogolyubov and Tyablikov[1] first proposed the so-called random-phase-approximation (RPA) decoupling scheme for the particular case of $S=\frac{1}{2}$. Following the same ideas, Takir-Kheli and ter Haar[2] extended this decoupling procedure for all values of spin. Later, Callen[3] proposed a symmetrical decoupling method guided by a physical criterion, and obtained good results for the case $S \geq 1$.

All these decoupling procedures have special difficulties for the case $S=\frac{1}{2}$. In RPA decoupling, the low-temperature magnetization presents the well-known spurious T^3 term and the renormalization of quasiparticle energies does not give a correct spin-wave behavior at low-temperatures.[2] The good results of this decoupling are those obtained in the high-temperature region ($T=T_c$ and $T \gg T_c$).[1,2] In the case of Callen's approximation,[3] one obtains a good renormalization of quasiparticle energies (in the sense of Keffer and Loudon[4]), but the magnetization also presents a spurious term T^3, and the T^4 term (originating from dynamical interactions) does not appear. For low-spin systems, Callen's results in the high-temperature region are not in good agreement[3,5] with the results of more exact theories, although one can get an improvement in the Curie temperature with some modification in Callen's temperature-dependent parameter.[6] The difficulties for the $S=\frac{1}{2}$ case have also been shown[7] to exist in other thermodynamic quantities. Hewson and ter Haar[8] first attributed these problems to the violation of local kinematics with an intimate connection to the nature of a first-order decoupling.

A new first-order decoupling proposed by Dembinski[9] overcomes the difficulty of Callen's scheme for the special case of $S=\frac{1}{2}$. In this decoupling there is an additional term which takes into account the kinematical effects, resulting in a low-temperature series expansion of the magnetization free from the spurious term T^3 and with a T^4 term quite similar to the one obtained by Dyson.[10] The evaluated Curie temperature is equal to the RPA one, and there is also an improvement in the high-temperature zero-field susceptibility series,[5] which reveals the effect due to the kinematic interactions in this region of temperature. It is worth mentioning that the renormalization of quasiparticle energies in Dembinski's approximation was not explicitly evaluated, but the zero-order Green's function from the iterated solution presents poles identical with those obtained by Callen.[3] We should also mention that none of these first-order decouplings gives a wave-vector-dependent renormalization of the energy, which is an important feature in phase transitions, especially in low-dimensional systems, as was recently analyzed by Lines.[11] However, it should be noted that equally simple first-order decouplings in $\vec{k}$ space give good agreement with Dyson's theory at low-temperature,[12] even if spin variables are used.[13]

Owing to their simplicity, the first-order decoupling procedures have been applied in a wide variety of problems such as ferromagnets including dipolar interactions,[13,14] antiferromagnets,[15] phase transitions,[11] and light scattering of ferromagnets and antiferromagnets.[16] Therefore it is worth seeking a deeper understanding of its successes and failures. Very recently the present authors and Ramos[17] have presented a critical study of these first-order decoupling procedures based on the two-spin system coupled by exchange interaction. They compared the results of these approximations with the exact ones in a wide range of temperature, and almost all the agreements, departures, and discrepancies which appear in the application of those approximations to the Heisenberg ferromagnet are also present in this simple model. They also concluded that among those approximations, Dembinski's is the best for $S=\frac{1}{2}$. Recently, Ramos and Gomes[18] showed that the correlation functions should play a fundamental role in proposing decoupling procedures and have explicitly shown how Callen and Dembinski's decouplings neglect terms of order T^3 in the correlation function $\langle (S_g^-)^2 S_g^+ S_f^+ \rangle$. Based on this method they were able to reproduce Dembinski's decoupling. On the other side, several authors[19] have overcome those difficulties by employing a higher-order Green's-function approximation, and after tedious calculations they obtained Dyson's results for the spontaneous magnetization at low temperatures and the correlation function $\langle (S_g^-)^2 S_g^+ S_f^+ \rangle$ vanishes identically.

The aim of this paper is to investigate the results of a first-order decoupling procedure which gives a better treatment for the spin kinematics. In Sec. II, based on the correlation-function approach,[18] we introduce a first-order decoupling scheme which treats the correlation function $\langle (S_g^-)^2 S_g^+ S_f^+ \rangle$ correctly up to order T^3. However, the spontaneous magnetization evaluated by this scheme (Sec. III) presents a spurious term T^3 and the T^4 term is greater than Dembinski's. These results give evidence that a first-order decoupling procedure (in spin-operator formalism) cannot give simultaneously correct temperature dependence for the spontaneous magnetization and the correlation function $\langle (S_g^-)^2 S_g^+ S_f^+ \rangle$ up to order T^3. Note that this is not true in higher-order Green's-function approximations.[19]

While our low-temperature results are not very good, the high-temperature ($T=T_c$ and $T \gg T_c$) ones are better than those obtained with the first-order decoupling schemes previously mentioned. In Sec. III, using the simple decoupling introduced, we obtain very good results for the Curie temperature and for the high-temperature zero-field susceptibility series for the spin-$\frac{1}{2}$ Heisenberg ferromagnet, as compared to the ones of the laborious method of Ref. 20. Very recently a modification in Callen's decoupling has been proposed.[21] This decoupling, whose introduction is based on the best possible result for the Curie temperature, provides a good description of the system at high temperatures but does not change Callen's results at low and very high temperatures. In the present treatment the decoupling is obtained on a physical basis, and the high-temperature results are better than those of other first-order decouplings (except for the Curie temperature of Ref. 21). We have also applied our approximation to the two-spin system (Sec. IV), obtaining the best results when compared to the ones of other decoupling procedures.[17] These results reveal the importance of the spin kinematics in the high-temperature region. Finally, in Sec. V we present a discussion of the results.

