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Effects of Anisotropy on Thermodynamic Properties of Antiferromagnets*

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In antiferromagnets, because of interplay with exchange energy, a small anisotropy can have a huge effect on thermodynamic properties. Detailed calculations are given, using spin-wave theory, of this effect on sublattice magnetization, specific heat, and parallel susceptibility of a cubic or uniaxial antiferromagnet. Specific heat data are discussed and experiments are suggested on the relatively large spin specific heats of antiferromagnets with low Curie points. The theory is extended to orthorhombic symmetry, and reasonable agreement found with existing experimental data on $CuCl_2 \cdot 2H_2O$.

I. INTRODUCTION

RECENTLY Anderson[1] has revived interest in the spin-wave theory of antiferromagnetism, first introduced in 1936 by Hulthén.[2] In this theory it is assumed that the antiferromagnetic ground state may be described in terms of a two-sublattice picture, the spins of one sublattice all pointing up, those of the other all pointing down. Since the actual ground state is a highly degenerate spin-zero state, it is by no means clear that a two-sublattice treatment is at all adequate. However, by a careful study of the zero-point spin-wave energy, Anderson has shown that the ground state of a three-dimensional antiferromagnet approaches closely the presumed two-sublattice arrangement. Several authors[3–7] have extended Anderson's theory to higher temperatures, and in particular Kubo[3,8] has presented a detailed discussion of the abnormal fluctuations inherent in the spin-wave approach.

Whether these fluctuations are real properties of antiferromagnets or are dissembled by the approximate nature of the theory is not clear at present. Real or fictitious, the fluctuations are subdued by the introduction of crystalline anisotropy into the theory; and probably all antiferromagnets have some anisotropy. Furthermore, neutron diffraction experiments[9] indicate that the spins of antiferromagnets are actually arrayed in sublattices, at least for times longer than 10^{-13} second.

The two most-studied single crystals, MnF_2 and $CuCl_2 \cdot 2H_2O$, are believed to have anisotropy energies of the order of one percent of their respective exchange energies. Other antiferromagnets seem to have similar anisotropies.[10] Thus the thermodynamic properties of antiferromagnets at temperatures low with respect to their Curie points should be accounted for by spin-wave theory, if this theory is at all meaningful.

In this paper we examine theoretically the expected temperature dependence of three measurable properties of antiferromagnets: (1) the magnetization of a sublattice, which may be detected most accurately by its effect on nuclear resonance;[11] (2) the spin-system specific heat, which swamps the lattice specific heat if the Curie point is well below the Debye temperature; and (3) the parallel susceptibility, or susceptibility of a single crystal in a magnetic field oriented along the preferred spin axis. We show that the presence of a small anisotropy, through a coupling of anisotropy and exchange energies, has a marked effect on these thermodynamic properties, and we consider this effect in some detail.

II. EFFECTS OF ANISOTROPY

The size of the anisotropy constant K is conveniently measured in terms of

$$\alpha = 2K/z|J|, \tag{1}$$

where $J/2$ is the exchange integral and z is the number of nearest neighbors.

Although α may be only a few percent, it appears in the spin-wave energy as $\alpha^{\frac{1}{2}}$. This tremendous enhancement of the effects of anisotropy comes about through a subtle coupling of anisotropy and exchange energies, the nature of which has been discussed in detail elsewhere.[7]

We define a temperature T_{AE} to measure this strong effect of anisotropy-exchange interplay:

$$kT_{AE} = z|J|S(2\alpha)^{\frac{1}{2}} = 2(z|J|K)^{\frac{1}{2}}S, \tag{2}$$

where S is the spin quantum number of a single atom.

The energy of a spin wave for small wave number $\mathbf{k}$ is

$$E_{\mathbf{k}} = z|J|S(b\mathbf{k}^2 + 2\alpha + \alpha^2)^{\frac{1}{2}}. \tag{3}$$

Here $\mathbf{k}$ is referred to the first Brillouin zone of the sublattice, and the constant b is a structure factor depending upon the type of lattice considered. Equation (3) is

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[1] P. W. Anderson, Phys. Rev. 86, 694 (1952).

