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Heisenberg Model and Magnetic Insulators*

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The magnetization and specific heat are calculated on the basis of a Heisenberg model which is thought to be appropriate for the description of the magnetic properties of four insulating copper salts. The calculation includes first- and second-neighbor interactions and is based on a quasimagnon approximation which goes beyond the Hartree-Fock theory. The low-temperature expansions are exact to order T^4, and the results obtained for the transition temperature are consistent with those obtained from the high-temperature series-expansion theory. Good agreement between theory and experiment is obtained over most of the temperature range $T < T_c$.

INTRODUCTION

The Heisenberg model of magnetism is a phenomenlogical model based on the concept of localized spins interacting by means of an exchange mechanism. The discovery and subsequent study of the four copper salts $Cu(NH_4)Cl_4 \cdot 2H_2O$, $CuK_2Cl_4 \cdot 2H_2O$, $CuRb_2Cl_4 \cdot 2H_2O$, and $Cu(NH_4)_2Br_4 \cdot 2H_2O$ has made it possible to test the ability of the Heisenberg model to predict the magnetic properties of insulators. These copper salts are spin-one-half isotropic ferromagnets with the ferromagnetic ions positioned approximately on a body-centered-cubic (bcc) lattice. The transition temperature T_c of each of these salts is high enough so that measurements of the magnetization and specific heat can be obtained above and below T_c, but low enough so that phonon effects can be neglected. The relatively low T_c would make neutron-scattering experiments rather difficult but not impossible to do.

Measurements of the magnetization and specific heat have been obtained by Miedema *et al.*[1,2] They indicated that their low-temperature results could be fitted reasonably well to the low-temperature Heisenberg theory with nearest-neighbor interactions. In an attempt to extend the expression for the specific heat to higher temperature they used an expression which was incorrect because of a double counting of the Hartree-Fock magnon-magnon interaction energy.

Wood and Dalton[3] subsequently pointed out the necessity of including second-neighbor interactions because of the bcc magnetic structure. Their work showed that it was possible to obtain much better agreement between the experimental and theoretical predictions of certain critical constants if second-neighbor interactions were included. They did not attempt to incorporate the effects of magnon-magnon interactions in their analysis of the low-temperature behavior of the salts.

The first attempt at incorporating the effects of the magnon-magnon interaction was done by Loly, who used the Hartree-Fock result with first- and second-neighbor interactions.[4] The magnetization and specific heat were found to be double valued and the magnetization remained nonzero at all temperatures. Loly used a value of T_c obtained from high-temperature series expansions to plot his results as a function of T/T_c and found that he could get reasonable agreement with the experiments up to about 80% of the transition temperature. It was also pointed out that there was some slight dependence of the results on the ratio of second-to-first-neighbor exchange.

There are several problems connected with the use of the Hartree-Fock theory. First, it does not reproduce the correct low-temperature contributions to thermodynamic quantities made by the magnon-magnon interaction. Second, it is necessary to go outside the Hartree-Fock theory in order to obtain a value of the transition temperature so that the results can be obtained as functions of T/T_c.

It is the purpose of this paper to go beyond the Hartree-Fock theory and to obtain expressions for the magnetization and the specific heat for arbitrary $T < T_c$ which include the effects of first- and

second-neighbor interactions and which reproduce the exact leading temperature terms contributed by the magnon-magnon interaction. These results are then to be compared with the experimental results obtained by Miedema *et al.* for the copper salts.

The calculations are based on the Green's-function formalism, and the single-spin Green's function is discussed in the first section. Identities relating the magnetization and the specific heat to the single-spin Green's function are used to establish approximate formulas for these quantities for arbitrary $T < T_c$ in the second section. The third section contains the exact low-temperature expansions, and fourth section gives the results of the extrapolation to higher temperature. The contents of the paper are summarized in the last section. Because of the length and complexity of the equations involved, much of the intermediate work has been omitted.

SPIN GREEN'S FUNCTION

The Hamiltonian which is thought to describe these copper salts is the one appropriate for an isotropic bcc spin-one-half ferromagnet with first- and second-neighbor interactions

$$H = g\mu h \sum_{\vec{j}} S_{\vec{j}}^{z} - \sum_{\vec{j}} \sum_{\vec{\xi}} J_{\vec{\xi}} \vec{S}_{\vec{j}} \cdot \vec{S}_{\vec{j}+\vec{\xi}} \,. \tag{1}$$

The first term is the Zeeman energy due to the external field h, and the second term represents the exchange interaction. The double sum in (1) runs first over the neighbors of $\vec{j}$, $\vec{\xi}$ being the neighbor vector, and then over the lattice positions $\vec{j}$. The $J_{\vec{\xi}}$ are the exchange constants which are assumed to depend only on the magnitude of $\vec{\xi}$:

$$\begin{aligned} J_{\vec{\xi}} &= J_1 \quad \text{for nearest neighbors,} \\ &= xJ_1 \quad \text{for second neighbors,} \\ &= 0 \quad \text{otherwise} \quad . \end{aligned} \tag{2}$$

The constant x is the ratio of second-to-first-neighbor exchange.

