count for background absorption and the conversion of transmission into absorption intensity, then the observed line is found to have the expected, asymmetric shape.

(5) From the fact that the two magnons are on opposite sublattices, we expect that the frequency should be magnetic-field independent, as observed.

(6) A molecular-field calculation shows that the frequency will fall with temperature more slowly that the AFMR frequency.

(7) The line is expected to disappear for $T > T_N$ as observed.

(8) A search of all processes through fourth order in perturbation theory in a model including excitons, magnons, and phonons, and their interactions shows that no other process in the model can explain the line.

The process by which magnon-assisted optical absorption is expected to take place is shown in Fig. 2(c). Experimental evidence for the existence of lines due to this process in MnF_2 is described in the accompanying Letter.

The work is described in more detail in a series of forthcoming papers. The authors are indebted to their research advisors, Professor M. Tinkham and Professor C. Kittel, for suggesting some of the studies reported here and for advice, encouragement, and helpful suggestions. We are also grateful to R. L. Greene et al. for showing us the results of the optical-absorption experiments described in the accompanying Letter prior to publication.

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OBSERVATION OF A SPIN-WAVE SIDEBAND IN THE OPTICAL SPECTRUM OF MnF_2†

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We have found evidence for a spin-wave sideband in the absorption spectrum of antiferromagnetic MnF_2 (Néel temperature 67.7°K. To our knowledge this is the first observation of a spin-wave sideband and provides a new method for investigating the spin-wave spectrum.

A familiar feature of optical spectra in crystals is the presence of vibronic or phonon sidebands which accompany pure electronic transitions (no-phonon lines).[1] In many cases these sidebands are stronger than the pure electronic transitions because of certain selection rules. For example, if the ground and excited states have the same parity, then electric dipole transitions are forbidden and only the weaker magnetic dipole transitions can occur. An odd vibration can destroy the inversion symmetry and permit electric dipole transitions to occur. It seems plausible then that other collective excitations such as spin waves (magnons) could have similar effects.

The sharp-line absorption spectra at 2.2°K for the transition ${}^6A_{1g}({}^6S)$ to ${}^4T_{1g}({}^4G)$ of the manganese ion in MnF_2 is shown in Fig 1(a). The position, shape, and temperature dependence of the electric dipole transition at 18 477.1 cm^{-1} indicate that this line is a spin-wave, or magnonic, sideband of the 18 419.6-cm^{-1} magnetic dipole line. The two lower energy lines shown in Fig 1(a) are magnetic dipole transitions. These are the first pure electronic transitions observed in MnF_2; all the other features of the spectrum are vibronic in nature. In this paper we will restrict ourselves to a discussion of the spin-wave sideband and its associated no-magnon line.

The shape of the magnonic sideband is determined primarily by the magnon dispersion relation From time-dependent perturbation theory, it is found that the absorption coefficient $\alpha(\omega)$

