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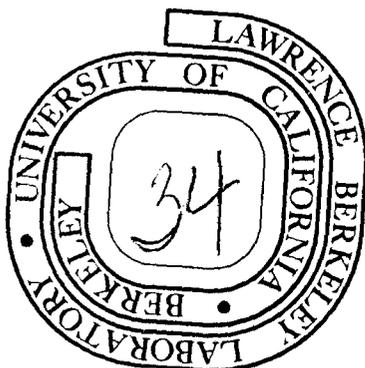
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ONE-MAGNON LUMINESCENCE SIDEBANDS OF THE
EXCITONIC TRANSITION IN MnF$_2$

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ABSTRACT

We report the first observation of the π-polarized one-
magnon excitonic luminescence sideband in MnF$_2$. The theory
of Loudon is used to fit the experimental spectrum quantita-
tively. An effective temperature of the crystal is deduced
from the simultaneously observed antiStokes sideband emission.

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One-magnon sideband of the excitonic transition in the $^6A_{1g}(^6S)$ — $^4T_{1g}(^4G)$ manifold in MnF_2 has been well studied in the literature.[1-3] In absorption, while the discrepancy between theory and experiment in the α- and σ-polarization spectra is small, it is quite large in the π-polarization. The discrepancy presumably results from ignoring the exciton-magnon interaction[4] in the theoretical calculation. In emission, since the exciton and magnon are not simultaneously present, the exciton-magnon interaction does not come into play. Then the theoretical calculation agrees very well with the experimental α- and σ-polarization spectra.[2] However, so far as we know, observation of the π-polarized one-magnon luminescence sideband, although predicted by theory,[3] has never been reported probably because of its much weaker intensity. Recently, in studying multi-magnon luminescence sidebands[5] in MnF_2, we have been able to observe clearly the π-polarized one-magnon sideband which is much weaker than those with α- and σ-polarizations. The spectrum can indeed be described almost perfectly by the theory without the exciton-magnon interaction. At higher temperatures, antiStokes luminescence of the sideband has also been observed. The temperature deduced from the Stokes-antiStokes ratio agrees with that obtained from the E_1 — E_2 exciton luminescence ratio.

The experiment was done using either a CW dye laser or a flash-pumped dye laser as the excitation source. The MnF_2 sample properly oriented was cooled by either gas or superfluid helium. Luminescence from the sample was analyzed by a double monochromator and detected by either a photon-counting system or a gated boxcar integrator.

Typical polarized luminescence spectra obtained with the pulsed laser are shown in Fig. 1. Impurity luminescence in this case was suppressed by

the gated detection scheme.[5,6] The observed exciton lines E_1 and E_2 in the $^4T_{1g} \rightarrow {}^6A_{1g}$ transitions and the α,σ-polarized one-magnon sidebands associated with E_1 agree well with those reported in the literature. The corresponding π-polarized one-magnon sideband is appreciably weaker and broader. At relatively higher temperatures, the antiStokes sideband emission is also clearly visible, and is relatively more intense for the π-polarization. In order to determine the sideband lineshape more accurately, we have also recorded the luminescence spectrum with the CW Ar$^+$ laser and photon-counting system. The result for the π-polarization is shown in Fig. 2, where in (a) the spectrum was obtained with the sample immersed in superfluid helium, and in (b) the spectrum was obtained with the sample in cold helium gas at 13°K. A strong impurity luminescence line is clearly present on the low-energy side of the sideband in Fig. 2(a), while the same line is apparently thermally quenched in Fig. 2(b).

The dispersions of the E_1 and E_2 excitons and the magnons in MnF_2 are shown in Fig. 3(a). To explain the observed sidebands, we use the theory of Loudon.[3] In his formalism, two-ion exchange interaction is responsible for the magnon creation or annihilation and the exciton-magnon interaction is neglected. If only the interaction between next-nearest neighbors on the opposite sublattice is taken into account, then the one-magnon sideband absorption $A^{nn}(\omega)$ and emission $E^{nn}(\omega)$ in the α or σ polarization are given, in the zero-temperature approximation, by