The quantities of interest in this paper can be calculated from the spin Green's function $\langle\langle S^{+}(t); S^{-}(t')\rangle\rangle$. The space-time transform of this Green's function has been calculated within a T-matrix formalism and was found to have the form[5]

$$G_1(\vec{q}; z) = M/[z - E^0(\vec{q}) - \Sigma(\vec{q}; z)], \tag{3}$$

where M is the relative magnetization and

$$E^0(\vec{q}) = g\mu h + J(0) - J(\vec{q}), \tag{4}$$

$$J(\vec{q}) = \sum_{\vec{\xi}} J_{\vec{\xi}}\, e^{i\vec{q}\cdot\vec{\xi}}, \tag{5}$$

$$\Sigma(\vec{q}; z) = [z - E^0(\vec{q})]\, \Lambda(\vec{q}; z) + \Sigma_D(\vec{q}; z). \tag{6}$$

Expressions for Λ and Σ_D were developed in Ref. 5 in terms of the zero-temperature boson single-particle Green's function and the T matrix for Dyson's magnon-magnon interaction. The same analysis can be carried through in terms of the T-matrix approximation for the boson Green's function. The result is that the leading temperature terms of Σ_D and Λ are given by the analytic continuation ($z_\nu \to z$) of

$$\Sigma_D(\vec{q}; z_\nu) = -\frac{2}{N\beta} \sum_{\vec{k},\nu'} G_1(\vec{k}; z_{\nu'}) \times T_{\vec{q}+\vec{k}}\left(\tfrac{1}{2}(\vec{k}-\vec{q}),\ \tfrac{1}{2}(\vec{k}-\vec{q});\ z_\nu + z_{\nu'}\right), \tag{7}$$

$$\Lambda(\vec{q}; z_\nu) = \frac{2}{N^2\beta} \sum_{\vec{k},\vec{p},\nu'} G_1(\vec{k}; z_{\nu'}) \times T_{\vec{q}+\vec{k}}\left(\tfrac{1}{2}(\vec{k}-\vec{q}),\ \vec{p};\ z_\nu + z_{\nu'}\right) \mathcal{G}_{\vec{q}+\vec{k}}(\vec{k}; z_\nu + z_{\nu'}), \tag{8}$$

respectively, where

$$\beta = 1/k_B T, \quad z_\nu = \pi\nu/(-i\beta), \quad \nu = \text{even integer}, \tag{9}$$

and N is the number of lattice sites. The function $\mathcal{G}$ is the space-time transform of $iG_1(\vec{j}; t)G_1(\vec{j}'; t)$. The equation for T is

$$T_{\vec{K}}(\vec{k}, \vec{k}'; z) = v_{\vec{K}}(\vec{k}, \vec{k}') + (1/N)\sum_{\vec{p}} T_{\vec{K}}(\vec{k}, \vec{p}; z) \times \mathcal{G}_{\vec{K}}(\vec{p}; z)\, v_{\vec{K}}(\vec{p}, \vec{k}'), \tag{10}$$

where

$$v_{\vec{K}}(\vec{k}, \vec{k}') = J(\tfrac{1}{2}\vec{K} + \vec{k}) + J(\tfrac{1}{2}\vec{K} - \vec{k}) - J(\vec{k} + \vec{k}') - J(\vec{k} - \vec{k}') \tag{11}$$

is Dyson's ideal magnon-magnon interaction. The results given above constitute what will be called the T-matrix approximation for G_1.

GENERAL RESULTS

The magnetization M and the specific heat C can be calculated from G_1 by means of the identities[6] ($S = \frac{1}{2}$)

$$M = 1 - (1/2\pi N)\sum_{\vec{q}} \int_{-\infty}^{+\infty} \rho(\vec{q}; \omega) f(\omega)\, d\omega, \tag{12}$$

$$C = \frac{d}{dT}\frac{\langle \mathcal{H} \rangle}{N}, \tag{13}$$

$$\frac{\langle \mathcal{H} \rangle}{N} = E_0 + \frac{1}{2\pi N}\sum_{\vec{q}} \int_{-\infty}^{+\infty} \tfrac{1}{2}[\omega + E^0(\vec{q})] \times f(\omega)\rho(\vec{q}; \omega)\, d\omega, \tag{14}$$

$$f(\omega) = (e^{\beta\omega} - 1)^{-1}, \tag{15}$$

$$\rho(\vec{q}; \omega) = i \lim_{\epsilon \to 0} [G_1(\vec{q}; \omega + \epsilon) - G_1(\vec{q}; \omega - i\epsilon)]. \tag{16}$$

The expression given in Eq. (3) can be rewitten in the form

$$G_1(\vec{q}; z) = \frac{[z - E^0(\vec{q})][M - 1 + \Lambda(\vec{q}, z)) + (1 - M)\Sigma_D(\vec{q}, z)}{[z - E^0(\vec{q}) - \Sigma(\vec{q}; z)]\,[z - E^0(\vec{q}) - \Sigma_D(\vec{q}, z)]}$$

$$+\frac{1}{z-E^0(\vec{q})-\Sigma_D(\vec{q};z)}\,. \tag{17}$$

The first term on the right-hand side of (17) contains the main contributions of the kinematic interaction. There are also kinematic terms in the last term coming from the G_1's appearing in (10) and (7). It is straightforward to prove by direct substitution that the kinematic terms contribute nothing to the magnetization or the specific heat to order T^4. This is a generalization of a similar result found for nearest-neighbor interactions.[7] With the kinematic terms neglected, the last term in (17) reduces to the T-matrix approximation of the single-particle Green's function G_D, calculated with respect to Dyson's ideal spin-wave Hamiltonian, and Σ_D reduces to the self-energy of these ideal spin waves.[8]

