Hot-magnon sideband in the fluorescence spectrum of $CsMnCl_3 \cdot 2D_2O$

Weiyi Jia* and W. M. Yen
Department of Physics, University of Wisconsin, Madison, Wisconsin 53706
(Received 27 October 1981)

We report the observation of hot-magnon sidebands in the fluorescence spectrum of the $^4T_{1g}$ state of deuterated $CsMnCl_3 \cdot 2H_2O$. A simplified theoretical model similar to that used in an earlier study of $CsMnCl_3 \cdot 2H_2O$ accounts well for the hot-magnon line shapes. Evidence is also presented for the existence of multimagnon sidebands in the cold-magnon emission of this compound.

In an earlier paper, we reported on the optical properties of $CsMnCl_3 \cdot 2H_2O$ (CMC-H) and commented that there existed some evidence of the presence of hot-magnon transitions accompanying the $^4T_{1g} \rightarrow {}^6A_{1g}$ Mn^{2+} pure excitonic transition.[1] We also mentioned there that considerable nonradiative relaxation occurred in the $^4T_{1g}$ via the H_2O vibrations leading to fluorescence quenching and to generally weak optical signals. We have since expanded our investigation to include $CsMnCl_3 \cdot 2D_2O$ (CMC-D) and find that the spectral properties are the same as for CMC-H in all respects with the exception that the fluorescence transitions are orders of magnitude stronger because of reduced quenching from the lower-frequency D_2O vibrations. This increase in signal allows us to analyze the spectral structure of CMC in greater detail and in this Report we present an analysis of the various hot bands accompanying the $^4T_{1g}$ transition of deuterated CMC.

The formation of hot-magnon sidebands in fluorescence corresponds to the simultaneous emission of $^4T_{1g} \rightarrow {}^6A_1$ exciton coupled with the annihilation of a thermally activated magnon in the opposite antiferromagnetic sublattice.[2] This process then involves an ion pair in contrast to cold-band-magnon emission which is a single-ion process in CMC.[1]

In Fig. 1, we show experimental traces of the fluorescence emission of CMC-D for the $^4T_{1g} \rightarrow {}^6A_1$ transition observed in α polarization and as a function of laser excitation power. Reference 1 discusses experimental details and procedures more extensively. The samples in this case were immersed in a superfluid He bath of 1.8 K, but because the phonon sidebands are pumped there is considerable local heating at the higher laser powers used in the experiment. For an Ar laser pump power below 40 mW, the observed fluorescence is the standard spectrum observed for CMC consisting of the pure transition accompanied by magnon- and phonon-assisted structure to the long-wavelength (lower-energy) side of excitonic line. As the power of the laser is increased, additional structure becomes evident on the high-energy side (Fig. 1): for example, with 120 mW a peak labeled H_2 in the figure is apparent and a small peak labeled H_1 is also discernible—we believe that these two peaks correspond to hot-magnon sideband emission. With 250 mW of pump power, both H_1 and H_2 are clearly visible. H_1 is separated from the exciton peak by 18 cm^{-1}, while H_2 has a separation of 4 cm^{-1}; this separation is of the order of the inhomogeneous width of the excitation and hence H_2 appears as a shoulder on the latter. Comparison of the relative strengths of H_1 and H_2 allows us to calculate the actual lattice temperature to be ~6.5 K under these

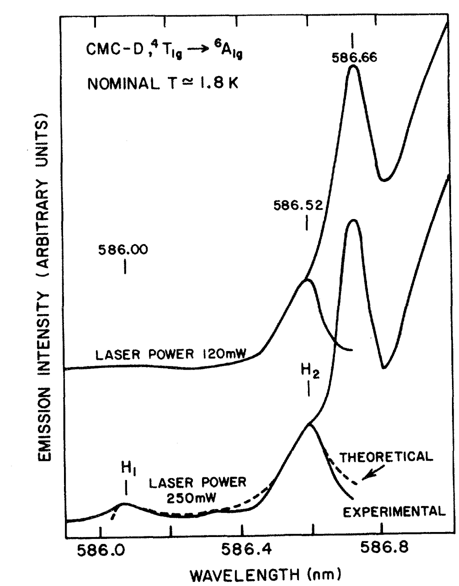

FIG. 1. Hot-band-magnon emission of CMC-D at 1.8 K.

pumping circumstances.

For the σ polarization similar results were obtained whereas in π ($\vec{E} \parallel b, \vec{H} \parallel a$) no additional hot-band peaks were observed even at 250-mW pump power levels.