[2] L. Hulthén, Proc. Roy. Acad. Sci. Amsterdam 39, 190 (1936).

[3] R. Kubo, Phys. Rev. 87, 568 (1952).

[4] T. Nakamura, Progr. Theoret. Phys. (Japan) 7, 539 (1952).

[5] J. R. Tessman, Phys. Rev. 88, 1132 (1952).

[6] J. M. Ziman, Proc. Phys. Soc. (London) A65, 540, 548 (1952); A66, 89 (1953).

[7] Keffer, Kaplan, and Yafet, Am. J. Phys. 21, 250 (1953).

[8] R. Kubo, Revs. Modern Phys. 25, 344 (1953).

[9] Shull, Strauser, and Wollan, Phys. Rev. 83, 333 (1951).

[10] See, for example, J. W. Stout and L. M. Matarrese, Revs. Modern Phys. 25, 338 (1953).

[11] N. J. Poulis and G. E. G. Hardeman, Physica 19, 391 (1953). Neutron diffraction may also be used to detect sublattice magnetization, but with less accuracy. See R. A. Erickson, Phys. Rev. 90, 779 (1953).

derived by Anderson[1] and by Kubo.[3] The total spin-wave energy is obtained by summing $E_{\mathbf{k}}$ over over Bose distributions in the (equivalent) first Brillouin zones of both sublattices. For small $\mathbf{k}$ this sum is approximated by an integral to infinity; and by proper normalization, the variable $b\mathbf{k}^2$ can be replaced by a single parameter λ^2. A similar approximation is used to obtain both the magnetization of a sublattice and the parallel susceptibility from the free energy. The procedure is given in detail in Kubo's 1952 paper,[3] which we use as a standard reference.

Thus Kubo finds for the temperature-dependent part of the total spin-wave energy of N spins:

$$E_T = NkTA_l\int_0^\infty \beta_\lambda(e^{\beta_\lambda}-1)^{-1}\lambda^{D-1}d\lambda, \qquad (4)$$

where

$$\beta_\lambda = (z|J|S/kT)(\lambda^2+2\alpha+\alpha^2)^{\frac{1}{2}}. \qquad (5)$$

Here A_l, the normalization factor discussed above, depends on the lattice structure; D is the dimensionality of the lattice, which we shall take as 3.

It should be noted that anisotropy is introduced into this theory as a crystalline field, that is, as involving the direction cosines between single spins and the crystal axes. A large part of the anisotropy, however, will originate in dipole-dipole and in anisotropic exchange forces, and hence involve the mutual orientation of pairs of spins. Ziman[6] has shown that such terms in the Hamiltonian lead to severe algebraic complications; and we shall not consider them in this paper. In making quantitative comparison of theory and experiment this limitation should be kept in mind.

Furthermore, we have restricted ourselves to calculations of sublattice magnetization and specific heat in the absence of an applied field.

A. Magnetization of a Sublattice

The temperature-dependent part of the sublattice magnetization, M_{ST}, in terms of the magnetization for complete alignment, M_∞, is given by:[3]

$$M_{ST} = -M_\infty(z|J|/kT)A(1+\alpha) \times\int_0^\infty \beta_\lambda^{-1}(e^{\beta_\lambda}-1)^{-1}\lambda^{D-1}d\lambda. \qquad (6)$$

We may evaluate Eq. (6) by expanding the Bose function,

$$(e^{\beta_\lambda}-1)^{-1} = \sum_{n=1}^\infty e^{-n\beta_\lambda},$$

and integrating term by term. Dropping α^2 as small compared to 2α, we make the substitution

$$(2\alpha)^{\frac{1}{2}}\cosh y = (\lambda^2+2\alpha)^{\frac{1}{2}}.$$

Thus, for $D=3$,

$$\frac{M_{ST}}{(M_\infty/S)} \approx -2\alpha A_l \sum_{n=1}^\infty \int_0^\infty \exp[-(nT_{AE}/T)\cosh y]\sinh^2 y\,dy = -2\alpha A_l \sum_{n=1}^\infty \frac{K_1(nT_{AE}/T)}{(nT_{AE}/T)}. \qquad (7)$$

Here K_1 is a Hankel function and the summation, which converges rapidly, is known as a Schlömilch series.

