the sc and bcc spectra could be followed as they respectively change to those of the fcc and sc under the application of second-neighbor interactions. Anisotropic effects would produce more singularities, e.g., by considering a dispersion relation of the form $\alpha(1-\cos x)+\beta(1-\cos y)+\gamma(1-\cos z)$ on a sc lattice.

While other methods might be used to obtain more precise spectra, the method used is very simple and quite sufficient for the investigations undertaken and has resulted in several new results. Computational facilities suitable for performing these calculations are readily available.

The understanding now reached for a single band can be of considerable help in appreciating complicated density-of-states spectra where branches may cross and bands overlap. Uses of the density-of-states spectra are so diverse that it would be difficult to survey the applications that might be made of our results.

A case that immediately comes to mind is that of spin waves in[13] EuS which has spins of magnitude $\frac{7}{2}$ on an fcc magnetic lattice, nn ferromagnetic exchange and suggested antiferromagnetic-second-neighbor exchange of strength to give $\eta \sim -\frac{1}{3}$. Somewhat differently, a forthcoming paper by Thorpe[20] on two-magnon bound states in EuO gives another possible application. That work suggests an experimental observation on the basis of the logarithmic singularity of the nn case, Fig. 2(b) part (v). It is not clear to what extent that behavior is essential for the prediction but our study of second-neighbor effects has obvious relevance to any real substance.

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Magnon-Photon Coupling in Antiferromagnets

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The coupling of photons with long-wavelength magnons in antiferromagnetic MnF_2 is discussed. The consequences of this coupling for reflection spectra are pointed out, and numerical results for the dispersion curves of the coupled modes are presented. The discussion is briefly generalized to more complicated situations.

It is now well known both from experiments and theory that phonons and photons can couple in ionic crystals to form coupled modes, namely, polaritons. An analogous coupling between magnons and photons can occur in magnetically ordered crystals, and while the possibility of such a coupling in ferromagnets has been discussed by several authors,[1-8] the more interesting case of antiferromagnets seems to have been generally ignored. In this paper we investigate the latter problem with specific reference to MnF_2. The results obtained are then briefly generalized.

The technique we shall be employing is formally similar to that previously used in connection with the spin-resonance problem.[3,5] Indeed, as will be seen later, the results obtained also have much in common. There is, however, an important conceptual difference between the spin-resonance problem and the one presently under discussion. In the former, one studies the spin motions in the presence of *external* static and rf fields. Here, on the other hand, we shall not be concerned with externally imposed fields. Rather, we shall examine the effect on the spin waves of the photon field associated with the *spin fluctuations themselves*. This interaction is usually ignored in the study of spin waves, and this is justified except when the magnon wave vector becomes very small. In that long-wavelength situation this interaction plays a significant role, and may be viewed as a magnon-photon coupling analogous to the phonon-photon coupling that one considers in ionic crystals to allow for the effects of retardation on the phonon spectrum. Our aim in this paper is to study this magnon-photon interaction in MnF_2 and to point out some of its consequences.

MnF_2 has the rutile structure and becomes antiferromagnetically ordered below 67 °K, with the spins of the Mn^{++} ions oriented parallel to the c axis.[9] The spin-wave spectrum has been thoroughly investigated, the long-wavelength modes by optical and microwave techniques,[10,11] and the short-wavelength modes by slow-neutron scattering.[12] For purposes of discussing the magnon-photon coupling we need consider only the long-wavelength part of the magnon spectrum. Let us first deduce the relevant equations from the conventional spin-wave equations, which do not allow for the effect discussed above. This deficiency will subsequently be corrected. The starting point for deriving the spin-wave equations is the Hamiltonian[13]

$$\mathcal{H} = -J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j - H_A g \mu_B \left(\sum_i S_{iz} - \sum_j S_{jz}\right). \quad (1)$$

Here J is the nearest-neighbor exchange integral,[14] μ_B the Bohr magneton, and g the gyromagnetic ratio. H_A denotes the anisotropic field and is directed along the z axis taken parallel to the c direction. The subscripts i and j label, respectively, the spins on the two sublattices. Thus $\sum_i S_{iz}$ indicates a summation of the z component of the spin operators pertaining to sublattice 1; a similar meaning applies to $\sum_j S_{jz}$. Furthermore, in the first term, $\langle ij \rangle$ implies that, corresponding to any given S_i on sublattice 1, the summation over j is restricted to nearest neighbors.