II. DERIVATION OF DECOUPLING

In this section we present the derivation of the decoupling equation used in the remainder of the paper. We use the well-known definition of the temperature-dependent commutator retarded and advanced Green's function for two Heisenberg time-shifted operators $A(t)$ and $B(t)$.[22] The analytic continuation of its Fourier transform $\langle\langle A;B\rangle\rangle_E^{(a,r)}$ to complex energies satisfies the following equation of motion:

$$E\langle\langle A;B\rangle\rangle_E=(i/2\pi)\langle[A,B]\rangle+\langle\langle[A,H];B\rangle\rangle_E\ . \tag{2.1}$$

We assume that the Hamiltonian H contains nearest-neighbor isotropic Heisenberg interactions and the interaction with an external field h

$$H=-J\sum_{\langle ij\rangle}\vec{S}_i\cdot\vec{S}_j-\mu h\sum_i S_i^z\ , \tag{2.2}$$

where μ is the Bohr magneton times the gyromagnetic ratio. With $A=S_g^+$, $B=S_l^-$, where the spin operators are defined by the usual commutation rules, we obtain from Eq. (2.1),

$$(E-\mu h)\langle\langle S_g^+;S_l^-\rangle\rangle_E=(i/2\pi)\langle[S_g^+,S_l^-]\rangle$$
$$+\ 2J\sum_{\langle j\rangle\neq g}(\langle\langle S_j^zS_g^+;S_l^-\rangle\rangle_E-\langle\langle S_g^zS_j^+;S_l^-\rangle\rangle_E)\ . \tag{2.3}$$

The key problem of a first-order decoupling procedure is essentially to express the Green's function $\langle\langle S_g^zS_j^+;S_l^-\rangle\rangle_E$ in terms of lower-order Green's functions, which enables one to solve the infinite chain of equations of motion in a approximate way. Callen[3] proposed a representation of the S_g^z operator in terms of the transverse spin operators (S_g^-, S_g^+) and of a temperature-dependent parameter, which for $S=\frac{1}{2}$ is

$$S_g^z=\langle S^z\rangle+\tfrac{1}{2}(1-2\langle S^z\rangle)S_g^+S_g^--\tfrac{1}{2}(1+2\langle S^z\rangle)S_g^-S_g^+\ . \tag{2.4}$$

For the low-temperature limit $(\langle S^z\rangle\approx\frac{1}{2})$, this equation gives

$$S_g^z=\tfrac{1}{2}-S_g^-S_g^+\ . \tag{2.5}$$

On the other hand, in the high-temperature limit, $(\langle S^z\rangle\approx 0)$ it gives

$$S_g^z=\tfrac{1}{2}(S_g^+S_g^--S_g^-S_g^+)\ . \tag{2.6}$$

Note that (2.5) and (2.6) are exact representations of the S^z operator for $S=\frac{1}{2}$. With the representation (2.4), the Green's function $\langle\langle S_g^zS_j^+;S_l^-\rangle\rangle_E$ is written

$$\langle\langle S_g^zS_j^+;S_l^-\rangle\rangle_E\underset{g\neq j}{=}\langle S^z\rangle\langle\langle S_j^+;S_l^-\rangle\rangle_E+\tfrac{1}{2}(1-2\langle S^z\rangle)$$
$$\times\langle\langle S_g^+S_g^-S_j^+;S_l^-\rangle\rangle_E-\tfrac{1}{2}(1+2\langle S^z\rangle)\langle\langle S_g^-S_g^+S_j^+;S_l^-\rangle\rangle_E\ . \tag{2.7}$$

The two higher-order Green's functions on the right-hand side of this equation have been treated in several ways. Callen[3] treated them using a symmetric decoupling. Ramos and Gomes[18] allowed a more general decoupling for them and, imposing some conditions on the correlation function $\langle(S_g^-)^2S_g^+S_j^+\rangle$, obtained Dembinski's decoupling equation. Here we use the general initial decoupling of Ref. 18 with different conditions on the correlation function. As in Ref. 18, we approximate the higher-order Green's functions of (2.7) to

$$\langle\langle S_g^+S_g^-S_j^+;S_l^-\rangle\rangle_E\underset{g\neq j}{=}\langle S_g^+S_g^-\rangle\langle\langle S_j^+;S_l^-\rangle\rangle_E+\langle S_g^-S_j^+\rangle$$
$$\times\langle\langle S_g^+;S_l^-\rangle\rangle_E\ +A\delta_{g,l}\langle\langle S_j^+;S_l^-\rangle\rangle_E\ , \tag{2.8a}$$

$$\langle\langle S_g^-S_g^+S_j^+;S_l^-\rangle\rangle_E\underset{g\neq j}{=}\langle S_g^-S_g^+\rangle\langle\langle S_j^+;S_l^-\rangle\rangle_E+\langle S_g^-S_j^+\rangle$$
$$\times\langle\langle S_g^+;S_l^-\rangle\rangle_E\ +B\delta_{g,l}\langle\langle S_j^+;S_l^-\rangle\rangle_E\ , \tag{2.8b}$$

which yield, when inserted into Eq. (2.7), the general basic equation of the decoupling:

$$\langle\langle S_g^zS_j^+;S_l^-\rangle\rangle_E\underset{g\neq j}{=}\langle S^z\rangle\langle\langle S_j^+;S_l^-\rangle\rangle_E-2\langle S^z\rangle\langle S_g^-S_j^+\rangle$$
$$\times\langle\langle S_g^+;S_l^-\rangle\rangle_E\ +\tfrac{1}{2}\delta_{g,l}\{A(1-2\langle S^z\rangle)$$
$$-B(1+2\langle S^z\rangle)\}\langle\langle S_j^+;S_l^-\rangle\rangle_E\ . \tag{2.9}$$

The parameters A and B of the correction terms can be determined by the conditions imposed on the correlation function $\langle S_g^-S_g^zS_j^+\rangle$. As pointed out in Ref. 18, there is no *a priori* reason for assuming that the low- and high-temperature values of A and B are the same. However, one can show that if the correlation function is to have the same form in both limits, the values of A and B in those limits must not be different. A first equation connecting A and B can be obtained subtracting (2.8a) and (2.8b) and using the high-temperature limit of (2.4). One obtains

$$\langle\langle S_g^zS_j^+;S_l^-\rangle\rangle_E\underset{g\neq j}{=}[\langle S^z\rangle+\delta_{gl}\tfrac{1}{2}(A-B)]\langle\langle S_j^+;S_l^-\rangle\rangle_E \tag{2.10}$$

and its associate correlation function for $g=l$,

TABLE I. Correlation function $\langle (S_g^-)^2 S_g^+ S_j^+ \rangle$ in the low-temperature limit from various first- and second-order decoupling schemes.