$$\begin{Bmatrix} A^{nn}_{\alpha,\sigma}(\omega) \\ E^{nn}_{\alpha,\sigma}(\omega) \end{Bmatrix} = \sum_{\vec{k}} \left[C^2 \cos^2\left(\frac{k_x a}{2}\right) \cos^2\left(\frac{k_y a}{2}\right) \right.$$
$$\left. + D^2 \sin^2\left(\frac{k_x a}{2}\right) \sin^2\left(\frac{k_y a}{2}\right) \right] \sin^2\left(\frac{k_z c}{2}\right) \begin{Bmatrix} u_k^2 \\ v_k^2 \end{Bmatrix} \delta[\hbar\omega - \varepsilon_0 \mp \varepsilon_m(\vec{k})] \tag{1}$$

and those in the π-polarization by

$$\begin{Bmatrix} A_\pi^{nn}(\omega) \\ E_\pi^{nn}(\omega) \end{Bmatrix} = F^2 \sum_{\vec{k}} \left[\sin^2\left(\frac{k_x a}{2}\right)\cos^2\left(\frac{k_y a}{2}\right) \right.$$
$$\left. + \cos^2\left(\frac{k_x a}{2}\right)\sin^2\left(\frac{k_y a}{2}\right) \right] \cos^2\left(\frac{k_z c}{2}\right) \begin{Bmatrix} u_k^2 \\ v_k^2 \end{Bmatrix} \delta[\hbar\omega - \varepsilon_0 \mp \varepsilon_m(\vec{k})] \tag{2}$$

where C, D, and F are coupling constants of the same order of magnitude,
a and c are lattice constants, ε_0 is the E_1 exciton energy, $\varepsilon_m(\vec{k})$ is the
magnon energy at $\vec{k}$, and u_k and v_k are defined in Ref. 3 and reproduced
here in Fig. 3(b). Similarly, one finds that the interaction between
nearest neighbors on the same sublattice contributes to the sideband ab-
sorption and emission in the α or σ polarization as

$$\begin{Bmatrix} A_{\alpha,\sigma}^{n}(\omega) \\ E_{\alpha,\sigma}^{n}(\omega) \end{Bmatrix} = G^2 \sum_{\vec{k}} \sin^2(k_z c) \begin{Bmatrix} v_k^2 \\ u_k^2 \end{Bmatrix} \delta[\hbar\omega - \varepsilon_0 \mp \varepsilon_m(\vec{k})] \tag{3}$$

but contributes nothing to the sideband absorption and emission in the π-
polarization, where G is again a coupling constant of the same order of
magnitude as C, D, and F.

The summations over $\vec{k}$ in the above equations are weighted heavily
towards the Brillouin zone edges by the large magnon density of states.
However, near the zone edges, v_k^2 tends to zero while u_k^2 remains finite as
shown in Fig. 3(b). It is then easy to see that

$$A_{\sigma,\alpha}(\omega) \cong A_{\sigma,\alpha}^{nn}(\omega) + A_{\sigma,\alpha}^{n}(\omega) \cong A_{\sigma,\alpha}^{nn}(\omega); \tag{5}$$

$$E_{\sigma,\alpha}(\omega) \cong E_{\sigma,\alpha}^{nn}(\omega) + E_{\sigma,\alpha}^{n}(\omega) \cong E_{\sigma,\alpha}^{n}(\omega); \qquad (6)$$

$$E_{\pi}(\omega) \cong E_{\pi}^{nn}(\omega) \quad ; \quad A_{\pi}(\omega) \cong A_{\pi}^{nn}(\omega) \qquad (7)$$

and $E_{\pi}(\omega) \ll A_{\pi}(\omega)$, $A_{\sigma,\alpha}(\omega)$, $E_{\sigma,\alpha}(\omega)$.

Dietz et al.[2] has used Eq. (3) to fit quantitatively the observed

one-magnon luminescence sideband in the α,σ-polarization. Similarly, we

can use Eq. (2) to fit the luminescence sideband in the π-polarization.

Figure 2(a) shows that the agreement between theory and experiment is in-

deed excellent. In the inset of Fig. 2, we also show the comparison be-

tween theory and experiment on the π-polarized one-magnon absorption side-

band. The discrepancy is obvious. Agreement in the luminescence case and

disagreement in the absorption case clearly indicates that the exciton-

magnon interaction is non-negligible in the absorption process. This in-

teraction should appreciably broaden the sideband absorption and shift it

to lower energy.[4] We notice that because of the difference in u_k^2 and v_k^2

associated with the sine and cosine terms in Eq. (2), the theoretical

lineshapes of the π-polarized absorption and emission sidebands are very

different. Also, the integrated strength ratio of absorption to emission

is about 23.

