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Relaxation in Antiferromagnets due to Spin-Wave Interactions

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For an antiferromagnet it is shown that within perturbation theory the Holstein–Primakoff and Dyson–Maleev transformations do not lead to identical results for either the static or dynamic properties. By examining the spin Green's functions we justify the use of the Dyson–Maleev transformation when there are few spin waves present. Using second-order perturbation theory we find the antiferromagnetic resonance linewidth to be

$$\Delta\omega_0=(64\omega_A\omega_0/\pi^3S^2\omega_E)(kT/\hbar\omega_E)^2\exp(-\hbar\omega_0/kT)\quad\text{for}\quad kT\ll\hbar\omega_0$$

and

$$\Delta\omega_0=[40\omega_A\zeta(3)/\pi^3S^2](kT/\hbar\omega_E)^3\quad\text{for}\quad\hbar\omega_0\ll kT\ll\hbar\omega_E,$$

in qualitative agreement with the experimental results for MnF_2.

RECENTLY the dynamical properties of magnetic insulators have been the subject of several experimental[1] and theoretical[2–6] investigations. The aim of these studies was to determine the imaginary or absorptive part $\chi''(\omega,k)$ of the wave vector and frequency-dependent susceptibility. Although the theoretical interpretation might be expected to be simpler for ferromagnets, technical factors have thus far influenced the experimentalists to study antiferromagnets such as MnF_2. However, the various calculations which can be found in the literature of $\chi''(\omega,k)$ for an antiferromagnet[2–5] are in disagreement with one another. The reasons for these discrepancies are partly due to algebraic difficulties and partly due to different methods of calculation.

To illustrate the latter point we now discuss for a ferromagnet the relative merits of the Holstein–Primakoff[7] (HP) as opposed to the Dyson–Maleev[8] (DM) transformation to bosons. Neglecting kinematic effects, Oguchi[9] has shown that if the terms in the perturbation series for the free energy are arranged in powers of $1/S$, then the HP and DM transformations lead to identical low-temperature results, at least to order $1/S$. One can also compare the lifetime of HP and DM bosons. We write $\mathcal{H}=\mathcal{H}_0+V$ with

$$\mathcal{H}_0=\sum_{\mathbf{k}}\epsilon_{\mathbf{k}}a_{\mathbf{k}}^+a_{\mathbf{k}}=2JzS\sum_{\mathbf{k}}(1-\gamma_{\mathbf{k}})a_{\mathbf{k}}^+a_{\mathbf{k}},\tag{1}$$

where $\gamma_{\mathbf{k}}=z^{-1}\sum_{\delta}\exp(i\mathbf{k}\cdot\boldsymbol{\delta})$ in the usual[9] notation. The form of V depends on which transformation to bosons is used:

$$V_{\rm DM}=(Jz/2N)\sum_{\lambda\sigma\rho}a_{\sigma+\lambda}^+a_{\rho-\lambda}^+a_{\rho}a_{\sigma}(\gamma_{\lambda-\rho}+\gamma_{\lambda+\sigma}-\gamma_{\lambda}-\gamma_{\lambda+\sigma-\rho}),\tag{2a}$$

$$V_{\rm HP}=V_{\rm HP}^{(0)}+(1/S)V_{\rm HP}^{(1)}+(1/S)^2V_{\rm HP}^{(2)},\tag{2b}$$

$$V_{\rm HP}^{(0)}=\tfrac{1}{2}(V_{\rm DM}+V_{\rm DM}^+).\tag{2c}$$

The lifetime of spin waves follows from the golden rule formula:

$$\tau(\mathbf{k})^{-1}=(\pi/2h)\sum_{\lambda\rho}\{\langle\mathbf{k}\lambda\,|\,V\,|\,\mathbf{k}+\rho,\lambda-\rho\rangle\times\langle\mathbf{k}+\rho,\lambda-\rho\,|\,V\,|\,\mathbf{k}\lambda\rangle[\exp(\beta\epsilon_\lambda)-1]^{-1}\times[1-\exp(-\beta\epsilon_{\mathbf{k}})]\delta(\epsilon_{\mathbf{k}}+\epsilon_\lambda-\epsilon_{\mathbf{k}+\rho}-\epsilon_{\lambda-\rho})\},\tag{3}$$

where $|\mathbf{k}\mathbf{k}'\rangle=a_{\mathbf{k}}^+a_{\mathbf{k}'}^+|0\rangle$ and $|0\rangle$ is the state with no spin waves. Following Oguchi[9] we write $V_{\rm DM}=V_{\rm HP}^{(0)}+A$, where

$$A=(Jz/4N)\sum_{\lambda\rho\sigma}a_{\sigma+\lambda}^+a_{\rho-\lambda}^+a_{\rho}a_{\sigma}(\epsilon_\rho+\epsilon_\sigma-\epsilon_{\sigma+\lambda}-\epsilon_{\rho-\lambda}).\tag{4}$$