Starting from (1) it is straightforward to obtain the following equation of motion for the spins on the first sublattice:

$$\dot{\vec{S}} = \vec{S}_i \times \left(J \sum_j \vec{S}_j + g\mu_B \vec{H}_A\right) , \quad (2)$$

the summation over j being restricted as usual to the first neighbors of $\vec{S}_i$. Upon multiplying (2) by $g\mu_B$ we get

$$\dot{\vec{\mu}}_i = \vec{\mu}_i \times \left(J \sum_j \vec{S}_j + g\mu_B \vec{H}_A\right) , \quad (3)$$

which is the equation of motion for the magnetic moment $\vec{\mu}_i$ at the site i in the first sublattice. At this point we remind ourselves that the spin fluctuations associated with spin waves are of small amplitude, and are essentially confined to the x and y components, the z component remaining fixed. This amounts to setting

$$\dot{S}_{iz} = 0, \quad \dot{\mu}_{iz} = 0 , \quad (4)$$

and is in the spirit of a classical treatment of the spin-wave problem. We now make the further assumption that the spins in each sublattice are precessing around the z axis with very nearly the same phase. This allows us to replace each $\vec{S}_i$ and $\vec{S}_j$ by their local spatial averages $\langle \vec{S}_i \rangle$ and $\langle \vec{S}_j \rangle$, which vary only slowly in space. In effect this implies that we are considering a long-wavelength mode of spin fluctuations.

Now we write

$$\tfrac{1}{2} g \mu_B \langle \vec{S}_i \rangle = \tfrac{1}{2} \langle \vec{\mu}_i \rangle = \vec{M}_1 \Omega , \quad (5)$$

where $\Omega = V/N$, with V being the volume of the crystal and N the number of magnetic ions in it. It is clear that $\vec{M}_1$, defined above, denotes the magnetization associated with the first sublattice. In particular, M_{1z} denotes the saturation magnetization, while M_{1x} and M_{1y} represent the fluctuations about the equilibrium value. Using (5) and (4) in (3) we obtain

$$\dot{\vec{M}}_1 = \vec{M}_1 \times \left(J \sum_j \langle \vec{S}_j \rangle + g\mu_B \vec{H}_A\right)$$

$$= g\mu_B \vec{M}_1 \times \left(\frac{J\Omega n}{(g\mu_B)^2} \vec{M}_2 + \vec{H}_A \right) , \quad (6)$$

where n is the number of nearest neighbors and $\vec{M}_2$ is defined similar to Eq. (5). It is to be remembered of course that $\dot{M}_{1z} = \dot{M}_{2z} = 0$. Introducing the notations

$$g\mu_B = \gamma$$

and

$$J\Omega n/(g\mu_B)^2 = \lambda , \quad (7)$$

Eq. (6) becomes

$$\dot{\vec{M}}_1 = \gamma \vec{M}_1 \times (\lambda \vec{M}_2 + \vec{H}_A) . \quad (8)$$

Remembering that $\lambda M_{2z} = H_E$ is the exchange field acting on sublattice 1 due to the magnetization of

sublattice 2, we may rewrite (8) in component form as

$$\dot{M}_{1x} = \gamma[M_{1y}(H_E + H_A) - \lambda M_{1z} M_{2y}] \ , \tag{9a}$$

$$\dot{M}_{1y} = \gamma[M_{1z}(-\lambda M_{2x}) - M_{1x}(H_E + H_A)] \ , \tag{9b}$$

$$\dot{M}_{1z} = 0. \tag{9c}$$

A similar set of equations may be written for the second sublattice.