We now introduce the variable

$$\theta = kT/z|J|S \approx \tfrac{1}{3}(S+1)(T/T_c), \qquad (8)$$

where the exchange integral $J/2$ is approximated from Van Vleck's[12] molecular field theory of the Curie temperature, T_c. Equation (7) may be written

$$\frac{M_{ST}}{(M_\infty/S)} = -A_l\theta^2(\pi^2/6)M(T/T_{AE}), \qquad (9)$$

with

$$M(T/T_{AE}) = \frac{6}{\pi^2}\frac{T_{AE}}{T}\sum_{n=1}^\infty \frac{1}{n}K_1\left(\frac{nT_{AE}}{T}\right). \qquad (10)$$

Equation (9) is also given, but not plotted, in Kubo's paper;[3] we have derived it here to make clear some new relations which follow. In the absence of anisotropy $M(T/T_{AE})$ goes to 1, and Eq. (9) reduces to the T^2 law given by Kubo. The variation of $M(T/T_{AE})$ with T/T_{AE} is plotted as the dashed line in Fig. 1.

B. Specific Heat

By the same process which led from Eq. (6) to Eq. (7), we may bring Eq. (4) into the form (for $D=3$):

$$\frac{E_T}{Nz|J|S} = 4\alpha^2 A_l \sum_{n=1}^\infty \frac{d^2}{d(nT_{AE}/T)^2}\left\{\frac{K_1(nT_{AE}/T)}{(nT_{AE}/T)}\right\} = 4\alpha^2 A_l \sum_{n=1}^\infty \left\{\frac{K_3(nT_{AE}/T)}{(nT_{AE}/T)} - \frac{K_2(nT_{AE}/T)}{(nT_{AE}/T)^2}\right\}. \qquad (11)$$

The specific heat, C_T, is the derivative of (11) with respect to θ and has the value

$$C_T/Nk = A_l\theta^3(4\pi^4/15)C(T/T_{AE}), \qquad (12)$$

with

$$C(T/T_{AE}) = \frac{15}{8\pi^4}\left(\frac{T_{AE}}{T}\right)^4 \sum_{n=1}^\infty \{K_2(nT_{AE}/T) + K_4(nT_{AE}/T)\}. \qquad (13)$$

In the absence of anisotropy $C(T/T_{AE})$ goes to 1, and Eq. (12) reduces to the T^3 law given by Kubo.[3] The variation of $C(T/T_{AE})$ with T/T_{AE} is plotted as the solid line in Fig. 1.

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[12] J. H. Van Vleck, J. Chem. Phys. 9, 85 (1941).

C. Parallel Susceptibility

In calculating the parallel susceptibility $\chi_{\parallel}$ one must restrict the applied field H_0 to values less than $kT_{AE}/g\mu_0$. Otherwise the spins can lower their free energy by flopping over to the hard axis, and one will find oneself confronted with $\chi_{\perp}$.

As shown by Kubo[3]

$$\chi_{\parallel}=(Ng^2\mu_0{}^2/4kT)A_l\int_0^{\infty}(\coth^2\tfrac{1}{2}\beta_{\lambda}-1)\lambda^{D-1}d\lambda. \quad (14)$$