If it is assumed that the kinematic terms continue to play a minor role in the calculation of C and M as the temperature is increased, then the only relevant term in (17) is the last term, which is

$$G_D(\vec{q};z)=1/[z-E^0(\vec{q})-\Sigma_D(\vec{q};z)], \tag{18}$$

where from here on Σ_D represents the self-energy for Dyson's ideal spin waves which is given by (10) and (7) with G_1 replaced by G_D. Then, neglecting the kinematic terms, Eq. (14) can be written in the form

$$\frac{\langle\mathcal{H}\rangle}{N}=E_0+\frac{1}{2\pi N}\sum_{\vec{q}}\int_{-\infty}^{+\infty}f(\omega)\,\Gamma(\vec{q};\omega)\times\left(\frac{\omega-E^0(\vec{q})-\Sigma_D(\vec{q};\omega)}{[\omega-E^0(\vec{q})-\Sigma_D(\vec{q};\omega)]^2+[\Gamma(\vec{q};\omega)]^2}\right)d\omega$$
$$+\frac{1}{2\pi N}\sum_{\vec{q}}\int_{-\infty}^{+\infty}f(\omega)[E^0(\vec{q})+\tfrac{1}{2}\Sigma_D(\vec{q};\omega)]\rho_D(\vec{q};\omega)\,d\omega, \tag{19}$$

where

$$\lim_{\epsilon\to 0}\Sigma_D(\vec{q};\omega\pm i\epsilon)=\Sigma_D(\vec{q};\omega)\mp i\,\Gamma(\vec{q};\omega) \tag{20}$$

and ρ_D is the spectral function for G_D.

At low temperatures the width of the spectral function Γ is small and the last term in (19) dominates. It also follows that ρ_D can be evaluated within a quasiparticle approximation. As the temperature, Γ increases and these approximations would eventually become invalid. There is some indication, however, that for the ferromagnet with nearest-neighbor interactions Γ remains sufficiently small so that the quasiparticle approximation could be valid up to at least $\frac{3}{4}\,T_c$.[9] The inclusion of second-neighbor interactions should not affect this result appreciably. Moreover, since Γ does not contribute anything to the leading temperature terms of M or C and since it cannot be expressed in a simple analytical form it seems appropriate as a first approximation to neglect it entirely, in which case

$$\rho_D(\vec{q};\omega)\simeq 2\pi\delta(\omega-E(\vec{q}))\,, \tag{21}$$

where $$E(\vec{q})=E^0(\vec{q})+\Sigma_D(\vec{q};E(\vec{q})) \tag{22}$$

is the quasimagnon energy. The last term in (22) represents the magnon-magnon interaction energy. Substitution of (10), with G_1 replaced by G_D, into (7) gives the leading terms

$$\Sigma_D(\vec{q};z)=\Sigma_1(\vec{q})+\Sigma_2(\vec{q};z)\,, \tag{23}$$

where

$$\Sigma_1(\vec{q})=\frac{2}{N}\sum_{\vec{p}}f(E(\vec{p}))\times[J(\vec{q})+J(\vec{p})-J(\vec{p}+\vec{q})-J(0)], \tag{24}$$

$$\Sigma_2(\vec{q};z)=\frac{2}{N^2}\sum_{\vec{p},\vec{k}}f(E(\vec{p}))\times\frac{T_{\vec{p}+\vec{q}}(\tfrac{1}{2}(\vec{p}-\vec{q}),\vec{k};z+E^0(\vec{p}))}{z+E^0(\vec{p})-E^0(\tfrac{1}{2}(\vec{p}+\vec{q})+\vec{k})-E^0(\tfrac{1}{2}(\vec{p}+\vec{q})-\vec{k})}\,. \tag{25}$$

In deriving (25) all temperature terms except for $f(E(\vec{q}))$ have been neglected. The term Σ_2 is generated by multiple scattering terms in the T-matrix theory. If Σ_2 is neglected in (23), Σ_D reduces to the Hartree-Fock energy.

As a result of the rather long list of approximations made above, (12) and (14) reduce to

$$\frac{\langle\mathcal{H}\rangle}{N}=E_0+\frac{1}{N}\sum_{\vec{q}}f(E(\vec{q}))[E^0(\vec{q})+\tfrac{1}{2}\Sigma_D(\vec{q};E(\vec{q}))], \tag{26}$$

$$M=1-(2/N)\textstyle\sum_{\vec{q}}f(E(\vec{q}))\,. \tag{27}$$

The average energy of the system is then the statistical average of the quasimagnon energy where the $\frac{1}{2}$ in (26) corrects for the double counting of the magnon-magnon interaction energy. If the multiple scattering term Σ_2 is neglected, (25) and (27) reduce to the self-consistent Hartree-Fock results investigated by Loly.