	Present work	Dembinski[a] or RPA	Callen[b]	Second-order[c] Green's function
$\langle (S_g^-)^2 S_g^+ S_j^+ \rangle$	≈ 0 [d]	$\approx \zeta^2(\frac{3}{2})\tau^3$	$\approx 2\zeta^2(\frac{3}{2})\tau^3$	0

[a] References 1 and 9.
[b] Reference 3.
[c] Reference 19.
[d] Up to order τ^3.

$$\langle S_g^- S_g^z S_j^+ \rangle \underset{g \neq j}{=} [\langle S^z \rangle + \tfrac{1}{2}(A-B)]\langle S_g^- S_j^+ \rangle \quad . \tag{2.11}$$

With (2.11) and the correlation function of (2.9), we arrive at

$$A + B = -2\langle S_g^- S_g^+ \rangle \quad . \tag{2.12}$$

Equations (2.11) and (2.12) make now the problem for evaluating the parameters A and B a simple one, once we impose some approximation on the correlation function $\langle S_g^- S_g^z S_j^+ \rangle$.

The choice of the parameters A and B must be guided by a physical criterion in both high- and low-temperature limits. It is well known that in the low-temperature region[3]

$$\langle S^z \rangle \approx \tfrac{1}{2} - a_0 \tau^{3/2} \tag{2.13}$$

and

$$\langle S_g^- S_j^+ \rangle \approx a_0 \tau^{3/2} \quad , \tag{2.14}$$

where a_0 is a constant and τ is a reduced temperature defined by Callen.[3] Substituting these expressions in Eq. (2.11), one gets

$$\langle S_g^- S_g^z S_j^+ \rangle \approx \tfrac{1}{2}\langle S_g^- S_j^+ \rangle - a_0^2 \tau^3 + \tfrac{1}{2}(A-B)\langle S_g^- S_j^+ \rangle \quad . \tag{2.15}$$

The simplest choice is $A = B$, which corresponds to the random-phase approximation for the correlation function $\langle S_g^- S_g^z S_j^+ \rangle$ in both limits of temperature. This choice reproduces Dembinski's basic equation as shown in Ref. 18. We notice that in this case the low-temperature limit of S_g^z in Eq. (2.5) gives, in (2.15)

$$\langle (S_g^-)^2 S_g^+ S_j^+ \rangle \approx a_0^2 \tau^3 \quad . \tag{2.16}$$

Obviously this is a spurious term, because for $S = \frac{1}{2}$ we should have $(S_g^-)^2 = 0$. As pointed out in Ref. 18, Eq. (2.16) is an improvement in the theory as compared to that of Callen, which makes an error of $2a_0^2\tau^3$. The achievement of this approximation was to obtain a magnetization free from the spurious term T^3 and a T^4 term quite similar to Dyson's results. We should mention that in the high-temperature region this approximation gives a molecular-field-theory-type behavior for the correlation function $\langle S_g^- S_g^z S_j^+ \rangle$.

Now we shall introduce a new approximation which treats the correlation function $\langle (S_g^-)^2 S_g^z S_j^+ \rangle$ correctly up to order T^3 in the low-temperature limit. Since the approximation in the correlation function $\langle S_g^- S_g^z S_j^+ \rangle$ is valid for both limits of temperature, we must choose the parameters A and B in such a way that the T^3 term does not appear and that a molecular-field-theory-type behavior in the high-temperature limit is obtained. These two condictions are satisfied if we make

$$A - B = 4\langle S^z \rangle \langle S_g^- S_j^+ \rangle \quad . \tag{2.17}$$

With this condition and using the low-temperature expansions of $\langle S^z \rangle$ and $\langle S_g^- S_j^+ \rangle$, we obtain from Eq. (2.15),

$$\langle S_g^- S_g^z S_j^+ \rangle = \tfrac{1}{2}\langle S_g^- S_j^+ \rangle + O(\tau^{9/2}) \tag{2.18a}$$

and consequently

$$\langle (S_g^-)^2 S_g^+ S_j^+ \rangle = 0 + O(\tau^{9/2}) \quad . \tag{2.18b}$$

The results for the correlation function $\langle (S_g^-)^2 S_g^+ S_j^+ \rangle$ in the low-temperature limit, using different approximations, are presented in Table I.

In the high-temperature limit we have, from Eqs. (2.11) and (2.17), the molecular-field-theory-type behavior for the correlation function,

$$\langle S_g^- S_g^z S_j^+ \rangle = \langle S^z \rangle \langle S_g^- S_j^+ \rangle \Gamma \quad , \tag{2.19a}$$

where

$$\Gamma = 1 + 2\langle S_g^- S_j^+ \rangle \quad . \tag{2.19b}$$

The parameters A and B are easily obtained from (2.12) and (2.17):

$$A = \langle S^z \rangle \langle S_g^- S_j^+ \rangle - \langle S_g^- S_g^+ \rangle \quad , \tag{2.20a}$$

$$B = -\langle S^z \rangle \langle S_g^- S_j^+ \rangle - \langle S_g^- S_g^+ \rangle \quad . \tag{2.20b}$$

Finally, the basic equation of the decoupling to be used is, from Eq. (2.9) and (2.20),

$$\begin{aligned}\langle\langle S_g^z S_j^+ ; S_l^- \rangle\rangle_E \underset{g \neq j}{=} & \langle S^z \rangle \langle\langle S_j^+ ; S_l^- \rangle\rangle_E - 2\langle S^z \rangle \langle S_g^- S_j^+ \rangle \\ & \times \langle\langle S_g^+ ; S_l^- \rangle\rangle_E + 2\delta_{g,l} \langle S^z \rangle (\langle S_g^- S_g^+ \rangle \\ & + \langle S_g^- S_j^+ \rangle) \langle\langle S_j^+ ; S_l^- \rangle\rangle_E \quad . \end{aligned} \tag{2.21}$$

We note that besides Dembinski's correction for the spin kinematics, which is proportional to $\langle S_g^- S_g^+ \rangle$ (same lattice sites), this decoupling introduces a correction which also takes into account the correlations between different lattice sites $\langle S_g^- S_j^+ \rangle$.

III. RESULTS IN HEISENBERG MODEL

In this section we use the decoupling equation to break the chain of equation (2.3) in first order. The aim is to use the Green's function to calculate the low-temperature spontaneous magnetization, the Curie temperature, and the high-temperature zero-field susceptibility for a Heisenberg ferromagnet. The comparison of these thermodynamic quantities with other theories will reveal the ability of the proposed decoupling to describe the Heisenberg model.