As previously remarked, Eqs. (9) (as also the corresponding set of equations for the second sublattice) do not give a proper representation of the spin motions in the long-wavelength limit since they do not allow for the coupling with the photon field associated with the spin motions. These equations are analogous to the long-wavelength lattice dynamical equations of ionic crystals obtained by ignoring the effects of electric fields generated by the vibrations themselves. The deficiency noted above concerning the lack of interaction with the photon field is, however, readily corrected by slightly modifying Eqs. (9) to accommodate the effects of the fluctuating field $\vec{H}$ associated with the spin motions, and by further requiring $\vec{H}$ to obey Maxwell's equations. Formally, the interaction between the spin motions and the photons could be described by adding a term $-\mu_B g(\sum_i \vec{S}_i + \sum_j \vec{S}_j) \cdot \vec{H}$ to the Hamiltonian. Thus, for a proper discussion of the long-wavelength motions, we must really consider the equations

$$\dot{M}_{1x} = \gamma[M_{1y}(H_E + H_A) - M_{1z}(H_y + \lambda M_{2y})] \ , \tag{10a}$$

$$\dot{M}_{1y} = \gamma[M_{1z}(H_x - \lambda M_{2x}) - M_{1x}(H_E + H_A)] \ , \tag{10b}$$

$$\dot{M}_{1z} = 0, \tag{10c}$$

$$\dot{M}_{2x} = \gamma[M_{2y}(-H_E - H_A) - M_{2z}(H_y - \lambda M_{1y})] \ , \tag{11a}$$

$$\dot{M}_{2y} = \gamma[M_{2z}(H_x - M_{1x}) - M_{2x}(-H_E - H_A)] \ , \tag{11b}$$

$$\dot{M}_{2z} = 0 \ , \tag{11c}$$

supplemented, of course, by Maxwell's equations. A consistent solution of all these equations will provide a proper answer to the long-wavelength magnon problem. In writing these equations we have taken note of the fact that like the spin fluctuations, $\vec{H}$ has only x and y components.

It is pertinent to remark here that Eqs. (10) and (11) are similar to those usually employed in the discussion of antiferromagnetic resonance (AFMR), [15,16] the difference being that, whereas in the AFMR problem $\vec{H}$ is the oscillatory field applied externally, here $\vec{H}$ represents the spontaneously generated field associated with the spin-wave motions.

Presently we wish to solve Eqs. (10) and (11) in conjunction with Maxwell's equations. Formally, this is similar to the approach of Pincus[3] and of Kittel, [5] who had earlier solved the spin-resonance equations for ferromagnets in conjunction with Maxwell's equations in order to delineate certain aspects of the resonance problem. Let us consider as solutions, plane waves of the form

$$\begin{pmatrix} M_{1x} \\ M_{1y} \\ M_{2x} \\ M_{2y} \\ H_x \\ H_y \end{pmatrix} = \begin{pmatrix} M_{0,1x}(k,\omega) \\ M_{0,1y}(k,\omega) \\ M_{0,2x}(k,\omega) \\ M_{0,2y}(k,\omega) \\ H_{0,x}(k,\omega) \\ H_{0,y}(k,\omega) \end{pmatrix} e^{i(\vec{k}\cdot\vec{r} - \omega t)} \ . \tag{12}$$