LOW-TEMPERATURE PROPERTIES

The low-temperature magnetization and specific heat can be obtained by expanding (26) and (27) in powers of the temperature. In order to obtain results which are exact up to order T^4, it is only necessary to know the leading temperature and momentum term of the magnon-magnon interaction energy. An exact expression for this quantity can be obtained by expanding the result given in the Appendix by (A7) in powers of the temperature and momentum. Then

$$\Sigma_D(\vec{q};E(\vec{q}))=J_1(qa)^2\,2\pi\nu\zeta(\tfrac{5}{2})Q(x)\times[\tau(x)]^{5/2}+\cdots, \tag{28}$$

$$\tau(x) = 3k_B T/16\pi\nu J_1(1+x) \quad \nu = 3\times 2^{-4/3}, \tag{29}$$

$$Q(x) = 4\sum_{n=1}^{4} A_n \equiv 1 + \tfrac{4}{3}x + S(x)\,, \tag{30}$$

where the A_n's are defined in the Appendix (A11) and $\zeta(m)$ is the Riemann zeta function. The term given in (28) will generate the exact lowest-order contribution to M and C resulting from magnon-magnon interactions.

The quantity $Q(x)$ is a generalization of the Q defined by Dyson for the nearest-neighbor interaction, and it is trivial to show that $Q(x=0)$ reduces to Dyson's result. The first two terms on the right-hand side of (30) represent the Hartree-Fock results, and the remaining terms are corrections due to multiple scattering processes. The ratio

$$R(x) = Q(x)/(1+\tfrac{4}{3}x) \tag{31}$$

is plotted in Fig. 1 as a function of x. This curve indicates that the multiple scattering terms are almost half as big as the Hartree-Fock term and that their relative importance seems to decrease slowly with increasing x. On the basis of this result it might be expected that the Hartree-Fock approximation would improve with increasing range of the interaction. This is apparently the case, since the Hartree-Fock approximation can produce exact results for the infinite-range interaction.[10]

Substitution of (28) into (26) and (27) gives M and C correct to order T^4. The results are

$$M = 1 - 2\zeta(\tfrac{3}{2})\{\tau(x)\}^{3/2} - \tfrac{3}{2}\pi\nu\zeta(\tfrac{5}{2})\{\tau(x)\}^{5/2} - 2\pi^2\omega_0^2\nu^2\zeta(\tfrac{7}{2})\{\tau(x)\}^{7/2} - 6\pi\nu Q(x)\zeta(\tfrac{3}{2})\zeta(\tfrac{5}{2})\{\tau(x)\}^4 + \cdots, \tag{32}$$

$$C/k_B = \tfrac{15}{4}\zeta(\tfrac{5}{2})\{\tau(x)\}^{3/2} + \tfrac{105}{16}\pi\nu\zeta(\tfrac{7}{2})\times\{\tau(x)\}^{5/2}[(1+\tfrac{4}{3}x)/(1+x)] + \tfrac{63}{4}\pi^2\nu^2\omega_0^2 \times(\tfrac{9}{2})\{\tau(x)\}^{7/2}\frac{\{1+\frac{656}{281}x+\frac{528}{281}x^2\}}{(1+x)^2} + 30\pi\nu Q(x)[\zeta(\tfrac{5}{2})]^2\{\tau(x)\}^4 + \cdots, \tag{33}$$

$$\omega_0^2 = \tfrac{281}{288}\,. \tag{34}$$

The τ^4 terms represent the leading contribution to M and C from the magnon-magnon interaction. The remaining terms were first given by Wood and Dalton and are generated by the noninteracting magnon energy $E^0(\vec{q})$.

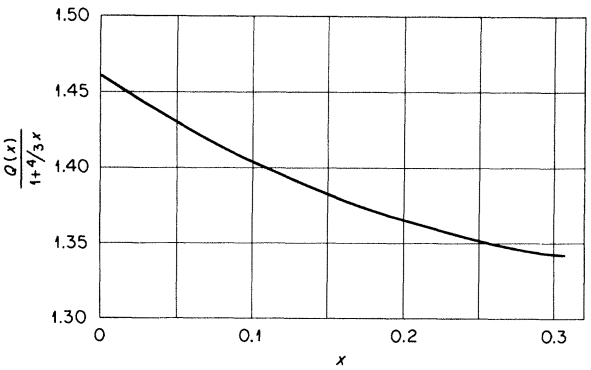

FIG. 1. The ratio of $Q(x)$ to the Hartree-Fock prediction for $Q(x)$, showing the relative importance of multiple scattering processes as a function of the ratio of second- to first-neighbor exchange.

EXTRAPOLATION TO HIGHER T

The extrapolation of the results for M and C to arbitrary temperatures is made on the basis of Eqs. (22), (23), (26), and (27). That is, all kinematic terms are neglected, and the remaining terms are treated within a quasiparticle approximation, where the lifetimes of the quasiparticle states are neglected.

The quantities $E(\vec{q})$ and Σ_D are to be calculated self-consistently by means of Eqs. (22) – (25). This set of equations can be formally solved, but because of Σ_2 the result is too complicated to work with numerically. An approximation for Σ_D which can be used to evaluate (26) and (27) is obtained in the Appendix in terms of $f(E(\vec{q}))$. The result is given by (A7)

$$\Sigma_D(\vec{q}; E(\vec{q})) \simeq -J_1\sum_{n=1}^{4} A_n I_n(T)\epsilon_n(\vec{q})\,, \tag{35}$$

where from (A13)

$$I_n(T) = (1/N)\textstyle\sum_{\vec{q}} f(E(\vec{q}))\epsilon_n(\vec{q})\,. \tag{36}$$

The quantities A_n and ϵ_n are defined in the Appendix. Equations (35) and (22) should not be used as a self-consistent set of equations to solve for $E(\vec{q})$, since this would greatly amplify the errors inherent in (35).

