EVIDENCE FOR MAGNON-MAGNON INTERACTIONS IN $RbMnF_3$

P. A. Fleury
Bell Telephone Laboratories, Holmdel, New Jersey
(Received 27 May 1968)

We report the observation of light scattering from two-magnon excitations in $RbMnF_3$. The shape and polarization of the spectrum are compared with theoretical predictions of Elliott et al. and confirm the importance of magnon-magnon interactions. Temperature dependences of the frequency and linewidth of the two-magnon spectrum are also given.

Recent experiments on the scattering[1] and absorption[2,3] of light in antiferromagnets have provided rather direct access to two-magnon excitations. Processes involving the simultaneous creation of pairs of magnons (with essentially equal and opposite wave vectors) have been shown to probe most effectively magnons in regions of high symmetry in the Brillouin zone. For simply structured magnetic systems, knowledge of the crystal symmetry and the magnon dispersion relation (from neutron scattering) enables one to predict details of the two-magnon spectrum. It has already been shown that the simplest such calculations, which neglect magnon-magnon interactions, produce inadequate agreement with experiment.[1,3] The effects of magnon-magnon interactions have now been calculated by Elliott et al.[4] for the case of a simple cubic antiferromagnet, such as $RbMnF_3$.[5] Here we report the observation of the two-magnon scattering of light in antiferromagnetic $RbMnF_3$ that confirms the striking effects of magnon-magnon interactions on the spectrum. In addition, temperature dependence of the frequency and width of the two-magnon spectrum are presented.

The experimental apparatus is described in detail elsewhere.[1] Linearly polarized light at 4880 Å from an argon-ion laser (~200 mW) was focused into an oriented single crystal of $RbMnF_3$. Sample temperature was controlled by a flowing He-gas arrangement. Light scattered through 90° was frequency analyzed with a double monochromator and photoelectrically detected. The spectrum obtained at ~10°K appears as the solid line in Fig. 1.

In the preceding Letter, it was argued that the effects of magnon-magnon interactions should be more readily observable in spectral measurements than in thermodynamic or other experiments, primarily because of the local nature of the magnons' interaction with light. The nature and strength of the magnon-magnon interaction are already contained in the usual nearest-neighbor Heisenberg exchange Hamiltonian[4] and in the nonlinear character of the Holstein-Primakoff transformation.[6] Therefore, the effects of magnon-magnon interactions on light scattering contain no adjustable parameters. The case of magnon-magnon interactions is thus different from the magnon-exciton case[4] in which the sideband position may be adjusted by proper choice of the excited-state exchange parameter, J'.

The symmetry and selection rules of two-magnon light scattering are unchanged by the inclusion of magnon interactions.

On the basis of other two-magnon scattering experiments, in antiferromagnetic MnF_2 and FeF_2, we expect the two-magnon state responsible for light scattering to be composed of one magnon from each of the antiferromagnetic sublattices.[1] As mentioned by Elliott et al.,[4] this implies that the most important terms in the Hamiltonian describing scattering of the incident field

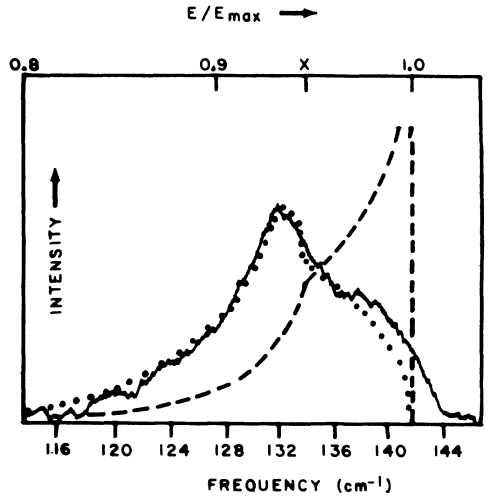

FIG. 1. Two-magnon light scattering in $RbMnF_3$ at ~10°K. The solid line shows experimental results with an instrumental width of less than 2 cm^{-1}. The Γ_3^+ component was observed with $\vec{E}$ parallel to [110], $\vec{E}'$ parallel to [$\bar{1}$10]. Dashed curve is theoretical prediction for Γ_3^+ without magnon-magnon interactions. Dotted curve is theory for Γ_3^+ with magnon-magnon interactions included. The frequency of magnons at the X point in the Brillouin zone is at X.