Using these in (10) and (11) it is easy to show that

$$M_{0x}(k,\omega) = M_{0,1x}(k,\omega) + M_{0,2x}(k,\omega)$$

$$= \frac{2\gamma^2 M_s H_A}{\omega_1^2 - \omega^2} \ H_{0x}(k,\omega) \ , \tag{13a}$$

$$M_{0y}(k,\omega) = M_{0,1y}(k,\omega) + M_{0,2y}(k,\omega)$$

$$= \frac{2\gamma^2 M_s H_A}{\omega_1^2 - \omega^2} \ H_{0y}(k,\omega) \ , \tag{13b}$$

where

$$M_s = M_{1z} = |M_{2z}|$$

denotes the saturation magnetization and

$$\omega_1 = \gamma[H_A(H_A + 2H_E)]^{1/2} \tag{14}$$

is the AFMR frequency. Note that if $\vec{H}$ were an applied oscillatory field, then (13a) and (13b) would define the transverse susceptibility $\chi_\perp(\omega)$, i.e.,

$$\chi_\perp(\omega) = \frac{2\gamma^2 M_s H_A}{\omega_1^2 - \omega^2} \ . \tag{15a}$$

Furthermore, since $M_z = M_{1z} + M_{2z} = 0$, we have the longitudinal susceptibility

$$\chi_\parallel(\omega) = 0 \ . \tag{15b}$$

Equations (13a) and (13b) show that the fluctuation magnetization is related to the fluctuating field by the transverse susceptibility.

Next we observe that from Maxwell's equations we may write

$$\frac{1}{c^2} \ddot{\vec{H}} - \nabla^2 \vec{H} = 4\pi \, \mathrm{grad} \, \mathrm{div} \, \vec{M} - \frac{1}{c^2} \ddot{\vec{M}} \ . \tag{16}$$

Substitution of the trial solution (12) leads to

$$\vec{H} = -4\pi \frac{[\vec{k}(\vec{k}\cdot\vec{M}) - (\omega^2/c^2)\vec{M}]}{(k^2 - \omega^2/c^2)} \ . \tag{17}$$

Combining (17) with (13), we obtain, using the notation (15), the equations

$$[4\pi\chi_\perp(\omega)(c^2k_x^2-\omega^2)+(c^2k^2-\omega^2)]M_{0x}+4\pi\chi_\perp(\omega)c^2k_xk_yM_{0y}=0\,, \tag{18a}$$

$$4\pi\chi_\perp(\omega)c^2k_xk_yM_{0x}+[4\pi\chi_\perp(\omega)(c^2k_y^2-\omega^2)+(c^2k^2-\omega^2)]M_{0y}=0\,. \tag{18b}$$

The requirement of nontrivial solutions for M_{0x} and M_{0y} then leads to the equation

$$\begin{vmatrix} 4\pi\chi_\perp(\omega)(c^2k_x^2-\omega^2)+(c^2k^2-\omega^2) & 4\pi\chi_\perp(\omega)c^2k_xk_y \\ 4\pi\chi_\perp(\omega)c^2k_xk_y & 4\pi\chi_\perp(\omega)(c^2k_y^2-\omega^2)+(c^2k^2-\omega^2) \end{vmatrix} = 0\,. \tag{19}$$

The roots of this determinantal equation yield the dispersion for the coupled magnon-photon modes or what may be termed the *magniton* modes, propagating in the direction $\hat{k}(=\vec{k}/k)$.

We shall now discuss the solutions of (19) for $\hat{k}$ along various directions. First, choosing $\hat{k}$ to be along the z axis, we obtain

$$\frac{k^2c^2}{\omega^2}=1+4\pi\chi_\perp(\omega)=\frac{\omega_2^2-\omega^2}{\omega_1^2-\omega^2}\,, \tag{20}$$

where

$$\omega_2^2=\omega_1^2+8\pi\gamma^2M_sH_A\,. \tag{21}$$

The dispersion curves appropriate to (20) are sketched in Fig. 1, where clearly each branch is doubly degenerate. The photonlike and the magnonlike parts of the dispersion curves are easily identified. We note that consequent on the magnon-photon coupling, there is a frequency gap ranging from ω_1 to ω_2. Furthermore, from (21) we have

$$\begin{aligned}\frac{\omega_2^2}{\omega_1^2}&=1+\frac{8\pi\gamma^2M_sH_A}{\omega_1^2}\\&=1+4\pi\chi_\perp(0)\\&=\mu_\perp(0)\,,\end{aligned} \tag{22}$$