Using Eq. (3) one sees that

$$\tau(\mathbf{k})^{-1}{}_{\rm HP}-\tau(\mathbf{k})^{-1}{}_{\rm DM}=(\pi/2h)\times\sum_{\lambda\rho}\{|\langle\mathbf{k}\lambda\,|\,A\,|\,\mathbf{k}+\rho,\lambda-\rho\rangle|^2[\exp(\beta\epsilon_\lambda)-1]^{-1}\times[1-\exp(-\beta\epsilon_{\mathbf{k}})]\delta(\epsilon_{\mathbf{k}}+\epsilon_\lambda-\epsilon_{\mathbf{k}+\rho}-\epsilon_{\lambda-\rho})\}.\tag{5}$$

Therefore, by Eq. (4) $\tau(\mathbf{k})_{\rm DM}=\tau(\mathbf{k})_{\rm HP}$.

For an antiferromagnet the DM Hamiltonian is

$$\mathcal{H}=K\sum_{\rm n}\{(2S-1)(a_{\rm n}^+a_{\rm n}+b_{\rm n}^+b_{\rm n})-a_{\rm n}^+a_{\rm n}^+a_{\rm n}a_{\rm n}-b_{\rm n}^+b_{\rm n}^+b_{\rm n}b_{\rm n}\}+J\sum{}'\{2S(a_{\rm n}^+a_{\rm n}+b_{\rm n'}^+b_{\rm n'}+a_{\rm n}^+b_{\rm n'}^++a_{\rm n}b_{\rm n'})-(b_{\rm n'}^+b_{\rm n'}b_{\rm n'}a_{\rm n}+b_{\rm n'}^+a_{\rm n}^+a_{\rm n}^+a_{\rm n}+2a_{\rm n}^+b_{\rm n'}^+a_{\rm n}b_{\rm n'})\},\tag{6}$$

where $a_{\rm n}^+$ ($b_{\rm n}^+$) creates spin deviations on the up (down) sublattice at $\mathbf{R}_{\rm n}$ ($\mathbf{R}_{\rm n}+\boldsymbol{\tau}$) with $\mathbf{R}_{\rm n}=(n_1\mathbf{i}+n_2\mathbf{j}+n_3\mathbf{k})a$ and $\boldsymbol{\tau}=\frac{1}{2}(\mathbf{i}+\mathbf{j}+\mathbf{k})a$. Here J and K are the exchange integral and the microscopic anisotropy

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constant so that the exchange and anisotropy frequencies, ω_E and ω_A, are $\hbar\omega_E=2JzS$ and $\hbar\omega_A=K(2S-1)$. The double sum is restricted so that $\mathbf{R}_n$ and $(\mathbf{R}_{n'}+\boldsymbol{\tau})$ are nearest neighboring lattice sites. If one attempts to transcribe to the case of an antiferromagnet the analysis given above, one finds that to order $(1/S)^0$ the free energy is independent of whichever transformation is used. However, by explicit calculation we have shown[10] that the perturbation series for the free energy using the DM and HP transformations differ in order $(1/S)$. To see that this result is not simply due to an algebraic error we apply the analog of Eq. (5) to the case of an antiferromagnet. We find that $\langle \mathbf{k}\lambda \mid A \mid \mathbf{k}+\boldsymbol{\tau}, \lambda-\boldsymbol{\tau}\rangle$ is in general nonzero even for $\epsilon_k+\epsilon_\lambda=\epsilon_{k+\tau}+\epsilon_{\lambda-\tau}$ so that $\tau_{DM}>\tau_{HP}$. The fact that DM bosons are longer lived than HP bosons is evidence that the former are more nearly the correct normal modes of the system. Two possible explanations of this inequivalence between the two transformations suggest themselves. Firstly, the two perturbation series may not converge, and secondly, the two transformations may induce differing contributions from the "unphysical" states. Accordingly it is necessary to discuss the kinematic effects.