We realize from Eq. (2) that in the π-polarization the antiStokes

sideband emission is simply the inverse process of the sideband absorption

only if the thermal population of magnons is properly taken into account.

Therefore, we can excpet to obtain the antiStokes sideband spectrum by

simply multiplying the experimental absorption sideband, normalized to yield

the correct sideband absorption-to-Stokes-emission ratio of 23, by a Bose-Einstein distribution function at a proper temperature. This is shown in Fig. 2(b). The theoretical antiStokes spectrum corresponding to a temperature of 13.3°K fits very well with the observed spectrum. This temperature is in good agreement with the one deduced from the luminescence intensity ratio of the E_1 and E_2 exciton lines. In the α- and σ-polarizations, deduction of the effective temperature from the antiStokes sideband emission is not possible because of lack of a normalization constant relating the strengths of the absorption and Stokes emission sidebands.

In summary, we have shown that the theory of Loudon gives an excellent description of the observed π-polarized one-magnon luminescence sideband in MnF_2. The effective temperature of the crystal can be deduced from the simultaneously observed π-polarized antiStokes luminescence sideband.
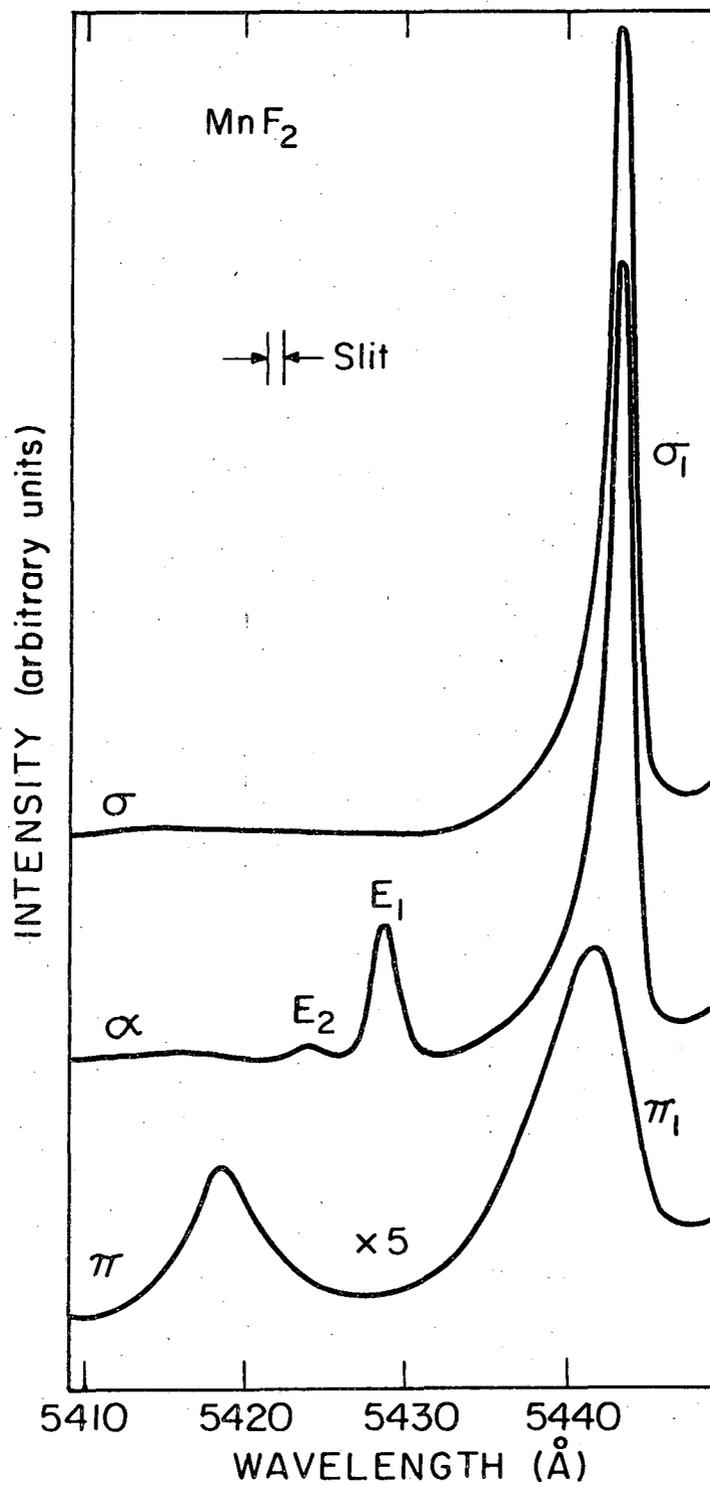
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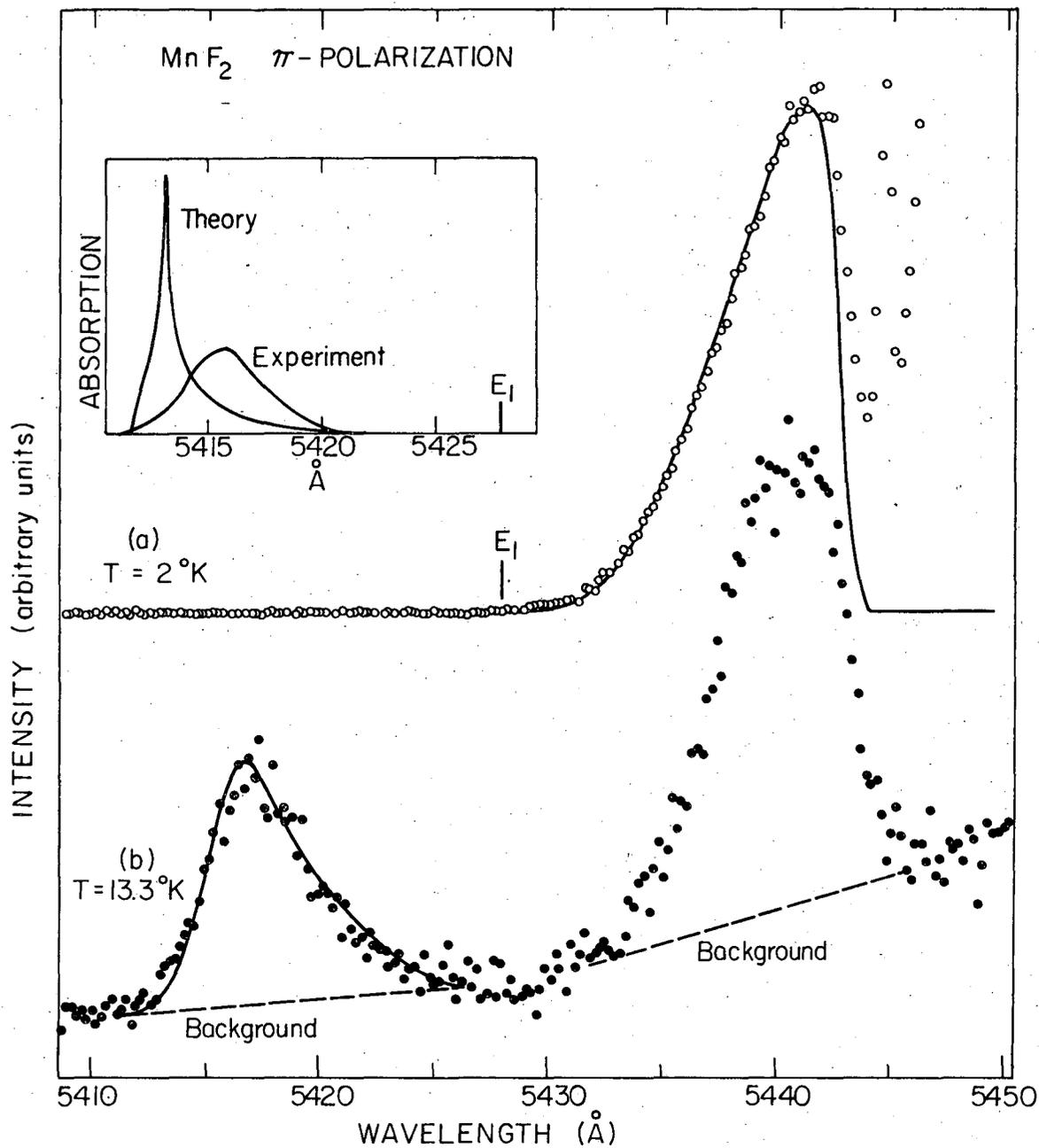
Figure Captions

Figure 1   Polarized intrinsic luminescence spectra of MnF_2 obtained

by the pulsed excitation-detection scheme. The laser excita-

tion had a wavelength of 5200 Å, a pulsewidth of 0.4 μsec,

a peak power of ∼ 30 MW/cm^2 for the π-polarization and ∼ 20

MW/cm^2 for the α- and σ-polarizations, and a repetition rate

of 6 pps. The boxcar used for detection had a gate width of

1 μsec. The sample was immersed in superfluid helium but

laser heating was still apparent. The effective sample tem-

perature was 12°K for the α- and σ-polarizations and 13.8°K

for the π-polarization.