The approximation that has been used here is to retain Σ_2 only to first order when calculating $f(E\times(\vec{q}))$. That is, (35) is substituted into (22) and the terms generated by Σ_2 are neglected in the temperature factors $I_n(T)$. The result is

$$f(E(\vec{q})) \simeq f\Big(J_1\sum_{n=1}^{4} K_n(T)\epsilon_n(\vec{q})\Big)\,, \tag{37}$$

where
$$K_n(T) = 1 - A_1\tilde{I}_1(T), \quad n=1$$
$$= x - A_2\tilde{I}_2(T), \quad n=2$$
$$= -A_n\tilde{I}_n(T), \quad n=3,4 \tag{38}$$

$$\tilde{I}_n(T) = (1/N)\textstyle\sum_{\vec{q}} f(E_1(\vec{q}))\epsilon_n(\vec{q})\,, \tag{39}$$

$$E_1(\vec{q}) = J_1\sum_{n=1}^{2} K_n(T)\epsilon_n(\vec{q})\,. \tag{40}$$

In order to simplify the results, the identity

$$E^0(\vec{q}) = J_1\epsilon_1(\vec{q}) + xJ_1\epsilon_2(\vec{q}) \tag{41}$$

has been used. The term $E_1(\vec{q})$ is simply the Hartree-Fock energy.

Substitution of these results into (26), (27), and (13) gives

$$M = 1 - \frac{2}{N}\sum_{\vec{q}} f\left(J_1 \sum_{n=1}^{4} K_n(T)\epsilon_n(\vec{q})\right) , \tag{42}$$

$$C = J_1 \sum_{n=1}^{4} K_n(T)\frac{dI_n(T)}{dT} , \tag{43}$$

$$I_n(T) = \frac{1}{N}\sum_{\vec{q}} f\left(J_1 \sum_{n=1}^{4} K_n(T)\epsilon_n(\vec{q})\right)\epsilon_n(\vec{q}). \tag{44}$$

These results can be shown to reduce to the exact low-temperature theory given in the previous section, and also they reduce to the Hartree-Fock theory investigated by Loly if the terms generated by Σ_2 are neglected.

When these results were evaluated on a computer, it was found that the relative magnetization was double valued, but that at most only one value of M for each temperature was positive. The temperature at which M became zero was taken as the transition temperature T_c. The specific heat was found to increase with temperature and apparently to diverge at the same T_c as that found from the magnetization curves. The calculations were made for various values of x in the range $0 \leq x \leq 0.3$ since it was felt that if $x > 0.3$ higher-neighbor interactions should be taken into account. The numerical results for

$$\theta = k_B T/J_1 \tag{45}$$

were found to vary linearly with x according to

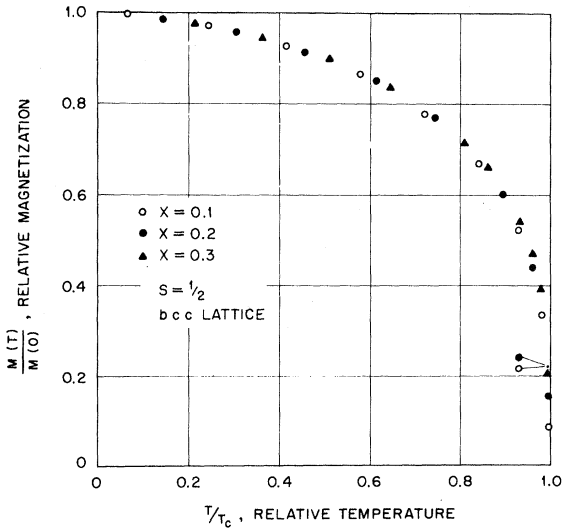

FIG. 2. Theoretical magnetization curve for a bcc lattice for three values of x, indicating the insensitivity of $M(T)/M(0)$ to x.

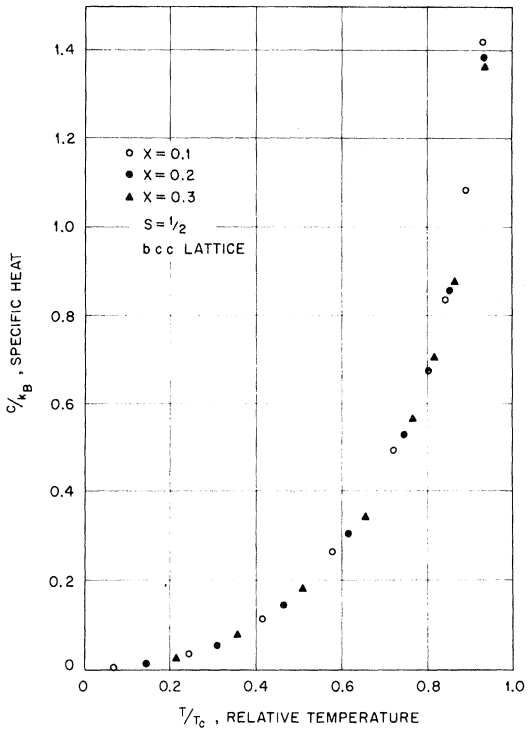

FIG. 3. Theoretical specific-heat curve for a bcc lattice for three values of x, indicating the insensitivity of C to x.

