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High-Temperature Series Expansion for Complicated Level Systems*

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Based on a Green's function formalism, a high-temperature series expansion technique applicable to systems with complicated energy levels is developed. The application of the method to magnetic systems with crystal fields is discussed. As an example, we have obtained the high-temperature series expansion of the susceptibility for a spin-1 ferromagnet with anisotropic exchange interactions and with a uniaxial crystal-field potential.

In this Letter we present a high-temperature expansion technique which is applicable to systems with complicated energy levels. The original motivation for this research was to seek an accurate method to estimate the crystal-field effect on the critical temperature in *real physical* systems such as rare earths and actinides. Intensive research on these compounds has been carried out for twenty or so years.[1] Indeed, with today's experimental techniques, especially with the advancement of neutron-scattering spectroscopy, it is no longer a difficult task to measure accurately the critical temperature and the crystal-field energy scheme of a compound. In fact, even the exchange-interaction parameter can be accurately deduced. The theoretical part of the research, however, seems to lag behind in some respects. For example, when it comes to calculating the critical temperature, the mean-field approximation is used even though the accuracy of the results is known to be very poor. Recently a Green's-function approach[2] and a correlated mean-field method[3] have been proposed, and calculations of critical temperatures for spin systems with a uniaxial crystal-field potential of second degree have been carried out. The critical temperatures thus obtained are presumably quite accurate. It is not easy, however, to assess the accuracy of the results so obtained except by comparing them with the known values, nor is it a simple matter to carry out the calculations to higher orders.

Up to date the most accurate estimations of the critical temperatures are those made by high-temperature series expansion techniques.[4] Such calculations not only provide critical temperatures accurately (generally within 1% uncertainty), it allows estimations of other critical parameters if the series is sufficiently long. The applications of the high-temperature expansion techniques, however, have been limited to simple Ising and Heisenberg models. It is therefore highly desirable to develop a general high-temperature series expansion technique applicable to a more general Hamiltonian such as that which contains a crystal-field potential.

Our high-temperature series expansion method is based on the Green's-function diagrammatic formalism for complicated level systems introduced by Yang and Wang[2] recently. It is different from the high-temperature expansion technique commonly used, but can be classified as a linked-cluster expansion method. The formalism is completely general and can be applied to other physical systems as well as magnetic systems. For convenience of presentation we still consider a magnetic system such as a rare-earth compound, with a Hamiltonian

$$\mathcal{H}=\sum_i V_{ci}-\sum_{ij}J_{ij}\vec{S}_i\cdot\vec{S}_j-\vec{h}\cdot\sum\vec{S}_i\,, \tag{1}$$

where V_{ci} is the crystal-field potential on the ith ion and $\vec{h}$ is the external magnetic field; the exchange pair interaction is represented by the second term with the double sum. $\vec{S}$ denotes the total angular momentum in the case of a rare-earth ion. If we assume a ferromagnetic ordering along the z axis, we split the Hamiltonian into two parts:

$$\mathcal{H}_0=\sum[V_{ci}-2J(0)\langle S^z\rangle-h]S_i^z \tag{2}$$

and

$$\mathcal{H}_1=-\sum_{ij}J_{ij}[S_i^+S_j^-+(S_i^z-\langle S^z\rangle)(S_j^z-\langle S^z\rangle)], \tag{3}$$

where $\langle S^z\rangle$ is the self-consistently determined ordering parameter and $J(0)=\sum_i J_{ij}$. It is a simple matter to obtain the eigenfunctions and eigenenergies of $\mathcal{H}_0$ which is a sum of single-body potentials. $\mathcal{H}_1$ describes the correlations of fluctuations. Ignoring $\mathcal{H}_1$ and solving $\mathcal{H}_0$ we obtain results in the mean-field approximation; the free energy F_0 is

$$-\beta F_0=\ln\sum_n\exp(-\beta\epsilon_n), \tag{4}$$

where ϵ_n are the eigenenergies of $\mathcal{H}_0$ and $\beta=(k_B\times T)^{-1}$. The corrections to F_0 when $\mathcal{H}_1$ is restored can be easily shown[5] to be