Figure 2   π-polarized CW luminescence spectra of MnF_2 obtained with a

92 mW, 5145 Å, Ar^+ laser light: (a) with the sample immersed

in superfluid He, and (b) with the sample at 13°K. The spec-

trum in (b) is amplified by 10 relative to that in (a). Solid

lines are theoretical curves with the background taken into

account. The inset shows the comparison between theoretical

and experimental absorption sideband spectra.

Figure 3   (a) Schematic dispersion curves of excitons and magnons in

MnF_2.

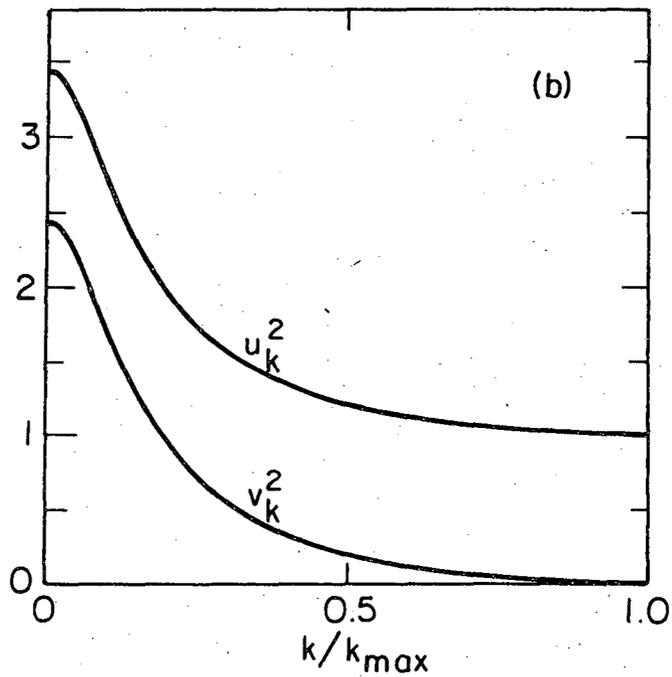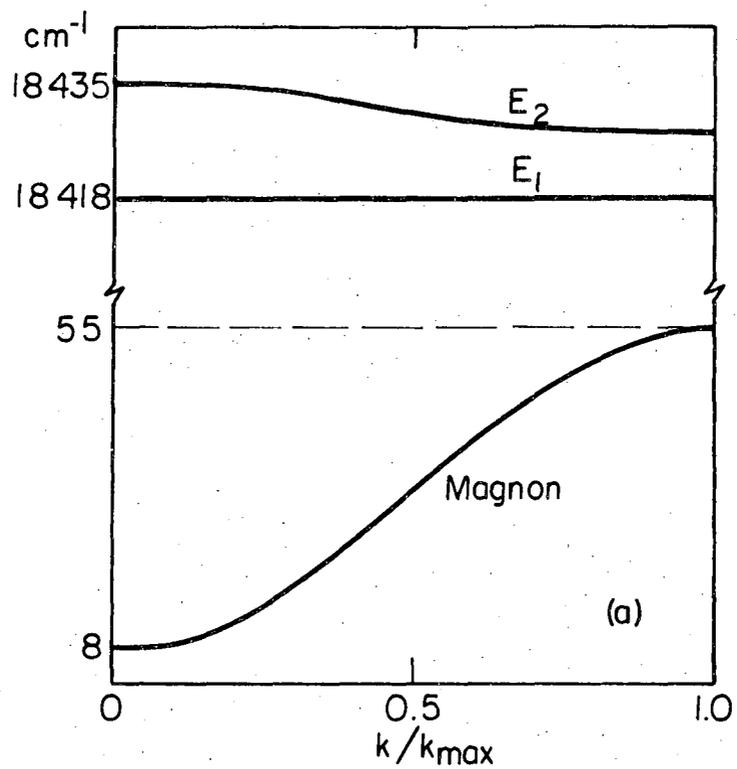(b) u_k^2 and v_k^2 as functions of k in the Brillouin zone.

Fig. 1

Fig. 2

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Fig. 3