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Note on "Approximate Free Energies for Heisenberg Ferromagnets"

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Radcliffe[1] has published a method of treating the Heisenberg ferromagnet by using Peierls's[2] theorem on the minimization of an approximate free energy, applying the theorem to a lattice composed of clusters. The largest cluster treated is the Bethe cluster[3] of $n+1$ atoms, where n is the coordination number of the lattice.

Following Radcliffe, a cluster Hamiltonian allows us to write the approximate density matrix as a product over clusters [his Eq. (V1)] and to obtain a free energy depending on the fields acting on the central atom and its neighbors in the cluster [Eq. (V5)].

Radcliffe then assumes that the partial derivatives of the free energy with respect to these two fields are both zero, i.e., that the fields are independent. This leads to a prediction of a transition temperature (see Table I), but the consistency condition that the average magnetizations of central and neighbor atoms is the same is not satisfied. This condition, in Radcliffe's notation, is

$$\frac{\partial F_\alpha}{\partial h_\alpha} = \frac{1}{n}\frac{\partial F_\alpha}{\partial h'_\alpha} \ ,$$

which yields h_α as a function of h'_α. We modify Radcliffe's procedure by imposing this consistency condition. If we now further require the minimization of the free energy in terms of these two dependent variables, we also obtain a condition that predicts a transition temperature.

To investigate the difference between the transition temperatures predicted by these two methods, we have used the classical spin model[4] to simplify the arithmetic. In our "modified Radcliffe" method, the equation for the Curie temperature T_C is

$$(n+1) - (n-1)L - \tfrac{1}{3}bn(n-1)(1+L) = 0 \ ,$$

where $L = \coth b - 1/b$

and $b = 2JS^2/kT$,

while Radcliffe's method gives (for classical spin)

$$1 - \tfrac{1}{3}b(n-1)[1 + (n-1)L^2] = 0 \ .$$

As shown in the table, the "improvement" of the numerical results that come from requiring consistency is in the right direction, but negligibly small. The reason for this smallness might be found in the following observation: The breakup of the lattice into Bethe clusters gives $N/(n+1)$ clusters and so $Nn/(n+1)$ exchange interactions in these clusters which are included correctly in the approximate product density matrix. The interactions between clusters involve $Nn(n-1)/2(n+1)$ exchange couplings which are considered uncorrelated, i.e., they are replaced by terms of the form $-2J\langle S_z\rangle^2$. It turns out that these are dominant in determining the Curie temperature because of their great number, and this is true whether the theory is self-consistent or not. Hence, the problem is with the method itself, in particular, with the requirement that the density matrix be written as a product.

TABLE I. Predicted transition temperature for a consistency condition.

kT_C/JS^2	$n = 2$	4	6	8
Radcliffe	0.889	2.418	3.824	5.195
Consistent Radcliffe	0.874	2.407	3.817	5.195
Bethe–Peierls–Weiss method	···	1.86	3.25	4.61

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Spin Relaxation via Modulation of the Orbital Energy

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In contrast to previous treatments, we present a spin-lattice-relaxation theory according to which the crystalline-modulated potential is considered as a quasistationary perturbation which entails modulation of orbitals and modulation of energy. One of these effects has been studied previously. We study here the second of them, namely, spin relaxation via modulation of the orbital energy.

In previous papers[1-3] it has been shown that when spin-lattice relaxation occurs, the transitions are induced between wave functions whose orbital part follows adiabatically the displacements of the crystalline complex. The correct way to study spin-lattice relaxation is thus to consider the crystalline-modulated potential as a quasistationary perturbation, whose effect is to entail polarization and rotation of the orbitals. These act secondarily on the spin system, through spin-orbit interaction.

We now consider another adiabatic relaxation mechanism by which the crystalline-modulated potential does not act directly on the spin system but indirectly, namely, relaxation via modulation of orbital energy.