$$-\beta\Delta F=\sum_{n=1}^{\infty}\frac{1}{n!}\int_0^\beta d\tau_1\int_0^\beta d\tau_2\cdots\int_0^\beta d\tau_n\langle T_\tau\mathcal{H}_1(\tau_1)\mathcal{H}_1(\tau_2)\cdots\mathcal{H}_1(\tau_n)\rangle_c\,, \tag{5}$$

where the angular brackets denote the canonical thermal averages over $\mathcal{H}_0$. The subscript c denotes the cumulant part of the τ-ordered product, or, in the diagram analysis, the contribution of the connected diagrams. Equations (4) and (5) constitute a series expansion of the free energy in powers of βJ from which other thermodynamic quantities can be obtained. To compute ΔF we need a method to evaluate the thermal averages of the τ-ordered products of spin operators. The method of Vaks, Larkin, and Pikins[6] is not applicable because the inclusion of the crystal-field potential in the unperturbed part of the Hamiltonian destroys the simple τ dependence of spin operators in the interaction representation. A generalized scheme has been furnished recently by Yang and Wang.[2] They used the standard basis operators $L_{mn}\equiv|m\rangle\langle n|$, which transfer an ion from the nth level to the mth level, to represent each spin operator. Because of the simple τ dependence of $L_{mn}(\tau)$ in the interaction representation a Wick-like theorem is possible. The price one pays is the effort in handling a greater number of operators, especially for large spin.

Employing the method of Yang and Wang,[2] the terms given in Eq. (5) can be represented by diagrams, which are constructed with the "semi-invariants" $\langle T_\tau S_i^\alpha S_i^\beta\ldots\rangle_c$ and the exchange interaction lines connecting S_i^z to S_j^z or $S_i^\pm$ to $S_j^\mp$. The semi-invariants are evaluated in the standard basis operator representation. As a consequence, each diagram may be more conveniently considered as consisting of a set of subdiagrams.[2] All the diagrams are evaluated in the Fourier space.

The rules for finding $-\beta\Delta F_n$ which is the term with $(\beta J)^n$ in Eq. (5) are as follows: (1) Construct all diagrams with n interaction lines joining the various semi-invariants. (2) Evaluate the semi-invariants by first replacing each spin operator by a linear combination of standard basis operators and then applying Wick's theorem to the latter operators. It will be more convenient at this point to represent the original diagrams by sets of subdiagrams.[2] (3) Attach a factor $\beta J(\vec{q})$ to each interaction line, $J(q)$ being a Fourier transform of J_{ij}. The momentum $\vec{q}$ and frequency ω_n should be assigned to an interaction line or a propagator in such a way that conservation of momentum and of frequency hold at each vertex. (4) Sum over all momentum and frequency variables. (5) Multiply the sum by the weight factor $P/n!$, P being the number of topologically distinct diagrams obtainable by permuting the indices of the diagram.

As an example of application, we calculate the high-temperature series expansion of the sus-

ceptibility for a spin-1 ferromagnet with a uniaxial crystal-field potential. The Hamiltonian is

$$\mathcal{H} = -D\sum (S_i^z)^2 - \sum_{ij} J_{ij}(S_i^z S_j^z + R S_i^+ S_j^-). \quad (6)$$

Here we also allow anisotropic exchange interaction, but we assume a magnetic ordering along the z axis below the critical temperature T_c. For simplicity, we also assume that the exchange interactions extend to the nearest neighbors only (this restriction can be easily removed). It is convenient to write the susceptibility χ in the form, $\chi^{-1} = \chi_c^{-1} - 2Jz$. The high-temperature expansion of χ_c is then

$$\chi_c/\chi_0 = \sum_{n,m} C_{nm}(t)(1+t/2)^{-n}(2\beta D)^{-m}x^n,$$

where the crystal-field-only susceptibility $\chi_0 = \beta(1+t/2)^{-1}$, $x = \beta J$, $t = e^{-\beta D}$, and the coefficients to the fourth order in βJ are