The integrand is equal to

$$\coth^2\tfrac{1}{2}\beta_{\lambda}-1=\frac{4e^{\beta\lambda}}{(e^{\beta\lambda}-1)^2}=-4\frac{\partial}{\partial(zJS/kT)}\times[(e^{\beta\lambda}-1)^{-1}(\lambda^2+2\alpha+\alpha^2)^{-\frac{1}{2}}],$$

and comparison with Eq. (6) shows that

$$\frac{\chi_{\parallel}}{(Ng^2\mu_0{}^2/kT)}=\frac{1}{(1+\alpha)}\frac{\partial}{\partial(zJS/kT)}\left(\frac{M_{ST}}{M_{\infty}/S}\right)=\frac{(2\alpha)^{\frac{1}{2}}}{(1+\alpha)}\frac{\partial}{\partial(T_{AE}/T)}\left(\frac{M_{ST}}{M_{\infty}/S}\right). \quad (15)$$

Applying the above to Eq. (9) we find

$$\frac{\chi_{\parallel}}{(Ng^2\mu_0{}^2/kT)}=A_l\frac{\pi^2}{3}\theta^3\chi\left(\frac{T}{T_{AE}}\right), \quad (16)$$

with

$$\chi\left(\frac{T}{T_{AE}}\right)=\frac{3}{\pi^2}\left(\frac{T_{AE}}{T}\right)^2\sum_{n=1}^{\infty}K_2\left(\frac{nT_{AE}}{T}\right). \quad (17)$$

In the absence of anisotropy $\chi(T/T_{AE})$ goes to 1, and Eq. (16) reduces to the T^2 law given by Kubo. The variation of $\chi(T/T_{AE})$ with (T/T_{AE}) is plotted as the dotted line in Fig. 1. For temperatures well below T_{AE} the susceptibility rises exponentially, a result obtained by Tessman.[5]

Equation (15) gives a general relation between the parallel susceptibility and the magnetization of a sublattice. It is interesting to compare this with the general relation given by Van Vleck's molecular field theory:[12]

$$\frac{\chi_{\parallel}}{(Ng^2\mu_0{}^2/kT)}=\frac{S^2B_S'(y_0)}{1+(S/\theta)B_S'(y_0)}. \quad (18)$$

Here, for comparison, the Brillouin function represents

$$SB_S(y_0)=(M_{S0}+M_{ST})/(M_{\infty}/S),$$

with

$$y_0=SB_S/\theta.$$

Thus, Van Vleck's relation may be brought into the form

$$\frac{\chi_{\parallel}}{(Ng^2\mu_0{}^2/kT)}=\frac{[\partial/\partial(1/\theta)][SM_{ST}/M_{\infty}]}{\{[(M_{S0}+M_{ST})/M_{\infty}]+(2/S\theta)[\partial/\partial(1/\theta)][SM_{ST}/M_{\infty}]\}}. \quad (18')$$

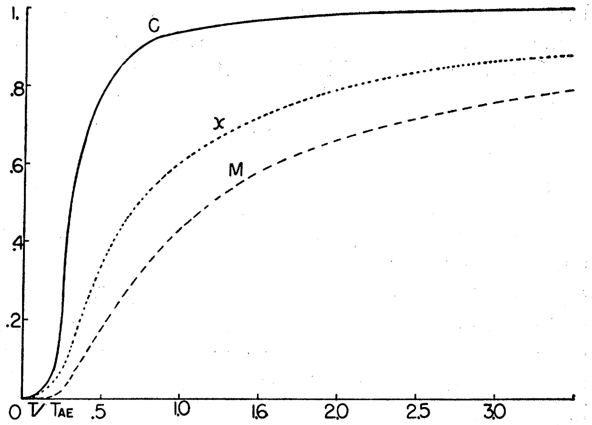

FIG. 1. The temperature variation of the three thermodynamic quantities for cubic or uniaxial symmetry showing the effect of anisotropy. The solid line is the specific heat $C(T/T_{AE})$, the dashed line is the magnetization $M(T/T_{AE})$, and the dotted line is the susceptibility $\chi(T/T_{AE})$.

At low temperatures (small θ) the denominator of Eq. (18′) is approximately 1 and, in the absence of anisotropy, Van Vleck's relation agrees with the spin-wave Eq. (15). (The correction for anisotropy is negligible.) Since the molecular field approximation is more accurate near the Curie point, Eq. (18′) might be valid at all temperatures.