where $\mu_\perp(0)$ denotes the static magnetic permeability. This result is analogous to the Lyddane-Sachs-Teller (LST) relation familiar in connection with long-wavelength vibrations in diatomic cubic crystals.[17] In fact, just as the longitudinal-optic frequency ω_{LO} occurring in the LST relation corresponds to a mode for which $\vec{D}=0$, we have that ω_2 is the frequency of a mode for which $\vec{B}=0$. It must be mentioned that the appearance of a gap at $k=0$ and the LST-type relation involving the limiting frequencies have previously been noted by Pincus[3] and Kittel[5] in their studies of ferromagnets.

Next let us consider $\hat{k}$ parallel to the x axis. Here we have several solutions, namely,

$$\omega=ck\,, \tag{23a}$$

$$1+4\pi\chi_\perp(\omega)=0,\quad \text{i.e.,}\quad \omega=\omega_2\,, \tag{23b}$$

and

$$\frac{k^2c^2}{\omega^2}=\frac{\omega_2^2-\omega^2}{\omega_1^2-\omega^2}\,. \tag{23c}$$

The dispersion curves appropriate to these are sketched in Fig. 2. We observe that, in contrast to Fig. 1, only one of the two magnon branches is affected. The remaining photon and magnon modes are unmixed. However, this unmixed magnon branch, though dispersionless, appears with an enhanced frequency ω_2 instead of ω_1.

The situation corresponding to $\hat{k}$ lying in the xz plane and making an arbitrary angle θ with the z axis is represented by the equations

$$\frac{k^2c^2}{\omega^2}=\frac{\omega_2^2-\omega^2}{\omega_1^2-\omega^2}\,, \tag{24a}$$

$$\omega^2=\tfrac{1}{2}\{\omega_2^2+k^2c^2\pm[(\omega_2^2+k^2c^2)^2-4k^2c^2(\omega_2^2\sin^2\theta+\omega_1^2\cos^2\theta)]^{1/2}\}\,. \tag{24b}$$

The dispersion curves for $\theta=45^\circ$ are sketched in

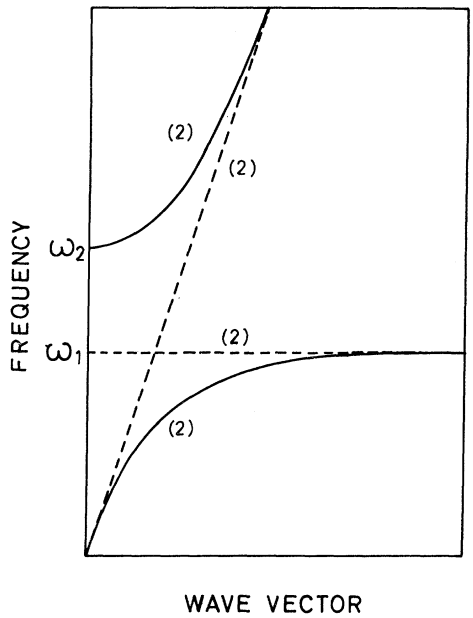

FIG. 1. Schematic drawing of the dispersion relations for coupled magnon-photon modes in MnF_2 for $\hat{k}$ parallel to the z axis. The dashed lines indicate the uncoupled modes, while the numbers in parentheses denote the degeneracies. ω_1 is the AFMR frequency.