The method of calculation we use is the same as Dembinski,[5,9] and we shall not reproduce the details. Following his notation, with the decoupling (2.19), we obtain from Eq. (2.3) the Green's function

$$G(E,\vec{\mathrm{k}}) = \frac{i\sigma}{\pi}\,\frac{1}{E-\tilde{\Omega}_k} + 2\sigma(2\sigma-R)\,J\,\frac{\omega_k}{E-\tilde{\Omega}_k}\,N^{-1} \times \sum_{\lambda}\frac{\gamma_\lambda}{\gamma_0}\,G(E,\vec{\lambda}) \quad , \tag{3.1}$$

where

$$G(E,\vec{\mathrm{k}}) = \sum_{g-l} e^{-i(\vec{\mathrm{g}}-\vec{\mathrm{l}})\cdot\vec{\mathrm{k}}}\langle\langle S_g^+; S_l^-\rangle\rangle_E \quad , \tag{3.2}$$

$$\sigma = \langle S^z\rangle \quad , \tag{3.3}$$

$$\gamma_k = \sum_{\delta} e^{i\vec{\delta}\cdot\vec{\mathrm{k}}} \quad , \tag{3.4}$$

$$\omega_k = \gamma_0 - \gamma_k \quad , \tag{3.5}$$

$$\tilde{\Omega}_k = \mu h + 2\sigma J\omega_k R \quad , \tag{3.6}$$

$$R = 1 + 2N^{-1}\sum_{\lambda}\frac{\gamma_\lambda}{\gamma_0}\,\psi(\vec{\lambda}) \quad , \tag{3.7}$$

$$\psi(\vec{\mathrm{k}}) = \sum_{g-l} e^{-i(\vec{\mathrm{g}}-\vec{\mathrm{l}})\cdot\vec{\mathrm{k}}}\langle S_l^- S_g^+\rangle \quad . \tag{3.8}$$

Here the indices k, g, l, λ, and δ are identified with vectors although the vector sign is left out for clarity of the notation. The key of Dembinski's method is to search for an iterated solution for the Green's-function equation. Carrying out a similar development for Eq. (3.1) and recalling

$$\sigma = \tfrac{1}{2}\langle S^- S^+\rangle \quad , \tag{3.9}$$

we find the following equations for the magnetization and the function R:

$$\tfrac{1}{2} - \sigma = 2\sigma N^{-1}\sum_{k}\Phi(\vec{\mathrm{k}}) + 2\sigma\sum_{n=1}^{\infty}[2\sigma(2\sigma-R)]^n\gamma_0^{-n}J^nN^{-n-1} \times \sum_{\lambda_1\cdots\lambda_{n+1}}\omega_{\lambda_1}\gamma_{\lambda_2}\omega_{\lambda_2}\cdots\gamma_{\lambda_n}\omega_{\lambda_n}\gamma_{\lambda_{n+1}} \times \sum_{m=1}^{n+1}[f'_{n+1}(\tilde{\Omega}_{\lambda_m})]^{-1}\Phi(\vec{\lambda}_m) \quad , \tag{3.10}$$

$$R = 1 + 4\sigma N^{-1}\sum_{k}\frac{\gamma_k}{\gamma_0}\Phi(\vec{\mathrm{k}}) + 4\sigma\sum_{n=1}^{\infty}[2\sigma(2\sigma-R)]^n \times \gamma_0^{-n-1}J^nN^{-n-1}\sum_{\lambda_1\cdots\lambda_{n+1}}\gamma_{\lambda_1}\omega_{\lambda_1}\cdots\gamma_{\lambda_n}\omega_{\lambda_n}\gamma_{\lambda_{n+1}} \times \sum_{m=1}^{n+1}[f'_{n+1}(\tilde{\Omega}_{\lambda_m})]^{-1}\Phi(\vec{\lambda}_m) \quad , \tag{3.11}$$

where

$$\Phi(\vec{\mathrm{k}}) = (e^{\beta\tilde{\Omega}_k} - 1)^{-1} \tag{3.12}$$

and

$$f_{n+1}(Z) = \prod_{m=1}^{n+1}(Z - \tilde{\Omega}_{\lambda_m}) \quad . \tag{3.13}$$

The set of equations (3.10) and (3.11) form the necessary equations to evaluate the thermodynamic quantities of interest. We should note that our Eqs. (3.10) and (3.11) go into Dembinski's equations by replacing $(2\sigma - R)$ with $(2\sigma - 1)$. At this point, it is worth mentioning that the iterative method used to solve Eq. (3.1) will not furnish the actual renormalization of quasiparticle energies. However, we note that the zero-order Green's function from the iterated solution presents poles identical with those obtained by Callen[3] and Dembinski.[9]

A. Low-Temperature Spontaneous Magnetization

The low-temperature spontaneous magnetization up to the T^4 term is evaluated from Eq. (3.10) and (3.11) (with $h=0$) by using the asymptotic expansions for the functions involved in these equations.[9] After some calculations, the result is

$$\sigma = \tfrac{1}{2} - \zeta(\tfrac{3}{2})\tau^{3/2} - \tfrac{3}{4}\pi\nu\zeta(\tfrac{5}{2})\tau^{5/2} - \pi^2\omega\nu^2\zeta(\tfrac{7}{2})\tau^{7/2} - 2\zeta^2(\tfrac{3}{2})\tau^3 - 3\{\tfrac{4}{3}+\tfrac{8}{3}[F(-1)-1]\} \times \pi\nu\zeta(\tfrac{3}{2})\zeta(\tfrac{5}{2})\tau^4 + O(\tau^{9/2}) \quad , \tag{3.14}$$

where ν and ω are well-known lattice structure parameters and $F(-1)$, defined by

$$F(n) = N^{-1}\sum_{k}\left(\frac{\omega_k}{\gamma_0}\right)^n \quad , \tag{3.15}$$

TABLE II. Comparison of the coefficients of T^3 and T^4 terms in the low-temperature expansion of $\langle S^z\rangle$ from different decoupling procedures. The terms are written in the form $q\ \zeta^2(\frac{3}{2})\tau^3$ and $-3\pi\nu\ \zeta(\frac{3}{2})\ \zeta(\frac{5}{2})Q\tau^4$.