$$\theta_c(x) = \theta_c(x=0)\ [1+x] ,$$
$$\theta_c(x=0) \simeq 2.64 . \tag{46}$$

The value of $\theta_c(x=0)$ is somewhat higher than that predicted by high-temperature series expansions (2.52 – 2.55), but the dependence of θ_c on x is in excellent agreement with the result found by Wood and Dalton,[3] namely,

$$\theta_c(x) = \theta_c(x=0)\ [1+0.99x]. \tag{47}$$

The results of the calculations for M and C are shown in Figs. 2 and 3, respectively. These functions are plotted with respect to $\theta/\theta_c = T/T_c$, where θ_c is obtained from (46), for various choices of x. Clearly, the results are essentially independent of x. An indication of why this is the case can be seen from Eqs. (32) and (33). The major x dependence is incorporated in the function $\tau(x)$, which from (29), (45), and (46) is

$$\tau(x) = \frac{3\theta(x)}{16\pi\nu(1+x)} = \frac{3\theta_c(x=0)}{16\pi\nu}\left(\frac{T}{T_c}\right) . \tag{48}$$

Thus if M or C is plotted as a function of T/T_c, most of the x dependence is missing. This is consistent with the experimental results obtained by Miedema *et al.*[1,2] for the copper salts. It also proves that if M and C are written as functions of θ/θ_c, there are essentially no adjustable parameters in the theory.

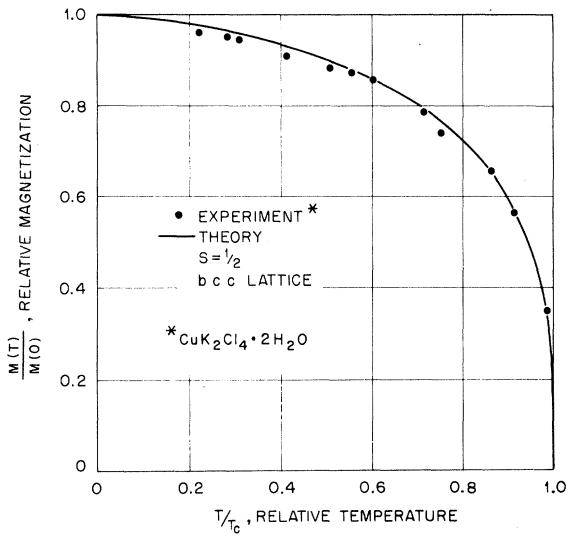

FIG. 4. The relative magnetization of $CuK_2Cl_4 \cdot 2H_2O$ compared with theory.

A comparison of the theoretical and experimental results for M and C is given in Figs. 4 and 5, respectively. The agreement is rather good considering the number of approximations that have been made. It should be pointed out here that for $T/T_c > 0.85$ the expression used for C amounts to the difference between two very large quantities and, therefore, the accuracy of values for C in this temperature range is probably not too good.

CONCLUSIONS

Before considering any interpretation of the results obtained in this paper, it is important to emphasize the fact that Eqs. (42) and (43) represent approximations to the actual behavior of M and C predicted by the Heisenberg model. The accuracy of these equations is certainly open to question. It is true that the approximations which were used to obtain these results are valid at low temperatures, and the low-temperature expansions of (42) and (43), given by (32) and (33), are exact or order T^4. It is also true that the value of $\theta_c(x=0)$ and the variation of θ_c with x are in good agreement with the high-temperature expansion results for the ferromagnet. This, of course, does not imply that the detailed behavior of M or C is correctly predicted near T_c, but it does indicate that the temperature extrapolation represented by (42) and (43) is not totally meaningless.

If one accepts that these expressions are reasonably good for arbitrary $T < T_c$, then the results of the last section are further evidence that the Heisenberg model is indeed a good model for magnetic insulators. These results indicate that if M (or C) is considered as a function of $\theta/\theta_c = T/T_c$, the result is essentially independent of x. This agrees with the experimental findings, and also indicates that the values of J_1 and x do not necessarily have to be the same for each of the copper salts.[3]

The inclusion of the terms beyond the Hartree-Fock theory treated by Loly allows one to obtain rigorous expressions for the leading terms contributed by the magnon-magnon interaction to the specific heat and the magnetization. The leading temperature term in the momentum expansion of the magnon energy can also be evaluated rigorously. The numerical results obtained here seem to be in reasonable agreement with the Hartree-Fock theory for $T/T_c \lesssim 0.9$, except for the slight variation of the Hartree-Fock results with x.[4] Above this temperature the results for positive M and C become double valued in the Hartree-Fock theory but remain single valued in the theory presented here, which explains why this theory predicts a transition temperature and the Hartree-Fock theory does not.