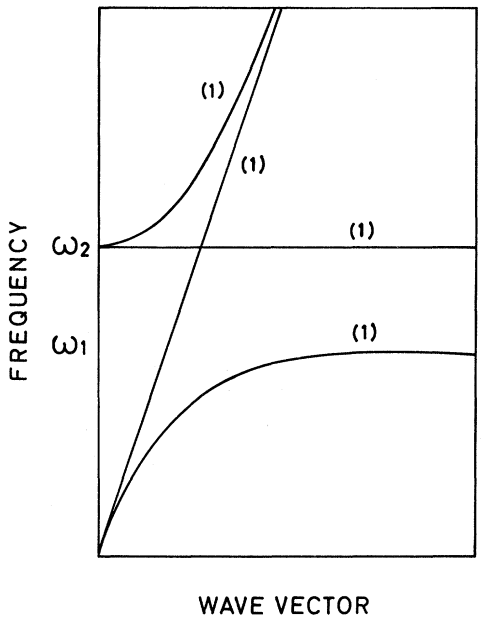

FIG. 2. Sketch of the dispersion curves for $\hat{k}$ parallel to the x axis. As compared to the previous figure, only one of the two magnon branches is coupled to the photon. The remaining magnon and photon branches are uncoupled, although the frequency of the magnon branch is enhanced.

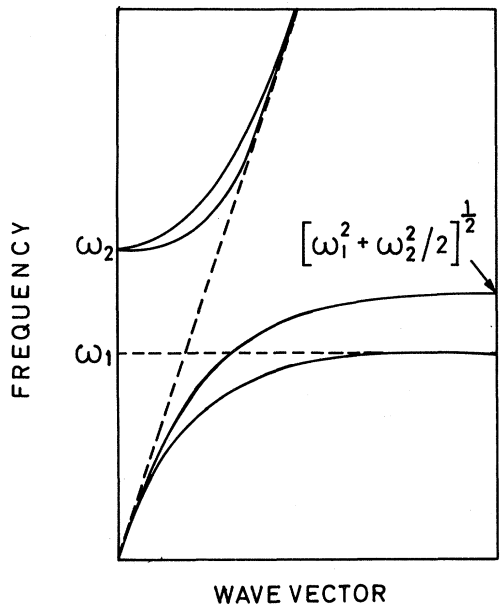

FIG. 3. Curves similar to the previous figures but for $\hat{k}$ making an angle of 45° with the z axis. One set of branches exhibits the same behavior as the corresponding set of the first two figures; the other set shows an intermediate behavior.

Fig. 3. Taking all three figures together, we note that whereas one set of branches remains unchanged as θ varies from 0° to 90°, the other set goes from a mixed to an unmixed state. This anisotropy arises specifically from the grad div$\vec{M}$ term in Eq. (16), and was not considered in earlier discussion of similar problems pertaining to ferromagnets.[3,5] It is also interesting to observe in passing that there is an almost exact parallelism between the problem under discussion and the formation of polaritons from the doubly degenerate infrared-active vibrational modes in uniaxial crystals of the ZnO type.[18]

There is one other feature common to polariton and magniton spectra which arises from the frequency gap mentioned earlier. Consider the reflection of electromagnetic radiation of frequency ω incident normally on a semi-infinite specimen having its c axis normal to the face. The reflectivity $R(\omega)$ is given by[19]

$$R(\omega)=\left|\frac{1-n(\omega)}{1+n(\omega)}\right|^2 , \tag{25}$$

where $n(\omega)$ is the refractive index appropriate to wave propagation along the c axis, and is itself given by[19]

$$n(\omega)=[\epsilon_\perp(\omega)\mu_\perp(\omega)]^{1/2} , \tag{26}$$

TABLE I. Parameters used in calculation of magnon-photon coupling in MnF_2. These are theoretical values (Ref. 20), and yield a value of 9.10 cm^{-1} for ω_1, which is in good agreement with experiments (Refs. 10 and 11).