III. DISCUSSION OF SPECIFIC HEATS

Postponing until the next section the interesting case of $CuCl_2\cdot 2H_2O$, we discuss here the specific heats of a few antiferromagnetic salts. First, the condition under which the spin-wave specific heat will be larger than the lattice specific heat should be pointed out. In the usual Debye approximation the lattice specific heat at low temperature is given by

$$C_L/N'k=(12/5)\pi^4(T/\Theta)^3, \quad (19)$$

where Θ is the Debye temperature. Comparison with Eq. (12) yields

$$C_T/C_L=(A_l/243)(S+1)^3(\Theta/T_c)^3\times C(T/T_{AE})(N/N'). \quad (20)$$

Here N/N' is the ratio of paramagnetic ions to total number of ions.

In Table I we apply Eq. (20) to a few antiferromagnetic salts with low Curie points. It is seen that, as a general rule of thumb, the spin-wave contribution to the specific heat will be larger than that of the lattice if

$$T_c<\sim\Theta/5. \quad (21)$$

However, in the region $T<T_{AE}$ the spin-wave specific heat will be sharply reduced by $C(T/T_{AE})$ [see Fig. 1]. Values of T_{AE} simply are not known at present. The value given in Table I for MnF_2 is a theoretical estimate[13] based chiefly on magnetic-dipole anisotropy.

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[13] F. Keffer, Phys. Rev. 88, 608 (1952).

Presumably T_{AE} of the other fluorides may go even higher than 15°K.[10] The two values given for $CuCl_2 \cdot 2H_2O$ will be discussed in the next section.

Stout and his collaborators have measured the specific heats of the fluorides in the vicinities of their Curie points.[14,15] Their published curves show the following temperature dependences for spin-plus-lattice specific heats just below the Curie points: MnF_2, $T^{1.7}$; FeF_2, T^2; CoF_2, $T^{2.9}$; NiF_2, T^3. Westrum and his collaborators[16,17] find a temperature dependence of $T^{3.2}$ for NpO_2 in the region between 15 and 24°K. Admittedly all these measurements are in a region where spin-wave theory is inapplicable; we quote them to indicate the general trend of specific heats and to emphasize the importance of obtaining measurements at lower temperatures. The measurements on MnF_2 were carried down to 15°K (vicinity of T_{AE}) with no appreciable change in the curious $T^{1.7}$ dependence. However, NpO_2 in the region between 15 and 11°K shows a marked drop in specific heat, perhaps indicating effects of anisotropy.

Mention should be made of $FeCl_2$, $CoCl_2$, and particularly $MnCl_2$ which have very low Curie points and should have large spin specific heats.

IV. ORTHORHOMBIC SYMMETRY

In an orthorhombic crystal, taking the z axis as preferred, the anisotropy is different for displacements towards the x and the y axes. As a measure of these anisotropies we introduce $\alpha(1)$ and $\alpha(2)$, to which correspond the two temperatures $T_{AE}(1)$ and $T_{AE}(2)$. The spin waves split into two sets, with approximate energies

$$E_{\mathbf{k}}(i) \cong z|J|S[b\mathbf{k}^2+2\alpha(i)]^{\frac{1}{2}}, \tag{22}$$

as shown by Nakamura.[4]

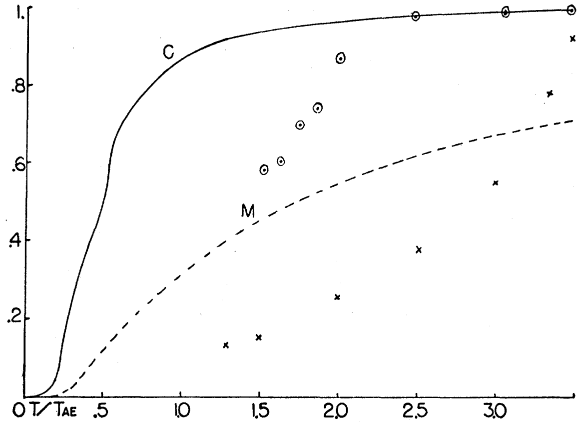

FIG. 2. The temperature variation of the two thermodynamic quantities for orthorhombic symmetry showing the effect of anisotropy. The solid line is the specific heat $C(T/1)+C(T/2)$, and the circles are the experimental points (see reference 19). The dashed line is the magnetization $M(T/1)+M(T/2)$, and the crosses are the experimental points (see reference 11).