For this purpose we study the spin Green's functions,

$$G_{\alpha\beta}(\mathbf{R},t;\mathbf{R}',t')\equiv -i\langle T(S_\alpha^+(\mathbf{R},t)S_\beta^-(\mathbf{R}',t'))\rangle.$$

Here α and β indicate the sublattice, $+$ or $-$, $\mathbf{R}$ indicates the unit cell, the times t and t' are restricted to the interval $(0, -i\beta)$ on the imaginary axis, and T orders the operators with increasingly negative imaginary times to the left. Wortis[11] has given an elegant treatment of the ferromagnet using these Green's functions. We follow his suggestion and write

$$G_{\alpha\beta}(\mathbf{k},\omega)=\sum_\gamma B_{\alpha\gamma}(\mathbf{k},\omega)D_{\gamma\beta}(\mathbf{k},\omega), \tag{7}$$

where we have taken space and time transforms. The equations of motion for $G_{\alpha\beta}(\mathbf{k},\omega)$ lead to coupled equations for $B_{\alpha\gamma}(\mathbf{k},\omega)$ and $D_{\gamma\beta}(\mathbf{k},\omega)$. It is possible to require that $B_{\alpha\gamma}(\mathbf{k},\omega)$ obey an equation which for $D_{\gamma\beta}(\mathbf{k},\omega)=\delta_{\alpha\beta}$ is identical to that of a Green's function of a system of bosons. Then $D_{\gamma\beta}(\mathbf{k},\omega)$ represents the kinematic effects which distinguish spins from bosons. This treatment is similar to, but more rigorous than that of Ref. 6. The advantage of the decomposition of Eq. (7) is that we can use a boson formalism to calculate $B_{\alpha\gamma}(\mathbf{k},\omega)$ and yet we can discuss the kinematic effect by analyzing $D_{\gamma\beta}(\mathbf{k},\omega)$. We find that in the approximation where we assume only a low density of spin waves, $B_{\alpha\gamma}(\mathbf{k},\omega)$ is identical to the DM boson Green's function. Furthermore $D_{\gamma\beta}(\mathbf{k},\omega)$ probably displays no resonance behavior, so that the variation in $\chi''(\mathbf{k},\omega)$ near resonance is mostly due to the variation in $B_{\alpha\gamma}(\mathbf{k},\omega)$. Accordingly we feel it justified to calculate $\chi''(\mathbf{k},\omega)$ using DM bosons providing ω is near resonance. Far from resonance and for the calculations of the thermodynamic functions it is necessary to calculate $D_{\gamma\beta}(\mathbf{k},\omega)$ accurately.

In analogy with the ferromagnet[6,11,12] one should sum the contributions of all the low-density diagrams. Unfortunately, for the antiferromagnet this does not seem to be feasible owing to the complex structure of the low-density diagrams. However, from Dyson's results[12] one sees that the resulting series in $(1/S)$ for a ferromagnet converges rapidly. Hence for spin S, e.g., for Mn^{++}, second-order perturbation theory will probably give adequate accuracy. Such contributions are of the form of Eq. (3) since it is easily seen[10] that it is impossible to conserve energy and momentum in processes where the number of spin waves is not conserved.

Results for nonzero k will be given elsewhere.[10] For $k=0$ we find the linewidth $\Delta\omega_k=4\pi/\tau_k$ by evaluating the right-hand side of Eq. (3), taking V from Eq. (6). The computations are straightforward but lengthy and hence cannot be given here. The results are

$$\Delta\omega_0=(64\omega_A\omega_0/\pi^3S^2\omega_E)(kT/\hbar\omega_E)^2\exp(-\hbar\omega_0/kT),\qquad kT\ll\hbar\omega_0, \tag{8a}$$

$$\Delta\omega_0=(40w_A\zeta(3)/\pi^3S^2)(kT/\hbar\omega_E)^3,\qquad \hbar\omega_0\ll kT\ll\hbar\omega_E, \tag{8b}$$

where $\omega_0=(2\omega_A\omega_E)^{\frac{1}{2}}$ and $\zeta(3)=\sum_{1,\infty}n^{-3}$. From the experimental results[13] for MnF_2 it is apparent that another relaxation mechanism, perhaps involving impurities, is operative. Therefore we have compared the experimental values of $[\Delta\omega(T)-\Delta\omega(0)]$ with Eqs. (8) and find qualitative agreement for temperature below 45°K. In contrast the results of other authors[2,5] overestimate the linewidth by a factor of two.

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