H_E	5.4×10^5 Oe
H_A	8.8×10^3 Oe
M_S	590 G

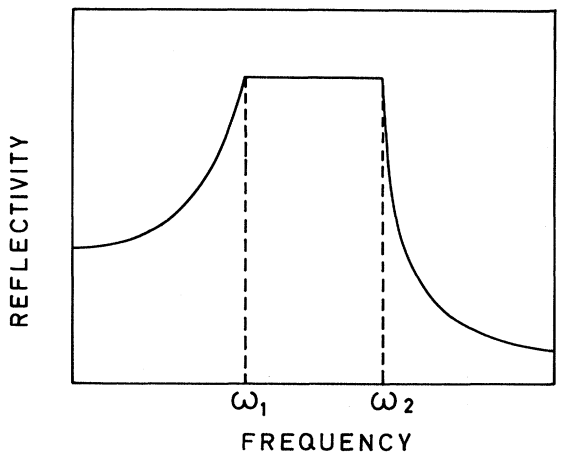

FIG. 4. Reflectivity spectrum $R(\omega)$ for light incident normally on a thick flat crystal having the c axis perpendicular to its face. The dielectric susceptibility $\epsilon_\perp(\omega)$ is assumed to have a constant value of, say ϵ. At $\omega=0$, the value of R is $|(\omega_1^2-\epsilon\omega_2^2)/(\omega_1^2+\epsilon\omega_2^2)|^2$, while at high frequencies it tends to $|(1-\epsilon)/(1+\epsilon)|^2$.

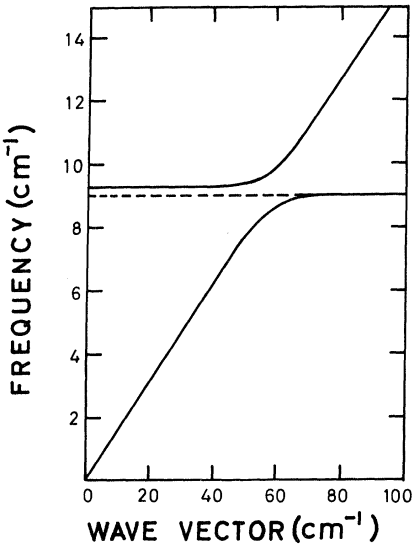

FIG. 5. Magniton-dispersion curves for MnF_2 for $\hat{k}$ parallel to the z axis. The parameters employed in the calculation are listed in Table I. The value of ω_1 is 9.10 cm^{-1}, while that of ω_2 is 9.2 cm^{-1}.

$\epsilon_\perp(\omega)$ being the transverse dielectric function. The reflectivity spectrum to be expected is sketched in Fig. 4, the characteristic feature of which is a flat-topped band extending from ω_1 to its "longitudinal counterpart" ω_2, exactly analogous to the reflectivity bands associated with infrared vibrational frequencies of crystals. The reflectivity spectra for other directions of incidence may be obtained in a similar manner.

Turning now to a quantitative estimate of the dispersion effects in MnF_2, we present in Fig. 5 the curves for $\hat{k}$ parallel to the z axis. The parameters employed in this calculation are summarized in Table I. Inspection of Fig. 5 shows that the frequency gap, though present, is disappointingly small for measurement by reflectivity experiments. Furthermore, in contrast to polariton spectra, the dispersion effects are also small, the transition between the photonlike and magnonlike regions being abrupt. Even so, it is conceivable that the sloping part may be accessible to forward-Raman-scattering experiments. The experimental problems will, however, be considerable in view of the small frequency shifts involved in the scattering.

Finally, we would like to observe that even though the effects of magniton formation in MnF_2 are rather small, it is possible they are much larger in other crystals. This of course can be confirmed only by detailed calculations. Formally, the equation to be solved can be obtained by combining (17) with

$$\vec{M} = \overleftrightarrow{\chi}(\omega)\vec{H} \ . \tag{27}$$

This leads to

$$\left(\frac{\omega^2}{c^2}\,\mu_{\alpha\beta}(\omega) - k^2\delta_{\alpha\beta} + k_\alpha k_\beta\right)\vec{H} = 0 \ ; \tag{28}$$

the requirement of nontrivial solutions for $\vec{H}$ then yields the determinantal equation

$$\left|\frac{\omega^2}{c^2}\,\mu_{\alpha\beta}(\omega) - k^2\delta_{\alpha\beta} + k_\alpha k_\beta\right| = 0 \ , \tag{29}$$

which is analogous to the general equation for exciton-photon and phonon-photon coupling.[21,22] By relating $\overleftrightarrow{\mu}(\omega)$ to the details of the magnon spectrum, the effects of retardation may be explicitly calculated for any given case, as we have done here for MnF_2.