Finally, it should be pointed out that the theoretical fits obtained in the last section do not de-

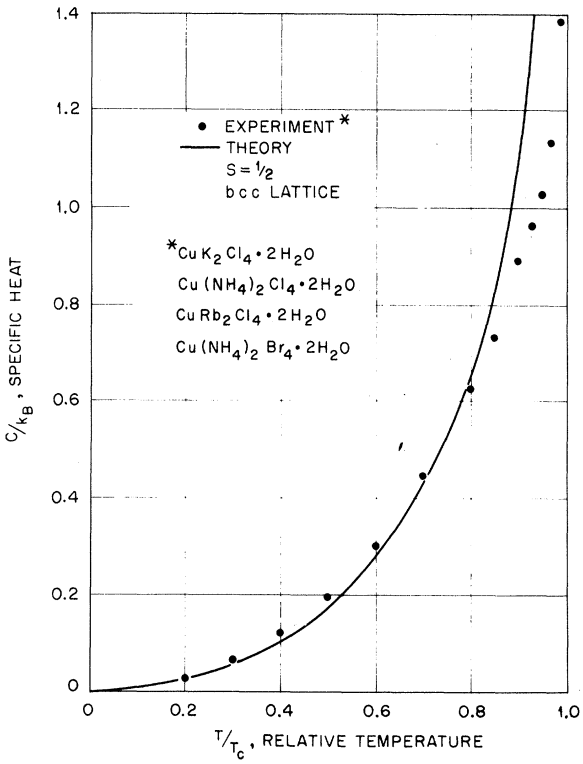

FIG. 5. The specific heat of the four copper salts compared with theory.

pend on any adjustable parameter, and they do not contain any of the contributions from the kinematic interaction, lifetimes of the quasimagnon states, or the magnon-magnon bound states. Presumably these contributions remain small over most of the temperature range $T < T_c$.

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APPENDIX

Expressions for Σ_2 and Σ_D are obtained in this Appendix. The leading temperature terms for Σ_D can be obtained by substituting the identity

$$G_D(\vec{q};\, z) = (1/2\pi) \int_{-\infty}^{+\infty} [\rho_D(\vec{q};\, \omega)/(z-\omega)]d\omega \quad \text{(A1)}$$

into (7), after replacing G_1 by G_D, and then using (21). The result is

$$\Sigma_D(\vec{q};\, E(\vec{q})+i\epsilon) \simeq \frac{2}{N}\sum_{\vec{p}} f(E(\vec{p}))$$
$$\times [J(\vec{p})+J(\vec{q})-J(\vec{p}+\vec{q})-J(0)]+\frac{2}{N^2}\sum_{\vec{p},\vec{k}} f(E(\vec{p}))$$
$$\times \frac{T_{\vec{p}+\vec{q}}(\frac{1}{2}(\vec{p}-\vec{q}),\, \vec{k};\, E^0(\vec{q})+E^0(\vec{p})+i\epsilon)\, v_{\vec{p}+\vec{q}}(\vec{k},\, \frac{1}{2}(\vec{p}-\vec{q}))}{E^0(\vec{q})+E^0(\vec{p})-E^0(\frac{1}{2}(\vec{p}+\vec{q})+\vec{k})-E^0(\frac{1}{2}(\vec{p}+\vec{q})-\vec{k})+i\epsilon}\,, \quad \text{(A2)}$$

where the T matrix is given by (10) with

$$\mathcal{G}_{\vec{K}}(\vec{k};\, z) = 1/[z - E^0(\tfrac{1}{2}\vec{K}+\vec{k}) - E^0(\tfrac{1}{2}\vec{K}-\vec{k})]\,. \quad \text{(A3)}$$

The T-matrix equation can be solved but the result is too complicated to treat numerically. An approximation which greatly simplifies the result and also leads to the correct low-temperature results for C and M is based on the matrix B, which enters into the solution for the T matrix:

$$B(\vec{\eta},\, \vec{\xi}) = D(\vec{\eta},\, \vec{\xi}) - \cos\tfrac{1}{2}(\vec{p}+\vec{q})\cdot\vec{\xi}\, D(\vec{\eta},\, 0)\,, \quad \text{(A4)}$$

$$D(\vec{\eta},\, \vec{\xi}) = -\frac{4J_1}{N}\sum_{\vec{k}}$$
$$\times \frac{\cos\vec{k}\cdot\vec{\eta}\cos\vec{k}\cdot\vec{\xi}}{E^0(\vec{q})+E^0(\vec{p})-E^0(\frac{1}{2}(\vec{p}+\vec{q})+\vec{k})-E^0(\frac{1}{2}(\vec{p}+\vec{q})-\vec{k})+i\epsilon}, \quad \text{(A5)}$$

where $\vec{\eta}$ and $\vec{\xi}$ represent neighbor vectors. The approximation mentioned above is to neglect the $\vec{p}$ and $\vec{q}$ dependence of B. This has the effect of discarding the effects produced by the magnon-magnon bound states. This also allows us to drop the ϵ dependence since the resulting expression for D, given by

$$D(\vec{\eta},\, \vec{\xi}) = \frac{2J_1}{N}\sum_{\vec{k}} \frac{\cos\vec{k}\cdot\vec{\eta}\cos\vec{k}\cdot\vec{\xi}}{E^0(\vec{k})}\,, \quad \text{(A6)}$$

converges in three dimensions.

