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ONE-MAGNON LUMINESCENCE SIDEBANDS OF THE EXCITONIC TRANSITION IN MnF₂

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ABSTRACT

We report the first observation of the π-polarized one-magnon excitonic luminescence sideband in MnF₂. The theory of Loudon is used to fit the experimental spectrum quantitatively. An effective temperature of the crystal is deduced from the simultaneously observed anti-Stokes sideband emission.

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One-magnon sideband of the excitonic transition in the $^6A_{1g}(^6S) \rightarrow ^4T_{1g}(^4G)$ manifold in MnF$_2$ has been well studied in the literature. In absorption, while the discrepancy between theory and experiment in the $\alpha$- and $\sigma$-polarization spectra is small, it is quite large in the $\pi$-polarization. The discrepancy presumably results from ignoring the exciton-magnon interaction 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 $\alpha$- and $\sigma$-polarization spectra. However, so far as we know, observation of the $\pi$-polarized one-magnon luminescence sideband, although predicted by theory, has never been reported probably because of its much weaker intensity. Recently, in studying multi-magnon luminescence sidebands in MnF$_2$, we have been able to observe clearly the $\pi$-polarized one-magnon sideband which is much weaker than those with $\alpha$- and $\sigma$-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 $E_1 \rightarrow E_2$ 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. The observed exciton lines \( E_1 \) and \( E_2 \) in the \( ^4T_{1g} \rightarrow ^6A_{1g} \) transitions and the \( \alpha,\sigma \)-polarized one-magnon sidebands associated with \( E_1 \) agree well with those reported in the literature. The corresponding \( \pi \)-polarized one-magnon sideband is appreciably weaker and broader. At relatively higher temperatures, the anti-Stokes sideband emission is also clearly visible, and is relatively more intense for the \( \pi \)-polarization. In order to determine the sideband lineshape more accurately, we have also recorded the luminescence spectrum with the CW \( \text{Ar}^+ \) laser and photon-counting system. The result for the \( \pi \)-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 \( 13^\circ \text{K} \). 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 \( \text{MnF}_2 \) are shown in Fig. 3(a). To explain the observed sidebands, we use the theory of Loudon. 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 \( A^{nm}(\omega) \) and emission \( E^{nm}(\omega) \) in the \( \alpha \) or \( \sigma \) polarization are given, in the zero-temperature approximation, by

\[
\begin{align*}
\begin{pmatrix}
A^{nm}(\omega) \\
E^{nm}(\omega)
\end{pmatrix} &= \sum_{k} \left( C^2 \cos^2 \left( \frac{k_x a}{2} \right) \cos^2 \left( \frac{k_y a}{2} \right) \\
&\quad + D^2 \sin^2 \left( \frac{k_x a}{2} \right) \sin^2 \left( \frac{k_y a}{2} \right) \right) \sin^2 \left( \frac{k_z c}{2} \right) \delta \left[ \hbar \omega - \varepsilon_0 \mp \varepsilon_m(k) \right]
\end{align*}
\]
and those in the $\pi$-polarization by

$$
\begin{align*}
\left\{ A^{nn}_{\pi}(\omega) \right\} &= \pi^2 \sum_{k} \left[ \sin^2 \left( \frac{k x a}{2} \right) \cos^2 \left( \frac{k y a}{2} \right) \\
&+ \cos^2 \left( \frac{k x a}{2} \right) \sin^2 \left( \frac{k y a}{2} \right) \right] \cos \left( \frac{k z c}{2} \right) \left[ \frac{u_k^2}{v_k^2} \right] \delta \left[ \hbar \omega - \varepsilon_0 \mp \varepsilon_m(k) \right]
\end{align*}
$$

(2)

where $C$, $D$, and $F$ are coupling constants of the same order of magnitude, $a$ and $c$ are lattice constants, $\varepsilon_0$ is the $E_1$ exciton energy, $\varepsilon_m(k)$ is the magnon energy at $k$, 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 absorption and emission in the $\alpha$ or $\sigma$ polarization as

$$
\begin{align*}
\left\{ A^{n\sigma}_{\alpha}(\omega) \right\} &= G^2 \sum_{k} \sin^2(k z c) \left[ \frac{v_k^2}{u_k} \right] \delta \left[ \hbar \omega - \varepsilon_0 \mp \varepsilon_m(k) \right]
\end{align*}
$$

(3)

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

The summations over $k$ 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

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

(5)
\[ E_{\sigma,\alpha}(\omega) \approx E_{\sigma,\alpha}^{nn}(\omega) + E_{\sigma,\alpha}^{n}(\omega) \approx E_{\sigma,\alpha}^{n}(\omega); \]  

\[ E_{\pi}(\omega) \approx E_{\pi}^{nn}(\omega); \quad A_{\pi}(\omega) \approx A_{\pi}^{nn}(\omega) \]  

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 \(\alpha,\sigma\)-polarization. Similarly, we can use Eq. (2) to fit the luminescence sideband in the \(\pi\)-polarization. Figure 2(a) shows that the agreement between theory and experiment is indeed excellent. In the inset of Fig. 2, we also show the comparison between theory and experiment on the \(\pi\)-polarized one-magnon absorption sideband. 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 interaction 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 \(\pi\)-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 \(\pi\)-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 expect 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 anti-Stokes 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 $\alpha$- and $\sigma$-polarizations, deduction of the effective temperature from the anti-Stokes 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 $\pi$-polarized one-magnon luminescence sideband in MnF$_2$. The effective temperature of the crystal can be deduced from the simultaneously observed $\pi$-polarized anti-Stokes luminescence sideband.

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References

5. T. C. Chiang, P. R. Salvi, J. Davies, and Y. R. Shen, to be published.
Figure Captions

Figure 1  Polarized intrinsic luminescence spectra of MnF$_2$ obtained by the pulsed excitation-detection scheme. The laser excitation had a wavelength of 5200 Å, a pulsewidth of 0.4 μsec, a peak power of $\sim 30$ MW/cm$^2$ for the $\pi$-polarization and $\sim 20$ MW/cm$^2$ for the $\alpha$- and $\sigma$-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 temperature was 12°K for the $\alpha$- and $\sigma$-polarizations and 13.8°K for the $\pi$-polarization.

Figure 2  $\pi$-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 spectrum 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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