The magnetization of a sublattice is obtained by a summation over all spin waves, and we now merely sum over the two independent sets. Thus

$$\frac{M_{ST}}{(M_\infty/S)} = -\tfrac{1}{2}A_l\theta^2\left(\frac{\pi^2}{6}\right) \times\left[M\left(\frac{T}{T_{AE}(1)}\right)+M\left(\frac{T}{T_{AE}(2)}\right)\right]. \tag{23}$$

Similarly, the specific heat is given by

$$\frac{C_T}{Nk} = \tfrac{1}{2}A_l\theta^3\left(\frac{4\pi^4}{15}\right)\left[C\left(\frac{T}{T_{AE}(1)}\right)+C\left(\frac{T}{T_{AE}(2)}\right)\right]. \tag{24}$$

One might expect the parallel susceptibility to be obtained on applying Eq. (15) to Eq. (23), but it turns out to be a much more complicated expression. Introducing

$$\epsilon = g\mu_0H_0/z|J|S \tag{25}$$

as a measure of the applied field H_0, we may distinguish two cases:

$$(A) \quad \epsilon^2>\alpha(2)-\alpha(1).$$

Here the applied field is large compared to the difference between the two anisotropy-exchange fields, and hence the normal modes of the spin waves are essentially determined by the applied field. The two sets of spin waves may be thought of as precessing, respectively, clockwise and counterclockwise about H_0. The energies of the spin waves are given approximately by[4]

$$E_{\mathbf{k}} \cong z|J|S\{[b\mathbf{k}^2+\alpha(1)+\alpha(2)]^{\frac{1}{2}}\pm\epsilon\}. \tag{26}$$

Accordingly, $\chi_{\parallel}$ will be given by Eq. (16) with T_{AE} replaced by $\tfrac{1}{2}[T_{AE}(1)+T_{AE}(2)]$.

$$(B) \quad E^2<\alpha(2)-\alpha(1)$$

Here the normal modes are essentially determined by the two widely different anisotropy-exchange fields. The spin-wave energies are given by adding terms in ϵ^2 to Eq. (22). The parallel susceptibility is field dependent in a complicated way. In the absence of a more precise introduction of anisotropy terms (especially since we are here dealing with a large difference in anisotropies), it hardly seems worthwhile to go further.

A remarkable series of measurements has recently been made at Leiden on single crystals of the orthorhombic $CuCl_2 \cdot 2H_2O$. From susceptibility measurements[18] it is possible to deduce values of $T_{AE}(1)$ and $T_{AE}(2)$. The smaller of these may be found from the critical field, that is, from the value of H_0 which is suf-

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[14] J. W. Stout and H. E. Adams, J. Am. Chem. Soc. 64, 1535 (1942).

[15] J. W. Stout and E. Catalano, Phys. Rev. 92, 1575 (1953).

[16] Westrum, Hatcher, and Osborne, J. Chem. Phys. 21, 419 (1953).

[17] D. W. Osborne and E. F. Westrum, Jr., J. Chem. Phys. 21, 1884 (1953).

[18] Van den Handel, Gijsman, and Poulis, Physica 18, 862 (1952).

ficient to overcome the anisotropy-exchange energy holding the spins to the preferred axis and to allow them to flop to the second-preferred xais. This is detected by an immediate jump of susceptibility from $\chi_{\parallel}$ to $\chi_{\perp}$. Inserting the measured value of 6500 oersteds for the critical field into

$$kT_{AE}(1)=g\mu_0(H_0)_{\text{crit}}, \tag{27}$$

we obtain $T_{AE}(1)\approx 1°\text{K}$.