$\vec{E}$ into the scattered field $\vec{E}'$ contain spin contributions of the form $S_{\vec{R}}^{+}S_{\vec{R}+\vec{r}}^{-}$. Here $\vec{R}$ and $\vec{R}+\vec{r}$ are vectors referring to sites on spin-down and spin-up sublattices, respectively. The most general form for such an interaction Hamiltonian is

$$\mathcal{H}' = \sum_{\vec{R}}\sum_{\vec{r}}\sum_{\alpha\beta\gamma\delta} E_\alpha E_\delta' B_{\alpha\delta\beta\gamma}(r) S_{\vec{R}}^{\beta} S_{\vec{R}+\vec{r}}^{\gamma}. \tag{1}$$

The symmetry of the magnetic crystal dictates the form of the B tensor. For a simple cubic perovskite antiferromagnet like $RbMnF_3$, the proper form of (1) is

$$\sum_{\vec{R}}\sum_{\vec{r}} \{B_1 \vec{E}\cdot\vec{E}' + B_3[(\vec{E}\cdot\vec{r})(\vec{E}'\cdot\vec{r}) - \tfrac{1}{3}\vec{E}\cdot\vec{E}']\}\vec{S}_{\vec{R}}\cdot\vec{S}_{\vec{R}+\vec{r}}. \tag{2}$$

The notation is the same as that of Ref. 4. The fact that terms multiplying B_1 and B_3 transform as Γ_1^+ and Γ_3^+, respectively, implies that the following polarization combinations will govern the scattering experiment:

$$\text{for } \Gamma_1^+,\ (\vec{E}\cdot\vec{E}')^2;$$
$$\text{for } \Gamma_3^+,\ (E_xE_x')^2 + (E_yE_y')^2 + (E_zE_z')^2 - E_xE_x'E_yE_y' - E_xE_x'E_zE_z' - E_yE_y'E_zE_z'. \tag{3}$$

The coordinate axes refer to those of the cubic unit cell.

The Γ_1^+ and Γ_3^+ contributions were separated experimentally by orienting the crystal so that light polarizations in [110] type directions were convenient. The data of Fig. 1 were obtained with $\vec{E}$ parallel to [110] and $\vec{E}'$ parallel to [$\bar{1}$10]. Since these are orthogonal polarizations, the Γ_1^+ component does not appear. From the fact that the same shape was obtained with both $\vec{E}$ and $\vec{E}'$ parallel to (100), we infer that B_3 is much greater than B_1 in $RbMnF_3$. The absolute scattering efficiency for the two-magnon peak in $RbMnF_3$ is of the same order as that in MnF_2, $\sim 10^{-12}$ cm^{-1} sr^{-1}.

As mentioned previously, these considerations are independent of magnon-magnon interactions. These interactions affect the details of the line shape. The shape of the Γ_3^+ component at ~10°K in $RbMnF_3$ is compared with the predictions of Elliott et al.[4] in Fig. 1. The experimental curve is the solid line; the dashed line represents the theory that ignores magnon-magnon interactions; the dotted line represents the theory including magnon-magnon interactions. For the theoretical curves the value of $E_{max} = 2nSJ$ was determined by[7] $S = \frac{5}{2}$, $n = 6$, and $J = 4.7$ cm^{-1}. The excellent agreement between the solid and dotted curves provides striking confirmation of the importance of interactions between magnon pairs excited by light. It should be emphasized that there are no shape or position determining parameters in the theory which can be adjusted to fit the experiment. Only the peak intensity of the theoretical curve was normalized to the experimental data.

Some other features of the two-magnon peak which have yet to receive theoretical attention have also been studied experimentally. We have measured the temperature dependence of both the frequency and linewidth of the two-magnon spectrum. The frequency of the peak-intensity point decreases smoothly from 133 cm^{-1} at 10°K to 92 cm^{-1} at 71°K. Some scattering is still observable at 80°K, although precise location of the peak is difficult. The Néel temperature of $RbMnF_3$ is 82.5°K. As T increases the two-magnon peak becomes more asymmetric and develops a low-frequency tail. The full width at half maximum increases from ~11 cm^{-1} at 10°K to ~52 cm^{-1} at 71°K. Both the width and peak frequency of the two-magnon spectrum are affected by a decrease in zone-edge magnon frequency[8] and an increase in the importance of magnon-magnon interactions as T is increased toward T_N. Generalizations of the theory to finite temperatures should provide additional interesting information on magnon interactions, and possibly on the magnetic phase transition itself.

Generalizations of the theory to the slightly more complicated tetragonal antiferromagnets, like MnF_2, is also desirable. Our results on $RbMnF_3$ make it quite likely that previously noted discrepancies[1] on the two-magnon line shapes will be resolved by including magnon-magnon in-

teractions.