The result for Σ_D then becomes

$$\Sigma_D(\vec{q};\, E(\vec{q})) \simeq -J_1\sum_{n=1}^{4} A_n I_n(T)\epsilon_n(\vec{q})\,. \quad \text{(A7)}$$

In defining the various terms in (A7), it is convenient to let $\{\vec{\xi}\}$ represent the set of nearest neighbors and $\{\vec{\eta}\}$ represent the set of second neighbors. Then let

$$D(\vec{\xi},\, \vec{\xi}') = a_1 \text{ if } \vec{\xi}=\vec{\xi}',\quad D(\vec{\eta},\, \vec{\eta}') = a_2 \text{ if } \vec{\eta}=\vec{\eta}',$$
$$= b_1 \text{ if } \vec{\xi}\neq\vec{\xi}',\qquad = b_2 \text{ if } \vec{\eta}\neq\vec{\eta}',$$
$$D(\vec{\xi},\, \vec{\eta}) = b_3,\quad D(\vec{\xi},\, 0) = \tfrac{1}{4}\alpha_1,\ D(\vec{\eta},\, 0) = \tfrac{1}{3}\alpha_2, \quad \text{(A8)}$$
$$\Gamma_1 = a_1 - b_1,\qquad \Gamma_2 = a_2 - b_2\,,$$
$$\mathcal{G}_1 = 3[b_3 - \tfrac{1}{4}\alpha_1],\quad \mathcal{G}_2 = 4[b_3 - \tfrac{1}{3}\alpha_2]\,, \quad \text{(A9)}$$
$$R = 1 + x\mathcal{G}_1 + \mathcal{G}_2\,.$$

The quantities α and Γ are generalizations of the α and Γ defined by Dyson for the nearest-neighbor interaction,[7] and they are functions of x since

$$E^0(\vec{q}) = J_1[\textstyle\sum_{\vec{\xi}} e^{i\vec{q}\cdot\vec{\xi}} + x\sum_{\vec{\eta}} e^{i\vec{q}\cdot\vec{\eta}}]\,. \quad \text{(A10)}$$

Then, the quantities A_n are defined by

$$A_1 = \frac{1}{4}\left[1 + \frac{1}{2}\alpha_1 + \frac{3}{2}\frac{\Gamma_1}{1-\Gamma_1} + 2x\mathcal{G}_1 R\left(\frac{\alpha_2}{3} - \frac{\alpha_1}{4} - \frac{1}{4}\right)\right],$$

$$A_2 = \frac{x}{3}\left\{1 + \frac{2}{3}x\alpha_2 + \frac{3}{2}\frac{x\Gamma_2}{1-x\Gamma_2} + 2x\mathcal{G}_2 R\left[x\left(\frac{\alpha_1}{4} - \frac{\alpha_2}{3}\right) - \frac{1}{3}\right]\right\}$$

$$A_3 = \frac{1}{6}\left[\frac{\alpha_1}{4} - \frac{1}{4}\frac{\Gamma_1}{1-\Gamma_1} + x\mathcal{G}_1 R\left(\frac{\alpha_2}{3} - \frac{\alpha_1}{4} - \frac{1}{4}\right)\right], \quad \text{(A11)}$$

$$A_4 = \frac{2}{3}xR\left[\frac{\alpha_1}{4} + \frac{\alpha_2}{3} + \frac{1}{2}\alpha_1\mathcal{G}_2 + \frac{1}{4}\mathcal{G}_2 + \frac{1}{3}\mathcal{G}_1\right.$$

$$+x\left(\frac{1}{3}\mathcal{G}_1\alpha_2+\frac{1}{4}\mathcal{G}_2\alpha_1\right)\Big] ;$$

ϵ_n by $\epsilon_1(\vec{q})=\sum_{\vec{\xi}}(1-e^{i\vec{q}\cdot\vec{\xi}})$,

$$\epsilon_2(\vec{q})=\sum_{\vec{\eta}}(1-e^{i\vec{q}\cdot\vec{\eta}}) ,$$

$$\epsilon_3(\vec{q})=\sum_{\vec{\xi}\neq\vec{\xi}'}\sin\frac{1}{2}\vec{q}\cdot\vec{\xi}'\,\frac{\vec{\xi}\cdot\vec{\xi}'}{|\vec{\xi}\cdot\vec{\xi}'|} , \qquad \text{(A12)}$$

$$\epsilon_4(\vec{q})=\frac{1}{2}\sum_{\vec{\eta},\vec{\xi}}\sin\frac{1}{2}\vec{q}\cdot\vec{\xi}\,\sin\frac{1}{2}\vec{q}\cdot\vec{\eta}\,\frac{\vec{\eta}\cdot\vec{\xi}}{|\vec{\eta}\cdot\vec{\xi}|}$$

and $I_n(T)$ by

$$I_n(T)=(1/N)\sum_{\vec{q}} f(E(\vec{q}))\epsilon_n(\vec{q}) , \qquad \text{(A13)}$$

where $E(\vec{q})$ is given by (22).

According to the prescription given in the text, $\Sigma_2(\vec{q})$ is the non-Hartree-Fock part of (A7), which from (40) can be written in the form

$$\Sigma_1(\vec{q})=\tfrac{1}{4}I_1(T)\epsilon_1(\vec{q})+\tfrac{1}{3}xI_2(T)\epsilon_2(\vec{q}) . \qquad \text{(A14)}$$

Then

$$\Sigma_2(\vec{q};E(\vec{q}))\simeq(A_1-\tfrac{1}{4})I_1(T)\epsilon_1(\vec{q})+(A_2-\tfrac{1}{3}x)\times I_2(T)\epsilon_2(\vec{q})+\sum_{n=3}^{4}A_nI_n(T)\epsilon_n(\vec{q}). \qquad \text{(A15)}$$

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