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Spin Dynamics in the One-Dimensional Antiferromagnet $(CD_3)_4NMnCl_3$

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Recent studies of the instantaneous magnetic correlations in $(CD_3)_4NMnCl_3$ using quasielastic-neutron-scattering techniques have shown that the $MnCl_3$ chains in this compound exhibit purely one-dimensional paramagnetic behavior down to 1.1 °K. The interactions between Mn^{2+} ions along the chain are such that a molecular field theory would predict an ordering at ~76 °K. It was found that both the spatial and thermal variation of the instantaneous correlations could be quantitatively accounted for using Fisher's theory for the classical Heisenberg linear chain. In this paper we report a detailed study of the time-dependent magnetic correlations in $(CD_3)_4NMnCl_3$ using inelastic-neutron-scattering techniques. It is bound that at low temperatures, for $q \gg \kappa$ and $\omega \neq 0$, the Van Hove scattering function $S(\vec{Q}, \omega)$ may be accurately described by spin-wave theory with a dispersion relation $\hbar\omega = 6.1/\sin\pi q_c^z$* meV over the entire one-dimensional Brillouin zone, even though there is no long-range order. As the temperature is increased from 1.9 to 40 °K these "spin waves" typically weaken in intensity and broaden asymmetrically, with the scattering increasing on the low-energy side. In no case were both well-defined spin waves and a central diffusive component observed simultaneously, although the latter, if weak, could have been masked by the large incoherent scattering.

I. INTRODUCTION

Recently, there has been considerable effort on the part of experimentalists and theorists to understand the dynamical behavior of the spins in Heisenberg paramagnets both near T_c and at higher temperatures. The most complete study to date has been on the compound $RbMnF_3$, which is an excellent example of a three-dimensional (3d) Heisenberg antiferromagnet.[1-9] One of the striking features of the spin dynamics near T_N in this system is the persistence of magnonlike modes into the paramagnetic regime.[1] This effect is more pronounced for systems of lower dimensionality, such as the 2d antiferromagnet K_2NiF_4[10] and the linear-chain system $CsMnCl_3 \cdot 2H_2O$.[11] The possible existence of paramagnetic spin waves, together with the fact that a truly 1d system cannot exhibit long-range order at nonzero temperatures,[12] make a complete study of the dynamics of an ideal 1d Heisenberg paramagnet particularly appealing.

In this paper we report a detailed study of the spin dynamics in the linear-chain antiferromagnet $(CD_3)_4NMnCl_3$ (TMMC).[13] Both bulk-susceptibility measurements[14] and measurements of the instantaneous correlations[15] using quasielastic-neutron-scattering techniques have shown that the $MnCl_3$ chains in this material exhibit remarkably good 1d paramagnetic behavior from high temperatures (the molecular-field-ordering temperature T_{MF} is ~76 °K) down to 1.1 °K. Furthermore, the instantaneous correlations can be quantitatively accounted for at all temperatures using Fisher's[16] exact solution for the classical Heisenberg 1d antiferromagnet with nearest-neighbor interactions. The dynamics of the spins in this system therefore should be particularly amenable to theory.

Theoretical work on the spin dynamics of the linear antiferromagnetic chain at other than infinite temperatures has until recently been wholly concerned with the $S = \frac{1}{2}$ case at 0°. Lieb, Schultz, and Mattis[17] derived the spectrum of first excited states for the XY model, and that for the Heisenberg model has been calculated exactly by des Cloizeaux and Pearson.[18] In both cases these states have been identified as magnonlike states with a simple sine dispersion curve, although the coefficients differ for the two models. des Cloizeaux and