$$C_{00} = 1, \quad C_2{}^0 = z(-4+t) + \tfrac{1}{2}R^2 z(-2t+t^2), \quad C_2{}^1 = 2R^2 z(-1+t^2),$$

$$C_{30} = \tfrac{1}{3}z(16-8t+t^2) + 2z^3\gamma_3(-4+t) - 2R^2 z t^2 + \tfrac{1}{6}R^3 z^3\gamma_3(2t+t^3),$$

$$C_{31} = 2R^2 z(4+t-2t^2) + R^3 z^3\gamma_3(-6t+4t^2-t^3),$$

$$C_{32} = 2R^2 z(-2-t+2t^2+t^3) + R^3 z^3\gamma_3 + R^3 z^3\gamma_3(-4+7t+4t^2-7t^3),$$

$$C_{40} = \tfrac{1}{12}z(-256+168t-30t^2+t^3) + \tfrac{3}{2}z^2(32-14t+t^2) + 2z^3\gamma_3(16-8t+t^2) + 4z^4\gamma_4(-4+t)$$
$$+R^2[\tfrac{1}{3}z(-6t+29t^2-2t^3) + \tfrac{2}{3}z^2(24t-16t^2+t^3) + \tfrac{1}{3}z^3\gamma_3(-6t-13t^2+t^3)]$$
$$+R^3[-\tfrac{1}{3}z^3\gamma_3(18t+7t^2+2t^3)] + R^4[\tfrac{1}{24}z(18t-35t^2+32t^3)$$
$$+\tfrac{1}{12}z^2(-18t+35t^2-32t^3) + \tfrac{1}{24}z^4\gamma_4(10t+9t^2+10t^3+t^4)],$$

$$C_{41} = R^2[2z(-8+9t^2-t^3) + z^2(16-10t-31t^2+4t^3) + 3z^3\gamma_3(8+2t-3t^2)] + R^3[2z^3\gamma_3(18t-t^2+4t^3)]$$
$$+R^4[\tfrac{1}{2}z(6t-17t^2+8t^3) + \tfrac{1}{2}z^2(12t+22t^2-22t^3+3t^4) - \tfrac{1}{2}z^4\gamma_4(6t+7t^2-2t^3+t^4)],$$

$$C_{42} = R^2[-4z(4+t^2+t^3) + 2z^2(16+6t-t^2) + 2z^3\gamma_3(-16-6t+t^2)]$$
$$+R^3[z^3\gamma_3(16-54t-13t^2+34t^3-t^4)] + R^4[z(-8-34t+6t^2-20t^3+5t^4)$$
$$+\tfrac{1}{2}z^2(8+82t-117t^2+62t^3-11t^4) - \tfrac{1}{2}z^4\gamma_4(30t-69t^2+38t^3-5t^4)],$$

$$C_{43} = R^2[4z(6+t-7t^2-t^3+t^4) + 2z^2(-16-6t+17t^2+6t^3-t^4)$$
$$+2z^3\gamma_3(16+6t-17t^2-6t^3+t^4)] + R^3[2z^3\gamma_3(-8+10t+15t^2-10t^3-7t^4)]$$
$$+R^4[z(20+54t-43t^2-54t^3+23t^4) + 2z^2(4-36t+22t^2+36t^3-26t^4)$$
$$-z^4\gamma_4(12-34t+13t^2+34t^3-25t^4)].$$