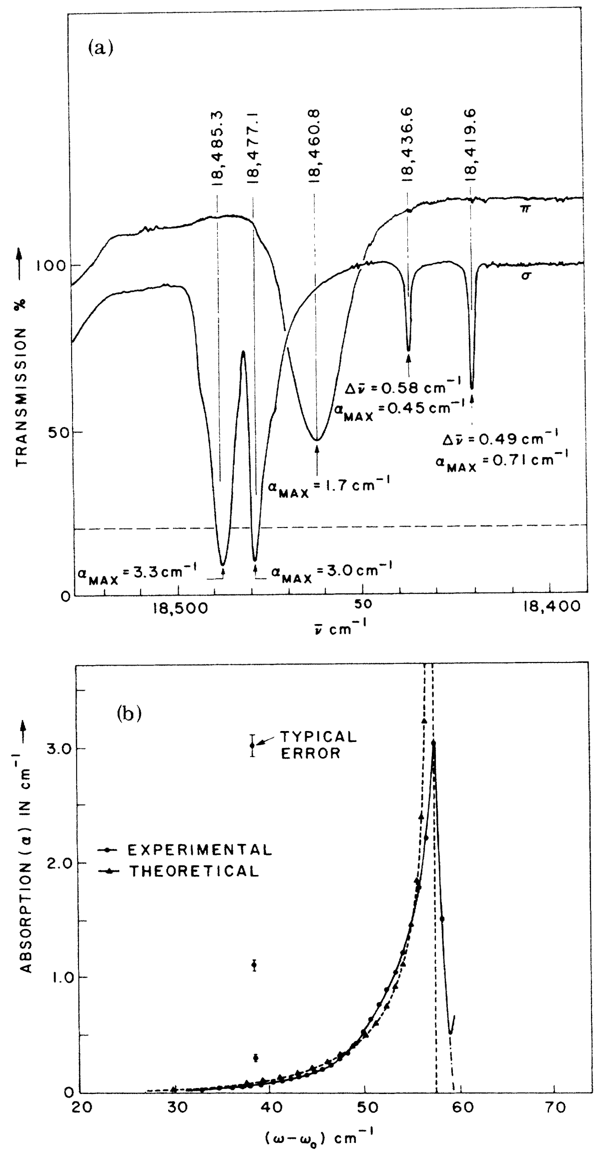

FIG. 1. (a) Sharp-line structure of $^6A_{1g}(^6S) \rightarrow {}^4T_{1g}(^4G)$ transition of Mn^{2+} in MnF_2 at 2.2°K in σ and π polarizations. A broad band (not shown) peaking at 19 400 cm^{-1} is also observed. The two lower energy lines are magnetic dipole transitions. The rest are electric dipole. The dipole character was determined through α (axial), π, and σ polarization studies. The baseline of the π-polarized transition has been displaced 20% for clarity. (b) Magnonic sideband line shape (peak at 18 477.1 cm^{-1}) at 2.2°K as determined experimentally and theoretically. Theoretical calculation is discussed in text.

is given by

$$\alpha(\omega) = |C|^2 k^2 (d\omega_{\vec{k}}/d\vec{k})^{-1} |s_{\vec{k}}|^2, \tag{1}$$

where $\omega = \omega_0 + \omega_k$; ω_0 is the pure electronic-transition frequency, and ω_k the magnon frequency. $|C|^2$ is a constant depending upon the detailed mechanism of the process, and $|s_{\vec{k}}|^2$ is a coupling parameter which depends on the magnon wave vector $\vec{k}$. For a two-sublattice antiferromagnet, the magnon dispersion is[2]

$$\omega_k = g\mu_B H_E [(1 + H_A/H_E)^2 - \gamma_k^2]^{1/2}, \tag{2}$$

where $\gamma_k = (Z)^{-1} \Sigma_\delta \exp(i\vec{k}\cdot\vec{\delta})$ and H_E and H_A are the usual exchange and anisotropy fields. The vector $\vec{\delta}$ connects a Mn^{2+} ion to its Z (eight for MnF_2) nearest neighbors on the opposite sublattice. The coupling parameter[3] is

$$s_{\vec{k}} = \sum_{\delta_k > 0} \sin k\delta_k, \tag{3}$$

where δ_k is the projection of $\vec{\delta}$ along the direction of spin-wave propagation. Equations (1) and (2) predict that the sideband peak should occur at the maximum magnon frequency, $\omega_M = g\mu_B(H_E + H_A)$, corresponding to $\vec{k}$ at the first Brillouin-zone boundary.[4] The neutron-scattering data of Low et al.[5] yield a value of $\omega_M = 54.8 \pm 0.7$ cm^{-1} at 4.2°K. Our experimental results [Fig. 1(a)] give a value of 57.5 ± 0.1 cm^{-1}.

A theoretical approximation to $\alpha(\omega)$ is obtained from Eq. (1) using values of H_E and H_A such that $\omega_M = 57.5$ cm^{-1}.[6] The theoretical and experimental absorption coefficients are compared in Fig. 1(b). The correspondence is good except near the peak where Eq. (1) does not apply because damping effects have not been considered and the theoretical peak goes to infinity. The free parameter $|C|^2$ has been chosen to fit the experimental value at $\alpha = \alpha_{max}/2$.