	Present work			Dembinski[a]			Callen[b]			RPA[c]			Second-order[d] Green's function		
	sc	bcc	fcc	sc	bcc	fcc	sc	bcc	fcc	sc	bcc	fcc	sc	bcc	fcc
Q	2.70	2.38	2.24	1.68	1.52	1.45		0			1		1.68	1.45	1.35[e]
q		−2			0			+2			−1			0	

[a] Reference 9.

[b] Reference 3.

[c] Reference 1.

[d] Reference 19.

[e] These listed values of Q coincide with the rigorous Dyson results.

TABLE III. Curie temperature ($1/\beta_c J$) for the spin-$\frac{1}{2}$ nearest-neighbor Heisenberg ferromagnet in cubic lattice given by different approximations.

Lattice	Present work	Baker *et al.*[a]	Dembinski[b] or RPA	Callen[c]	Copeland and Gersch[d]	Cooke and Gersch[e]	Swendsen[f]
sc	1.85	1.68	1.98	2.7	2.0	1.91	1.76
bcc	2.74	2.52	2.87	3.7	2.9	2.64	2.60
fcc	4.28	4.01	4.46	5.6	4.5	4.0	4.08

[a] Reference 20.
[b] References 1 and 9.
[c] Reference 3.
[d] Reference 6.
[e] Reference 19. Second-order Green's-function approximation.
[f] Reference 21.

has been evaluated by Watson.[23] The results for the coefficients of the T^3 and T^4 terms, using different approximations, are presented in Table II.

B. Curie Temperature

Equations (3.10) and (3.11) (with $h=0$) are now used in the limit $\sigma \rightarrow 0$ to obtain $\beta_C = (k_B T_C)^{-1}$, where T_C is the Curie temperature. The result is

$$\frac{1}{\beta_C J} = \frac{\gamma_0}{2} \frac{3F(-1)-2}{[2F(-1)-1]^2} \quad . \tag{3.16}$$

In Table III we present the values for the Curie temperature obtained using different approximations, and compare them with the value evaluated by Baker *et al.*[20] from a high-temperature series expansion.

C. High-Temperature Zero-Field Susceptibility

We use again Eqs. (3.10) and (3.11) now in the limit $h \rightarrow 0$, $\sigma \rightarrow 0$ to obtain the susceptibility χ from $\sigma/\mu h \rightarrow \chi/\mu^2$. After a laborious calculation, we find

$$\chi = \frac{\beta\mu^2}{4}\left[1 + \left(\frac{T_m}{T}\right) + \frac{\gamma_0 - 1}{\gamma_0}\left(\frac{T_m}{T}\right)^2 + \frac{\gamma_0^2 - 2\gamma_0 - \epsilon}{\gamma_0^2} \times \left(\frac{T_m}{T}\right)^3 + O\left(\frac{T_m}{T}\right)^4\right] \quad , \tag{3.17}$$

where $\epsilon = 2$ for sc and bcc, and 6 for fcc, and

$$T_m = J\gamma_0/2k_B \quad . \tag{3.18}$$

Once again the decoupling yields better results, this time in the last coefficient, when comparing with RPA, Callen (C), and Dembinski (D) results. In the present treatment,

$$a_3 = a_3^D - \gamma_0^{-2} = a_3^{\mathrm{RPA}} - 2\gamma_0^{-2} \quad . \tag{3.19}$$

In Table IV we list the coefficients given by different approximations and compare them with results of Baker *et al.*[20]

IV. RESULTS IN TWO-SPIN SYSTEM

Let us consider a ferromagnetic system consisting of two spins coupled through the exchange interaction, embedded in a external uniform magnetic field h. In this case the Heisenberg Hamiltonian reads

$$H = -2J\vec{S}_1 \cdot \vec{S}_2 - \mu h(S_1^z + S_2^z) \quad . \tag{4.1}$$

This simple system has been used by the present authors and Ramos[17] to study the validity of the Tyablikov, Callen, and Dembinski decoupling procedures. The advantage of this method is that the comparison, with the exact results, can be analytically done over the whole range of temperature. Now, the necessary equations to our problem are (2.1), (4.1), and (2.21). We refer to Ref. 17 for a better understanding of this problem and to more explicit numerical calculations. Here we shall present the results when using the proposed decoupling. We also present the results for the correlation function $\langle (S_2^-) S_2^+ S_1^+ \rangle$ when using RPA, Callen, and Dembinski decoupling procedures.

The transverse correlation functions are

$$\langle S_2^- S_1^+ \rangle = [\xi/(\tfrac{3}{2}\xi - \langle S^z \rangle)]\langle S^z \rangle \Delta^- \quad , \tag{4.2}$$

TABLE IV. Coefficients a_2 and a_3 of the susceptibility series $\chi = \frac{1}{4}\beta\mu^2 \sum_{n=0} a_n (T_m/T)^n$. The first coefficients a_0 and a_1 are equal to one in all approximations.

	Present work			Baker *et al.*[a]			Dembinski[b]			RPA[c]			Callen[d]		
	sc	bcc	fcc	sc	bcc	fcc	sc	bcc	fcc	sc	bcc	fcc	sc	bcc	fcc
a_2	0.83	0.88	0.91	0.66	0.75	0.83	0.83	0.88	0.91	0.83	0.88	0.91	1	1	1
a_3	0.61	0.72	0.792	0.40	0.54	0.63	0.64	0.73	0.799	0.66	0.75	0.806	0.97	0.98	0.99

[a] Reference 20.
[b] Reference 5.
[c] Reference 1.
[d] Reference 3.