The total Hamiltonian for the paramagnetic system coupled to the rigid lattice can be written as

$$\mathcal{H} = \mathcal{H}_0 + 2\beta\,\vec{S}\cdot\vec{H} + \beta\vec{L}\cdot\vec{H} + \lambda\,\vec{L}\cdot\vec{S} + V_c + \mathcal{H}_L, \quad (1)$$

where $\mathcal{H}_0$ is the Hamiltonian for the free ion, V_c is the crystalline potential, which can be expanded as a function of the deformations of the crystalline complex

$$V_c = V_c^{(0)} + V_c^{(1)}\epsilon + V_c^{(2)}\epsilon^2 + \cdots .$$

$\mathcal{H}_L$ is the Hamiltonian of the lattice vibrations $\mathcal{H}_L = \sum_p (a_p^+ a_p + \frac{1}{2})\hbar\omega$, and $\vec{H}$ is the external magnetic field.

We call φ_0 and φ_n the eigenfunctions of $\mathcal{H}_0 + V_c^{(0)}$, and E_0 and E_n the energies associated with these functions. As shown previously,[1-3] $V_c^{(1)}\epsilon$ for the direct process (and $V_c^{(2)}\epsilon^2$ for the Raman process) can be considered as a slow-varying perturbation, so that the energies associated with $\mathcal{H}_0 + V_c^{(0)}$

$+ V_c^{(1)}\epsilon$ are equal, to first order, to the following expressions:

$$\epsilon_0 = E_0 + \langle\varphi_0 | V_c^{(1)}\epsilon | \varphi_0\rangle,$$

$$\epsilon_n = E_n + \langle\varphi_n | V_c^{(1)}\epsilon | \varphi_n\rangle .$$

If we limit our study to the spin states associated with the ground orbital state, we can use an effective-Hamiltonian formalism. As usual,[4] the effective Hamiltonian can be written as[5]

$$\mathcal{H}_{eff} = \sum_{n\,\sigma} \frac{\mathcal{H}_{pert} | \varphi_n \sigma\rangle \langle \varphi_n \sigma | \mathcal{H}_{pert}}{\epsilon_0 - \epsilon_n} + 2\beta\,\vec{S}\cdot\vec{H}, \quad (2)$$

where $\mathcal{H}_{pert} = \lambda\,\vec{L}\cdot\vec{S} + \beta\vec{L}\cdot\vec{H}$.

Knowing the ability of $V_c^{(1)}\epsilon$ to induce phonon transitions, we can part $\mathcal{H}_{eff}$ into a sum of two terms, one being static and the other modulated. Since the perturbation energies are small with respect to E_0 and E_n, we get

$$\mathcal{H}_{eff} = \sum_{n\,\sigma} \left\{ \frac{\mathcal{H}_{pert} | \varphi_n \sigma\rangle \langle \varphi_n \sigma | \mathcal{H}_{pert}}{E_0 - E_n} \right\}$$

$$\times \left(1 - \frac{\langle\varphi_0 | V_c^{(1)}\epsilon | \varphi_0\rangle - \langle\varphi_n | V_c^{(1)}\epsilon | \varphi_n\rangle}{E_0 - E_n}\right) + 2\beta\vec{S}\cdot\vec{H} .$$

The term

$$\mathcal{H}_{eff}^0 = \sum_{n\,\sigma} \frac{\mathcal{H}_{pert} | \varphi_n \sigma\rangle \langle \varphi_n \sigma | \mathcal{H}_{pert}}{E_0 - E_n} + 2\beta\,\vec{S}\cdot\vec{H} \quad (3)$$

is the static effective Hamiltonian, while the term depending on ϵ is the dynamic one. If we put ϵ outside of the summation this latter one becomes

$$\tilde{\mathcal{H}}_{eff}$$

$$= -\epsilon \sum_{n\,\sigma} \frac{(V_c^{(1)}{}_{00} - V_c^{(1)}{}_{nn})\mathcal{H}_{pert} | \varphi_n \sigma\rangle \langle \varphi_n \sigma | \mathcal{H}_{pert}}{(E_0 - E_n)^2} . \quad (4)$$