Here z is the number of nearest neighbors of an ion; for the three cubic lattices sc (simple cubic), bcc, and fcc, respectively, we have $z = 6$, 8, and 12. $\gamma_n = N^{-1}\sum_k \gamma_k^n$ with $\gamma_k = z^{-1}\sum_{\vec{\delta}} \exp(i\vec{k}\cdot\vec{\delta})$ ($\vec{\delta}$ are vectors connecting an ion to its nearest-neighbor ions). For sc, bcc, and fcc, respectively, $\gamma_3 = 0$, 0, and $4z^{-2}$ and $\gamma_4 = 15z^{-3}$, $27z^{-3}$, and $45z^{-3}$. If we write

$$\chi/\chi_0 = \sum_n a_n(\beta D)x^n,$$

then

$$a_0 = 1, \quad a_1 = 2z(1+t/2)^{-1},$$

$$a_2 = C_2 + a_1{}^2, \quad a_3 = C_3 + 2a_1 C_2 + a_1{}^3,$$

$$a_4 = C_4 + 2a_1 C_3 + 3a_1{}^2 C_2 + a_1{}^4,$$

where $C_n = (1+t/2)^{-n}\sum_m (2\beta D)^{-m} C_{nm}$.

One can use the standard ratio method[4] to estimate the critical temperature T_c. We find that for all three lattices T_c should be accurate to within 2% of the exact value. (For fcc, the uncertainty is less than 1%.) Figure 1 shows, for the case of $R = 1$, $k_B T_c/Jz$ as a function of D/Jz.

To conclude, we stress that our method not only allows the linked-cluster expansion be performed for noncommuting operators, but is applicable to any complicated level system. Since the crystal-field potential can be included in the unperturbed part of the Hamiltonian, the strength of the crystal-field is unrestricted. Neither is the range of pair interaction limited since the quantities entering the calculation are sums in the reciprocal space of products of $J(q)$ and they

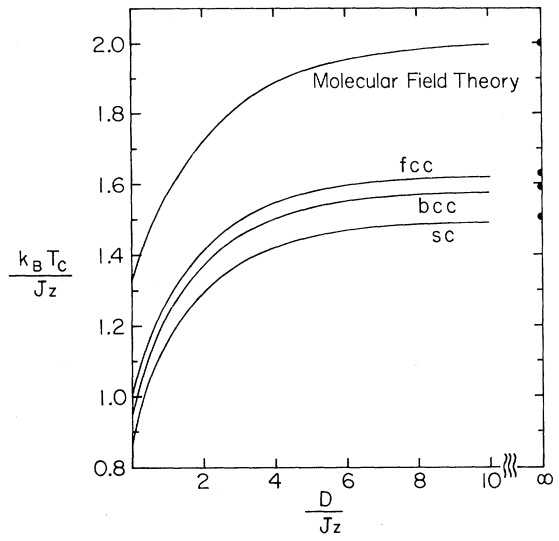

FIG. 1. Plot of critical temperature vs single-ion anisotropy strength for the spin-1 Heisenberg ferromagnet with easy-axis single-ion anisotropy, in sc, bcc, and fcc lattices. The curve showing the values in the molecular-field approximation is included for comparison.

can be computed analytically or numerically without difficulty. With all these flexibilities the method is up to now the only suitable one for the analysis of rare earths and actinides.

Work on higher-order terms of the spin-1 systems reported above and on a singlet-triplet model which can be extended to discuss Pr_3Tl and TbSb is in progress.

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Observation of Interactions between Two Superconducting Phase-Slip Centers*

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A superconducting microbridge has been used as a probe to detect quasiparticle diffusion currents and "heating", produced by a phase-slip center (PSC) in a second microbridge. The critical current of the detector is modulated by the voltage V_{PSC} across the PSC. This modulation provides a measure of quasiparticle current I_Q thru the PSC and gives $I_Q = V_{PSC}/R_D$ for low voltages, R_D being the high-current differential resistance of the PSC.

The dynamics of the voltage-sustaining state in thin-film superconducting microbridge weak links is poorly understood in contrast to Josephson tunnel junctions, and, hence, is the subject of much current investigation. These microbridges are inherently nonequilibrium devices, with their voltage-producing state being the result of a periodic collapse of the order parameter and its subsequent recovery to a state with a change of 2π in the phase difference across the bridge. This process is referred to as a phase slip and the microbridge as a phase-slip center (PSC). During these oscillations both the pair and normal-electron, or quasiparticle, densities and currents are out of equilibrium. Many of the properties of the bridge such as its current-voltage (I-V) curve