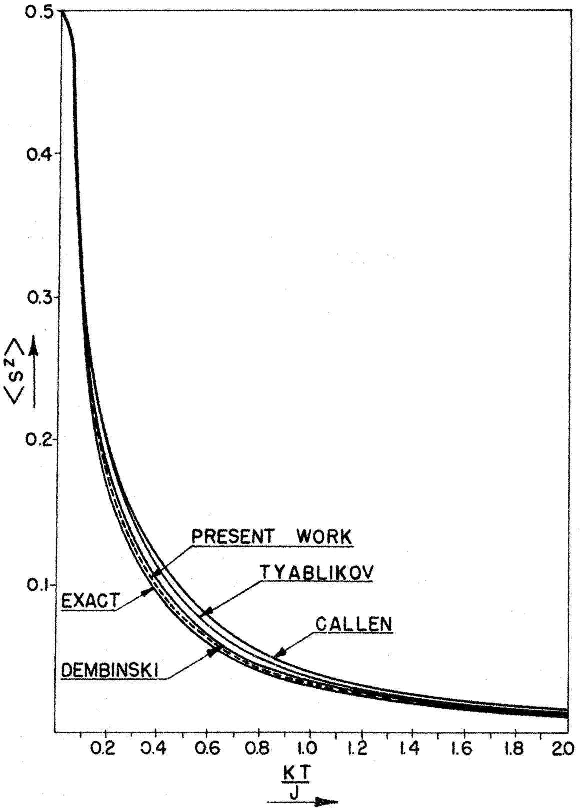

FIG. 1. Temperature dependence of the magnetization for the two-spin system, calculated for $S=\frac{1}{2}$ and $\mu h/J = 0.1$. The exact and the previous first-order decoupling curves (solid lines) were obtained in Ref. 17.

$$\langle S^- S^+\rangle = [\langle S^z\rangle/(\tfrac{3}{2}\xi - \langle S^z\rangle)][\tfrac{1}{2}(3\Delta^+ + \Delta^-)\xi - \langle S^z\rangle(\Delta^+ + \Delta^-)] \;, \tag{4.3}$$

$$\langle (S_2^-)^2 S_2^+ S_1^+\rangle = [\tfrac{1}{2} - \langle S^z\rangle(1 + 2\langle S_2^- S_1^+\rangle)]\langle S_2^- S_1^+\rangle \;, \tag{4.4}$$

$$\langle (S_2^-)^2 S_2^+ S_1^+\rangle_{\mathrm{RPA}} = (\tfrac{1}{2} - \langle S^z\rangle_{\mathrm{RPA}})\langle S_2^- S_1^+\rangle_{\mathrm{RPA}} \;, \tag{4.5}$$

$$\langle (S_2^-)^2 S_2^+ S_1^+\rangle_C = (\tfrac{1}{2} - 2\langle S^z\rangle_C^2)\,\langle S_2^- S_1^+\rangle_C \;, \tag{4.6}$$

$$\langle (S_2^-)^2 S_2^+ S_1^+\rangle_D = (\tfrac{1}{2} - \langle S^z\rangle_D)\langle S_2^- S_1^+\rangle_D \;, \tag{4.7}$$

where

$$\Delta^\pm = (e^{a/t} - 1)^{-1} \pm (\exp\{[a + 4\langle S^z\rangle \times (\tfrac{3}{2}\xi - \langle S^z\rangle)]/t\} - 1)^{-1} \;, \tag{4.8}$$

$$\xi = 1 + 2\langle S_2^- S_1^+\rangle = \frac{2\langle S^z\rangle}{\tanh(a/2t)} \;, \tag{4.9}$$

$$t = k_B T/J \;, \quad a = \mu h/J \;. \tag{4.10}$$

The magnetization is obtained from Eqs. (3.9), (4.3), (4.8), and (4.9) self-consistently. The zero-field magnetic susceptibility is the solution of the following equation:

$$8t^2\chi^3 - 3t\chi^2 + t\chi - \tfrac{1}{8} = 0 \;, \tag{4.11}$$

where

$$\chi = \frac{J}{\mu}\left(\frac{\partial\langle S^z\rangle}{\partial h}\right)_{h=0} \;. \tag{4.12}$$

It is worth mentioning that for the special case $S=\frac{1}{2}$ the exact solution can also be obtained by the Green's-function method dealing only with the first- and second-order equations of motion. The poles of the exact Green's function, which correspond to the exact excitation energies of the system, are $E_1 = \mu h$ and $E_{2,3} = \mu h \pm 2J$.[24] Here we present the temperature-dependent Green's-function poles obtained using the present and other approximations[17]:

$$E_1 = \mu h \;, \tag{4.13}$$

$$E_2 = \mu h + 2J\eta_{(i)}(T) \;, \tag{4.14}$$

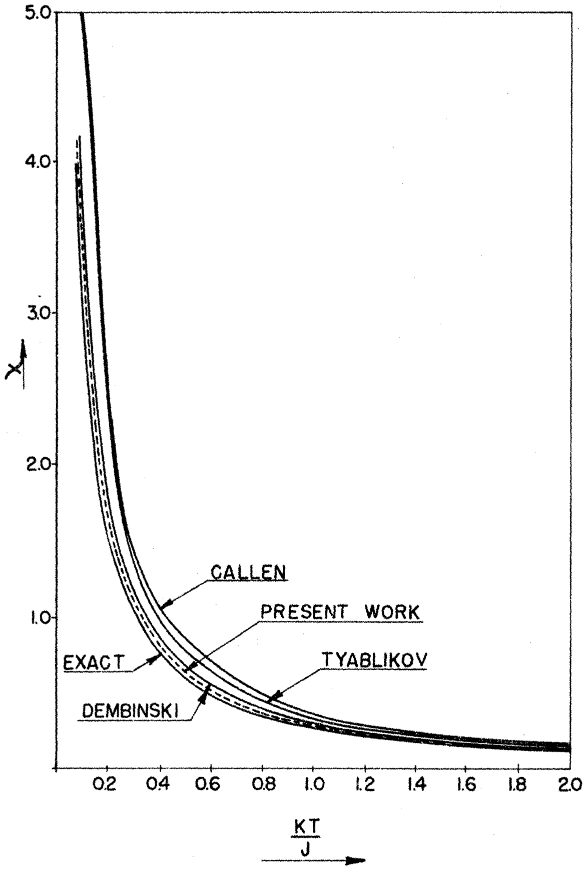

FIG. 2. Zero-field magnetic susceptibility (see the definition in the text) as function of temperature for the two-spin system ($S=\frac{1}{2}$). The solid curves were obtained in Ref. 17.

where

$$\eta(T) = 2\langle S^z\rangle(\tfrac{3}{2} + 3\langle S_2^- S_1^+\rangle - \langle S^z\rangle) \ , \qquad (4.15)$$

$$\eta_{\mathrm{RPA}}(T) = 2\langle S^z\rangle_{\mathrm{RPA}} \ , \qquad (4.16)$$

$$\eta_C(T) = 2\langle S^z\rangle_C(1 + 2\langle S_2^- S_1^+\rangle_C) \ , \qquad (4.17)$$

$$\eta_D(T) = 2\langle S^z\rangle_D[1 + 2\langle S_2^- S_1^+\rangle_D + (\tfrac{1}{2} - \langle S^z\rangle_D)] \ . \qquad (4.18)$$

The results of Eqs. (4.2)–(4.18) as well as the ones obtained with other decoupling schemes,[17] are shown in Figs. 1–4.