There exists a critical field for flop from the preferred to the third-preferred axis, and $T_{AE}(2)$ is given by this field. Since the spins will have flopped to the second-preferred axis before this field is reached, it cannot be measured directly, but must be inferred from measurement of the flop from the second- to the third-preferred axis when H_0 is along the second axis. This flop takes place at 10 690 oersteds. Now from Eq. (2) we see that the anisotropy is proportional to the square of the critical field, and since to first order the anisotropy constant for first- to third-preferred axis is the sum of the constants first- to second-preferred and second- to third-preferred, the critical fields will sum in the square. This yields a field of 12 500 oersteds corresponding to $T_{AE}(2)\approx 2°\text{K}$.

The Curie point of $CuCl_2\cdot 2H_2O$ is 4.3°K; thus the values of T_{AE} are very large, and effects of anisotropy will be strong. In Fig. 2 we have plotted $M(T/1)+M(T/2)$ as the dashed line and $C(T/1)+C(T/2)$ as the solid line. Experimental measurements of Poulis and Hardeman[11] on sublattice magnetization and of Friedberg[19] on specific heat are indicated; in plotting these points we have used the value $5/(2\pi^2)$ for the geometrical factor A_l, and we have evaluated J from molecular field theory, Eq. (8). Slight changes in J move all the points large distances up or down, but have little effect on the shape of the curves. None of the data are at sufficiently low temperatures for one to expect the spin-wave theory to be valid; nevertheless the general trend of things is encouraging. It is interesting to note the much stronger effect of anisotropy on sublattice magnetization than on specific heat.

Poulis and Hardeman[11] find that the sublattice magnetization closely follows a T^4 law over most of their range. We are unable to account for any such simple behavior.

TABLE I. Application of Eq. (20) to some representative antiferromagnetic salts.

Salt	Θ (°K) approx.	T_c (°K)	T_{AE} (°K)	S	N/N'	C_T/C_L from Eq. (20) (for $T>T_{AE}$)
MnF_2	450	70	15	5/2	1/3	7
FeF_2	420	78		2	1/3	3
CoF_2	360?	37		3/2	1/3	8
NiF_2	450	73		1	1/3	1
NpO_2	360	25		3/2?	1/3	10
$CuCl_2\cdot 2H_2O$	250	4.2	1 and 2	1/2	1/9	70

The anisotropy in $CuCl_2\cdot 2H_2O$ is due partly to anisotropic exchange, partly to magnetic dipole interactions, and partly to the anisotropy of the g factor (crystalline fields).[20] As noted in Sec. II, the theory used in this paper assumes the anisotropy can be represented as a sum of terms involving single spins only. Some of the detailed behavior of the thermodynamic properties may be lost in this simplification. However, since we have used the experimentally determined critical fields in the above, and these represent a sort of activation energy for spin waves, we doubt that a more involved treatment would change things significantly. We should note that one can obtain the critical fields from antiferromagnetic resonance frequencies, that is, from the activation energies of $\mathbf{k}=0$ spin waves. Using the data of Ubbink *et al.*,[21] Nagamiya[22] has deduced fields of 7200 and 13 000 oersteds, in good agreement with the susceptibility determination.

On the other hand the detailed nature of the anisotropy may be of considerable importance in the theory of parallel susceptibility, especially for applied fields of the same order of magnitude as the critical fields. The measurements[18] are in fields of this size and furthermore are all at 1.6°K or higher. Because of this, and since the susceptibility is field-dependent in such a complicated way, we shall drop the problem at this point.

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[19] S. A. Friedberg, Physica **18**, 714 (1952).

[20] T. Moriya and K. Yosida, Progr. Theoret. Phys. (Japan) **9**, 663 (1953). See also discussion remark of F. Keffer, Revs. Modern Phys. **25**, 337 (1953).

[21] Ubbink, Poulis, Gerritsen, and Gorter, Physica **18**, 361 (1952).

[22] T. Nagamiya (private communication).