It is a pleasure to thank Professor R. Loudon for informing me of the theoretical work of Ref. 4 and for continuing helpful discussions. I am also grateful to H. J. Guggenheim for a sample of $RbMnF_3$, to H. L. Carter for technical assistance, and to V. C. Wade for crystal polishing.

[1]P. A. Fleury and R. Loudon, Phys. Rev. 166, 514 (1968) and references cited therein.

[2]J. W. Halley and I. Silvera, Phys. Rev. Letters 15, 654 (1965).

[3]S. J. Allen, R. Loudon, and P. L. Richards, Phys. Rev. Letters 16, 463 (1966).

[4]R. J. Elliott, M. F. Thorpe, G. F. Imbusch, R. Loudon, and J. B. Parkinson, preceding Letter [Phys. Rev. Letters 20, 147 (1968)].

[5]S. Freeman and J. J. Hopfield, Bull. Am. Phys. Soc. 13, 388 (1968), have noted that for a cubic ferromagnet with spin $\frac{5}{2}$ the effects of magnon-magnon interactions are minor.

[6]See for example C. Kittel, Quantum Theory of Solids (John Wiley & Sons, Inc., New York, 1963), pp. 52-60.

[7]C. G. Windsor and R. W. H. Stevenson, Proc. Phys. Soc. (London) 87, 501 (1966).

[8]R. M. White, Phys. Letters 19, 453 (1965).

FREQUENCY-DEPENDENT DIELECTRIC FUNCTION OF A ZERO-GAP SEMICONDUCTOR*

D. Sherrington† and W. Kohn
Department of Physics, University of California, San Diego, La Jolla, California 92037
(Received 26 April 1968)

The frequency-dependent dielectric function $\epsilon(\omega)$ of an inherently zero-gap semiconductor, such as grey tin, has unusual properties at low temperatures. In undoped samples $\epsilon(\omega) \sim \omega^{-1/2}$; in doped samples interband effects considerably modify the coefficient of the additional ω^{-2} term from its free-carrier value, the sign being reversed for p-type impurities. These properties should be observable in reflectance measurements.

It has recently been shown[1-3] that the zero-temperature dielectric function $\epsilon(q)$ of a semiconductor whose highest conduction- and lowest valence-band[4] edges are inherently degenerate at a point in k space exhibits a $1/q$ singularity for small q. Here we consider the frequency-dependent dielectric function $\epsilon(\omega)$ and show that it has some interesting properties which should be observable by infrared reflectance measurements at low temperature. We find that the dielectric function of an undoped semiconductor of this type contains an $\omega^{-1/2}$ term. In a doped sample one finds, for intermediate frequencies, an additional ω^{-2} term whose coefficient differs from that of the Drude theory and for holes even has the opposite sign.

A semiconductor with this band structure is grey tin[5] in which the degeneracy is at $k = 0$ (see Fig. 1). Grey tin has the diamond structure and the bands shown are related to the valence bands of Si or Ge, with the important difference that the curvature of the light-mass band is reversed. We shall concern ourselves explicitly with this structure, which is also that of HgTe, HgSe.[6]

The dielectric function is conveniently split into two parts:

$$\epsilon(\omega) = \epsilon^{(a)}(\omega) + \epsilon^{(b)}(\omega), \tag{1}$$

where $\epsilon^{(b)}$ arises from transitions involving only the conduction and valence bands and $\epsilon^{(a)}$ is the contribution from all the other bands in transitions between themselves and with the conduction and valence bands. For frequencies much less than the direct gaps between these other bands and the conduction and valence bands $\epsilon^{(a)}(\omega)$ may be considered constant, and for grey tin it may be shown to be of the same order as the dielectric constant of Ge or Si.

We write

$$\epsilon^{(b)}(\omega) = \epsilon_1^{(b)}(\omega) + i\epsilon_2^{(b)}(\omega). \tag{2}$$

The contribution to $\epsilon_2^{(b)}$ of an electron in the state $|\vec{k}, i\rangle$, where $i = 1, \cdots, 4$ labels the doubly

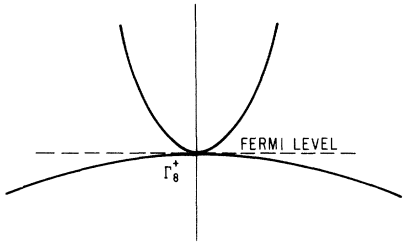

FIG. 1. Conduction- and valence-band extrema in intrinsic grey tin (after Groves and Paul, Ref. 5).