V. DISCUSSION OF RESULTS AND CONCLUSIONS

In this work we have investigated the results of a first-order decoupling procedure in the Green's-function theory of $S=\frac{1}{2}$ ferromagnetism devised to give a better treatment for the spin kinematics preserving a good renormalization of quasiparticle energies. The motivation is that it is known that the thermodynamical behavior of the Heisenberg model is dominated by the spin kinematics and the renormalization of the quasiparticle energies.[25] Previously developed decoupling procedures give good treatments of either of these two properties. As an example we note that both the RPA and Callen's ($S=\frac{1}{2}$) decoupling result in a spurious T^3 term (Table II). This is so because in the former the quasiparticle energies are equivalent to simple spin-wave energies renormalized by a factor proportional to the magnetization,[2] whereas the latter obscures the local-spin kinematics (Table I). On the other hand, in Dembinski's[9] approximation the spurious term does not appear because it treats spin kinematics as well as RPA and at the same time it improves the renormalization of energies. This improvement is reached through a renormalization factor for the spin-wave energies which is proportional to the thermodynamic energy (with a leading temperature term arising in $T^{5/2}$[10]) rather than the magnetization. This renormalization factor is expected in the low-temperature region[4] and was also obtained in Callen's decoupling.[3] However, at higher temperatures all these decoupling procedures renormalize the spin-wave energies by the average magnetization. Therefore a particular treatment given for the spin kinematics strongly influences the results in the high-temperature region. As an example we see that the same treatment for the spin kinematics given by RPA and

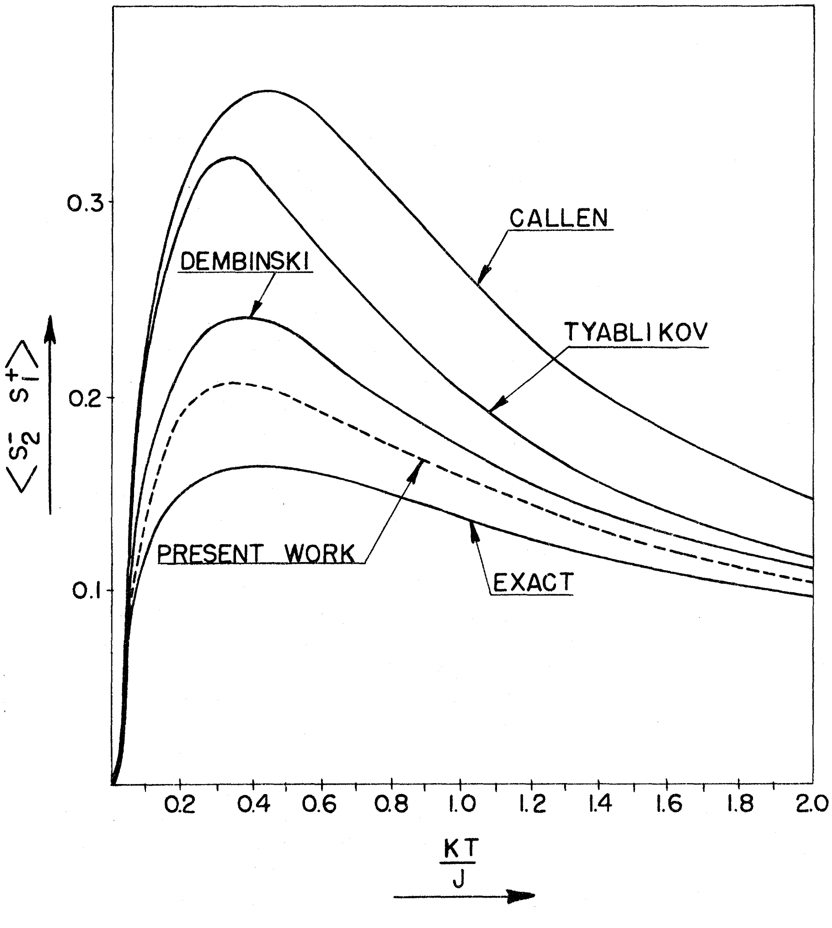

FIG. 3. Temperature dependence of the transverse correlation function for the two-spin system, calculated for $S=\frac{1}{2}$ and $\mu h/J = 0.1$. The solid curves were obtained in Ref. 17.