It should be pointed out that similar expressions can be used to deduce the shape of an acoustic phonon sideband. The possibility of the observed sideband being vibronic is ruled out for two reasons: (1) The phonon peak wave number would be displaced from the pure electronic transition by ~300 cm^{-1}, and (2) the sideband absorption would be appreciable 50 cm^{-1} or more away from the peak on the long-wavelength side.

The temperature dependence of the magnon-band peak, ω_M, is determined by that of H_E. Since H_E varies directly as the sublattice magnetization[7] $M(T)$, the ratio $\omega_M(T)/\omega_M(0)$ should equal $M(T)/M(0)$. In Fig. 2, the observed temperature dependence of $\omega_M(T)/\omega_M(0)$ is compared to $M(T)/M(0)$ determined by Jaccarino and Shulman from F^{19} nmr.[8] Data could not be taken above 30°K due to broadening of the sideband. The magnetic dipole lines have no measurable shift between 2.2 and 30°K.

The local symmetry at a Mn^{2+} ion is D_{2h}. The pure electronic transition considered here is spin and parity forbidden. Spin-orbit coupling relaxes the spin selection rule so that magnetic dipole transitions can occur. Electric dipole transitions can occur only if the local inversion symmetry is destroyed. A possible mechanism by which a spin wave destroys the inversion symmetry has been proposed by Halley.[9] He notes that each Mn^{2+} ion has a spin-dependent electric quadrupole moment due to spin-orbit coupling. A short-wavelength spin wave cants the spins so that the spins on either side of a given Mn^{2+} ion tip towards this ion. This destroys the inversion symmetry. The crystal field at a given Mn^{2+} ion thus has an odd component due to the quadrupole fields of the neighboring ions. This permits electric dipole transitions to occur.

Two points deserve further consideration. The difference between our measured value

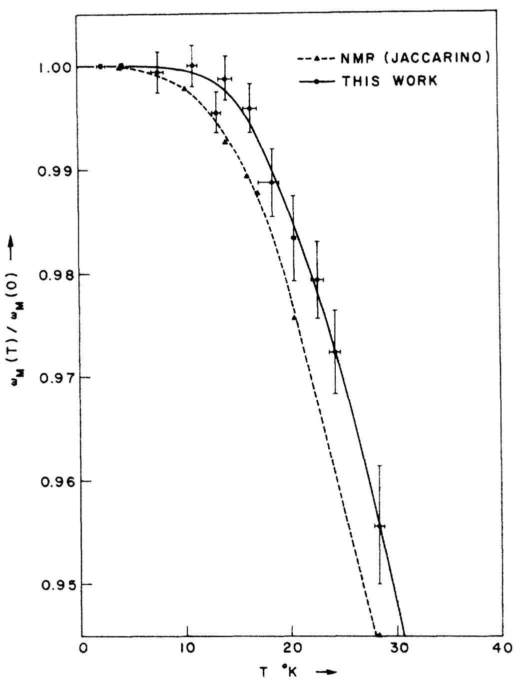

FIG. 2. Temperature dependence of the sideband peak position expressed as the ratio $\omega_M(T)/\omega_M(0)$. Comparison is made to the F^{19} nmr data of Jaccarino and Shulman (reference 8).

of ω_M and that obtained from neutron scattering is 2.5 cm^{-1}. This may be the result of exciton dispersion. The electronic states of a condensed system such as MnF_2 should be described in terms of exciton theory. According to Halley, the interaction responsible for the magnonic sideband results in the emission of a magnon and exciton with equal and opposite wave vectors. Hence our measured value of ω_M would be $\omega_M = \omega_{magnon} + \omega_{exciton}$ (both values at the zone boundary). Our results would then indicate that the difference in energy between a $k=0$ and a $k=\pi/c$ exciton is no more than 2.5 cm^{-1}.