The intensity of the hot-band-magnon emissions is governed by expressions similar to those of the cold-band-magnon absorption with some modifications. For the hot-band magnon, a Bose-Einstein factor describing the thermal distribution of magnon state density should be included, i.e.,

$$\langle n_k \rangle = \{\exp[E_m(k)/k_B T] - 1\}^{-1} \quad . \tag{1}$$

For the α polarization, the intensity of the hot-band emission can be written as

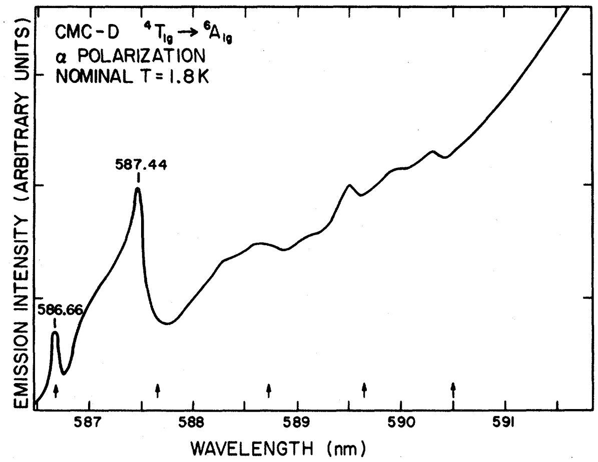

FIG. 2. Multimagnon sideband in CMC-D at 1.8 K.

$$E_H^\alpha = \sum_k^{BZ} (D_2^2 \sin^2 k_b b + D_3^2 \cos^2 \tfrac{1}{2} k_c c) \langle n_k \rangle U_k^2 \delta(\hbar\omega - E_e(k) - E_m(k))$$
$$+ \sum_k^{BZ} (C_4 \cos \tfrac{1}{2} k_a a \cos k_b b + C_6 \sin \tfrac{1}{2} k_a a \sin \tfrac{1}{2} k_c c)^2 \langle n_k \rangle V_k^2 \delta(\hbar\omega - E_e(k) - E_m(k)) \quad . \tag{2}$$

The notation is as in Ref. 1. Assuming the exciton dispersion can be neglected, $E_e(k) = E_0$. The magnon dispersion $E_m(K)$ is available at the absolute zero temperature.[1] At higher temperature the magnon dispersion should be renormalized[2]:

$$E_m(K,T) = R_k(T) E_m(k) \quad . \tag{3}$$

In our case, for simplicity, we will neglect the effect of renormalization and take $R_k(T) = 1$.

In hot-band-magnon emission, the exciton and the magnon coexist. The interaction between exciton and magnon causes the position of the magnon peaks to shift towards the origin.[3,4] As in the case of the cold-band-magnon absorption,[1] this effect is equivalent to the decrease of the number of the nearest neighbors. Using formulas and parameter values of Ref. 1 and taking $T = 6.5$ K, and $\alpha = 0.38$, the theoretical profile of the hot-band magnon on the α polarization has been calculated, as shown in Fig. 1 (dashed line). The agreement between the calculation and experiment is excellent.

At the lower-energy side of the exciton in the emission spectrum, in addition to the strong one-magnon sideband, there are a series of fluorescence peaks (Fig. 2). They form a regular progression. The peaks can be divided into four groups. Each one has two peaks (or one peak and one shoulder), which correspond to the shape of the magnon-state density. They are separated from each other by ~ 25 cm^{-1} which is the maximum magnon energy of CMC. In a one-dimensional magnetic system such as CMC, the excitation is localized and impurity lines will not appear. We have shown experimentally that all the peaks discussed above are independent of the impurities content of the sample and have the same lifetime as the exciton. We concluded that these structures are intrinsic and that they are multimagnon sidebands similar to those observed in $KMnF_3$ and MnF_2.[5,6] Arrows in Fig. 2 indicate the cutoff boundary of the multimagnon sidebands.

In conclusion, then, we have observed and analyzed hot-magnon bands in the emission of deuterated CMC and find agreement with a simplified theoretical model.[1] In addition, there appears to be evidence of multimagnon cold-band emission accompanying the $^4T_{1g} \to {}^6A_1$ transition of Mn^{2+} in CMC-D.

We acknowledge with thanks discussions with Dr. E. Strauss and support from the National Science Foundation under Grant No. DMR 78-20070.

*Present Address: Institute of Physics, Chinese Academy of Sciences, Beijing, People's Republic of China.

[1] Weiyi Jia, E. Strauss, and W. M. Yen, Phys. Rev. B 23, 6075 (1981).

[2] R. S. Meltzer, M. Lowe, and D. S. Mclure, Phys. Rev. 180, 561 (1969).

[3] R. J. Elliott, M. F. Thorpe, G. F. Imbusch, R. Loudon, and J. B. Parkinson, Phys. Rev. Lett. 21, 137 (1968).

[4] J. B. Parkinson and R. Loudon, J. Phys. C 1, 1568 (1968).

[5] E. Strauss, V. Gerhardt, and H. Riederer, J. Lumin. 12–13, 239 (1976).

[6] T. C. Chiang, P. Salvi, J. Davies, and Y. R. Shen, Solid State Commun. 26, 527 (1978).