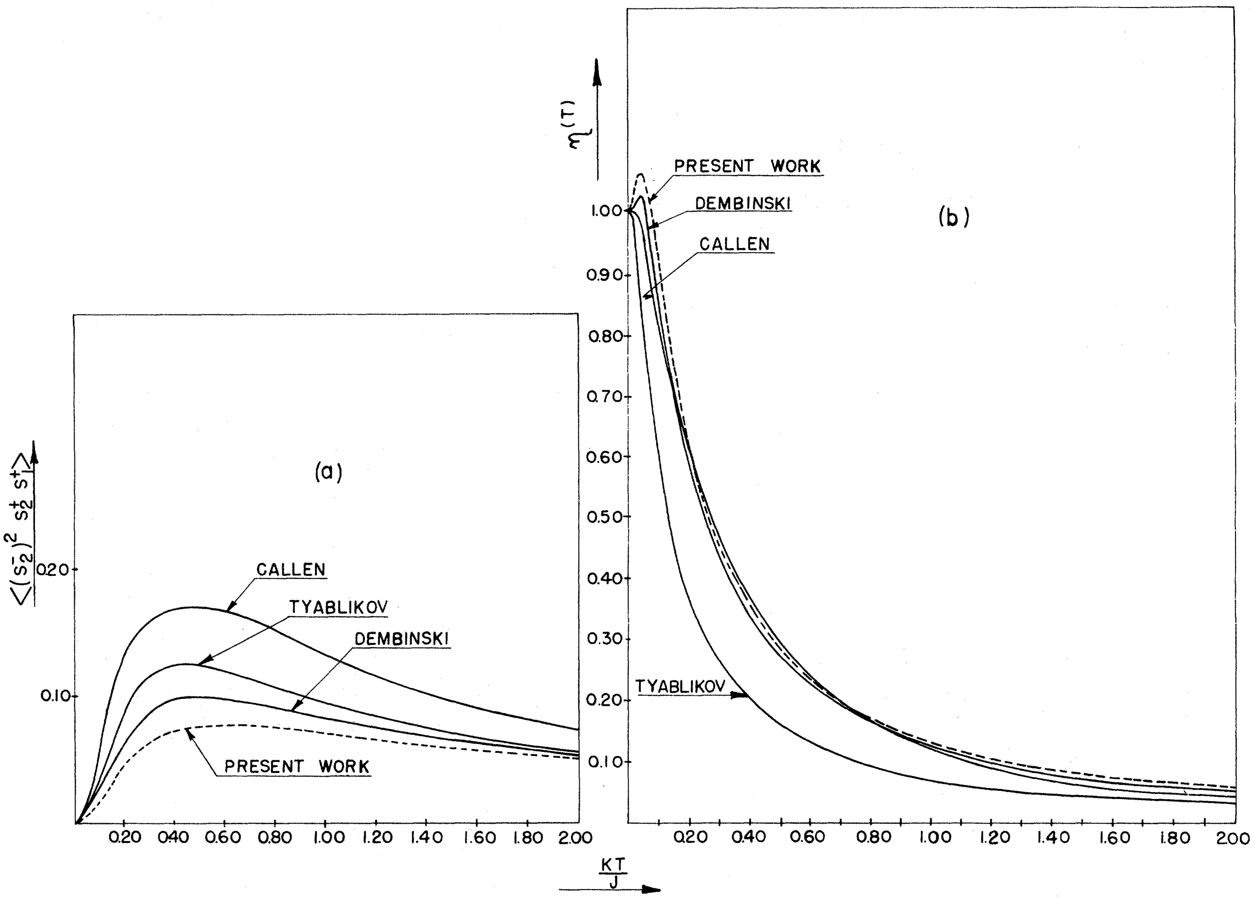

FIG. 4. Temperature dependence of the correlation function $\langle (S_2^-)^2 S_2^+ S_1^+ \rangle$ and the function $\eta(T)$ (see the definition in the text) for the two-spin system ($S=\frac{1}{2}$): (a) treatment of spin kinematics in the various approximations; (b) behavior of the renormalized energies in the various approximations. Note that the exact energies correspond to $\eta=1$ and $\eta=0$.

Dembinski's decouplings (Table I) led them to obtain the same Curie temperature which is better than Callen's temperature (Table III).

The decoupling introduced here was generated with the requirement of treating the correlation function $\langle (S_g^-)^2 S_g^+ S_j^+ \rangle$ correctly up to order T^3, that is zero up to order T^3, to treat better the spin kinematics. At the same time in our treatment the quasiparticle energies are equivalent to simple spin-wave energies renormalized by a factor proportional to the thermodynamic energy at low-temperatures and to the average magnetization at high-temperatures, as in the Callen and Dembinski approximations. This is shown by the zero-order Green's-functions poles in Eq. (3.1). The improvement in the local spin kinematics results in better high-temperature characteristics, such as the Curie temperature and zero-field susceptibility (Tables III and IV). This is a consequence of the fact that at high-temperature the number of spin deviations in the systems is larger and the local-spin kinematics becomes very important.[10] On the other hand, this improvement is responsible for the spurious term T^3 in the magnetization (Table II) which shows up in our treatment. These results seem to indicate that in a first-order decoupling procedure one cannot improve the spin kinematics to obtain better results for all thermodynamics quantities in all ranges of temperatures. In this respect we point out that the first-order decoupling proposed by Dembinski[9] reaches an equilibrium point in treating the spin kinematics at low-temperatures. As a consequence it has no spurious T^3 term in the low-temperature spontaneous magnetization. However, this equilibrium is not maintained at high temperatures.

To understand better the decoupling introduced we have applied it to the two-spin system coupled by exchange. This model is very convenient because it can be solved analytically in the whole range of temperatures. Of course its limitation is that it is not a realistic model for an infinite ferromagnetic crystal. Compared to other first-order decouplings, the one of the present work gives better results for the magnetization, the susceptibility, and the transverse correlation function over a wide range of temperature (Figs. 1–3). Figure 4(a) shows that the spin kinematics is also treated better in the whole temperature range. Figure 4(b) reveals that the improvement of this property results in a variation of the low-temperature renormalized energy. This fact also occurs in the Heisenberg model, but it has no influence in the evaluation of the low-temperature spontaneous magnetization up to order T^4.

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Magnetocrystalline Anisotropy and Spin-Orbit Coupling in Nonspherical Crystal Potentials

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The effects of the crystal field on the magnetocrystalline anisotropy energy are investigated. The matrix elements of $(\alpha^2/4)\vec{\sigma}\cdot[\vec{E}\times(-i\vec{\nabla})]$, the general coupling term between spin and orbit, are derived for nonspherical types of potentials. In the case of cubic transition metals, the anisotropy is shown to arise only from the *d* electrons; it is found that the nonsphericity at a lattice site of the crystal potential cannot give significant changes to the theoretical values of the ferromagnetic anisotropy derived with only the $\vec{L}\cdot\vec{S}$ coupling term. However, this might not be so in other crystals.

I. INTRODUCTION

In ferromagnetic materials, the crystalline energy depends upon the orientation, relative to the crystal axes, of the magnetization. In cubic crystals, symmetry considerations require that the energy has the form[1]

$$E(\alpha_1,\alpha_2,\alpha_3)=E_0+K_1(\alpha_1^2\alpha_2^2+\alpha_2^2\alpha_3^2+\alpha_3^2\alpha_1^2)+K_2(\alpha_1^2\alpha_2^2\alpha_3^2)+\cdots, \tag{1}$$

where α_1, α_2, and α_3, are the direction cosines of the magnetization. The coefficients K_1 and K_2, which depend upon the material and the temperature, have to be determined from the differences between energies measured at various orientations of the magnetization. For nickel at 4.2 °K, Kor-