The difference between the observed values of the ratios $\omega_M(T)/\omega_M(0)$ and $M(T)/M(0)$ is larger than the experimental error. This difference is not accounted for by line pulling due to the broadening of the 18 485.3-cm^{-1} transition, nor is it accounted for by considering the effects of the exciton dispersion. In reality, these two ratios measure slightly different characteristics of the spin system. The sublattice magnetization is a measure of the thermal equilibrium population of the magnon modes. At low temperatures, this quantity is relatively insensitive to the shape of the dispersion relation near the zone boundary. The ratio $\omega_M(T)/\omega_M(0)$, however, is clearly related to the temperature dependence of the dispersion at the zone boundary. The slight deviation from the predicted theoretical behavior is then not surprising.

Several features of this transition are not well understood. The nature of the electronic states, the absence of a sideband to the 18 436.6-cm^{-1} line, and the identification of the 18 460.8-cm^{-1} and 18 485.3-cm^{-1} transitions are currently under investigation.

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BALLISTIC MEAN FREE PATH MEASUREMENTS OF HOT ELECTRONS IN Au FILMS

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This letter describes a new experimental approach whereby the mean free path of excited electrons in metals can be directly obtained. Measurements have been made of the mean free path of 0.85-eV electrons in Au films deposited on Ge and Si substrates. The electrons are injected into Au films of varying thicknesses from a Si point contact. On the other side of the film, a constant fraction of the electrons which have not undergone a collision in the gold are collected over the Au-semiconductor Schottky barrier at the substrate-film interface. The size of this fraction is shown to be in good agreement with theoretical estimates, and the temperature-dependent portion of the mean free path is identified quantitatively with the electron-phonon conductivity mean free path. Transport of the above kind has been reported by Atalla and Kahng,[1] but has not been sufficiently stable or reproducible to be used in a quantitative fashion.

A cantilever-beam mechanism[2] (Fig. 1) capable of contact-separation adjustments as low as 10 Å has been used to contact metal films as thin as 100 Å without appreciable deformation of the films in order to make measurements of the incremental emitter-to-collector current-transfer ratio, α, as a function of film thickness. The cantilever beam and supporting yoke were constructed from a single piece of steel to provide a stable clamp for the beam. The yoke also served as a vibration-free support for the micromanipulators required to make the base contacts to the Au films and to make coarse adjustments of the contact position. Low-temperature measurements were made in an atmosphere of dry nitrogen with samples on a small liquid-nitrogen container which in turn was mounted on the cantilever beam.

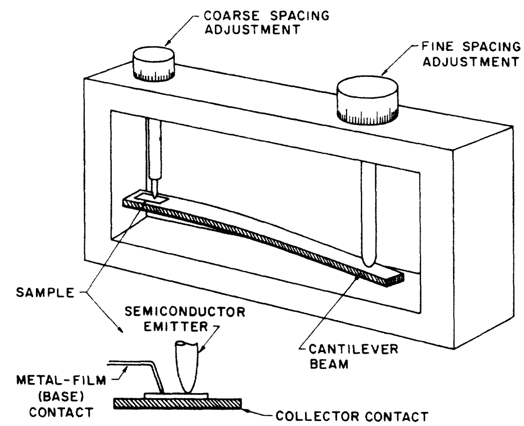

FIG. 1. Cantilever mechanism for fine adjustment of semiconductor emitter to metal-base spacing.

Au films of several thicknesses were deposited simultaneously by evaporation on chemically cleaned ⟨111⟩ Ge and Si surfaces in a Vac-Ion system at $\approx 10^{-8}$ Torr. Film thicknesses were measured by multiple-beam interferometry and a Taylor "Talysurf." The two measurements agreed within ±20 Å. A metal mask was used to define metal base regions ≈0.01 in. in diameter. The Si point had a radius of curvature of ≈0.001 in. The point was cleaned immediately before each set of measurements. The values of α were sensitive to the pressure of the point. The maximum α was obtained when the point just maintained contact with the Au film. The maximum values were reproducible and independent of the current for currents in excess of 0.5 mA.

Figure 2 shows the measured results. It is apparent that α varies exponentially with film