Title
REPLY TO ""COMMENT ON TWO-MAGNON RESONANT RAMAN SCATTERING IN MnF2""

Author
Amer, Nabil M.

Publication Date
1976-02-01

Submitted to Physical Review Letters

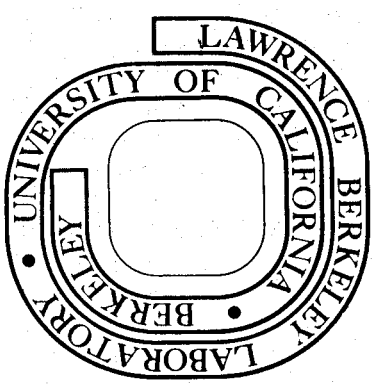
LBL-4917
Preprint

REPLY TO 'COMMENT ON TWO-MAGNON RESONANT RAMAN SCATTERING IN MnF_2"

Nabil M. Amer, Tai-chang Chiang, and Y. R. Shen

February 1976

Prepared for the U. S. Energy Research and
Development Administration under Contract W-7405-ENG-48



DISCLAIMER

LBL-4917

Reply to "Comment on Two-Magnon Resonant Raman Scattering in MnF_2"

Nabil M. Amer, Tai-chang Chiang, and Y. R. Shen†
Department of Physics, University of California
Berkeley, California

and

Materials and Molecular Research Division
Lawrence Berkeley Laboratory, Berkeley, California 94720

ABSTRACT

We found two-magnon modes in the resonant Raman spectra of both our MnF_2 sample and the purest MnF_2 sample used by Rousseau et al in the preceeding paper. We present here raw spectra obtained from both samples.

†On leave at The Miller Institute of University of California at Berkeley.

In the preceeding paper,[1] Rousseau et al reported their failure of observing the two-mangon resonant Raman scattering (RRS) in various samples of MnF_2. They suspect that the observed two-magnon modes in Ref. 2 are the results of experimental artifact. Recently, we exchanged samples with Rousseau et al. In our preliminary measurement on their purest sample (#1 in Table I of Ref. 1), we were able to readily detect the two-magnon modes at resonance.

Here, we present the resonant Raman and luminescence spectra of their sample (sample A) together with the spectra of our sample (sample B) measured in the same run.[3]

The experimental arrangement was the same as in Ref. 2. We used a Z(XY)Z geometry. The dye laser power on the sample was about 15 mW. Figure 1 shows the raw spectra (Raman + luminescence) obtained with the laser frequency at 18476 cm^{-1} which is very close to the peak of the σ_1 absorption band. Figure 1b and 1c are spectra for samples A and B respectively, while Fig. 1a shows the effect of small optical misalignment on the spectrum of sample A. The scan rate used for these spectra was 0.83 cm^{-1} per channel and the spectrometer had a resolution of 1.5 cm^{-1}. As seen in Figs. 1b and 1c, the luminescence spectra of the two samples were quite different, but the two-magnon mode, denoted by M, appeared in both spectra at the same position. This M peak in Fig. 1 was reproducible, shifted when the laser frequency was varied, and gradually disappeared as the temperature was raised. On the other hand, the luminescence peaks, σ_{1L} (magnon sideband) and I (impurity modes), always appeared at fixed frequencies. The difference in the luminescence

spectra of the two samples is mainly due to different impurity concentrations. Sample A has a high Zn^{2+} concentration which gives rise to the impurity mode at 18384 cm^{-1} and the rising tail above 18363 cm^{-1} in Fig. 1b. Sample B has a higher Mg^{2+} concentration as evidenced by the strong impurity line at 18369 cm^{-1} in Fig. 1c. The σ_{1L} peak of sample A is almost twice more intense than that of sample B. This is probably due to more effective quenching of intrinsic luminescence by impurities in sample B.[4]

Figure 2 shows another set of spectra with the laser excitation at 18478 cm^{-1} which is in the valley between the σ_1 and σ_2 absorption bands. Again, Fig. 2a shows that no two-magnon mode was visible if the optical alignment was poor. With better alignment, then two two-magnon modes were seen on both samples as expected.[2] Their frequencies agreed well with those obtained earlier on sample B.[2]

The integrated intensities of the two-magnon modes were always much weaker than that of the σ_{1L} peak, more so in sample A, although the peak intensities might become comparable. In Ref. 2, we showed that the two-magnon modes could have a resonance enhancement of a factor of 7. This is of course true only when the raw data were corrected by absorption at the laser and Stokes frequencies.[6]

Rousseau et al[1] have also commented that omission of exciton-magnon interaction in our theoretical interpretation makes our results suspicious. Our answer is as follows. 1) Inclusion of exciton-magnon interaction may slightly shift and change the two-magnon modes, but should not cause the modes to disappear. It is therefore not a valid reason

to reject our observation. 2) In fluorescence only the magnon sideband of E_1 can be seen. Contrary to what is stated in Ref. 1, no one knows whether the sidebands of E_1 and E_2 have the same shape or not. 3) The stress-dependent luminescence experiment of Dietz et al.[5] shows the importance of exciton-magnon interaction, but as far as we know, there is no quantitative estimate on how strong the stress-induced mixing of E_1 and E_2 is. It is doubtful that an E_2 dispersion of 7 cm^{-1} can be detected in that experiment. 4) We stated in Ref. 2 that our results suggested a negigible dispersion of E_2 near the zone edges although the total dispersion appeared to be 7 cm^{-1}. This is not in contradiction to the results in Fig. 5 of Ref. 5.

We do not know exactly why Rousseau et al. could not observe the two-magnon modes. As suggested in Figs. 1 and 2, it could be the result of poor alignment, inadequate resolution, or insufficient signal-to-noise ratio. Multiple scattering of the strong luminescence in the crystal makes alignment of RRS in MnF_2 somewhat more difficult than in other crystals. In order to obtain reliable results, the experiment should certainly be done with utmost care.

This work was supported by the U. S. Energy Research and Development Administration.

REFERENCES

1. D. L. Rousseau, R. E. Dietz, and P. F. Williams, Phys. Rev. Lett., the preceeding paper.

2. N. M. Amer, T. C. Chiang, and Y. R. Shen, Phys. Rev. Lett. 34, 1454 (1975). (In Fig. 2, the second spectrum should read ω_ℓ = 18476.6 cm^{-1}, and the third spectrum has a different scan rate such that the two M modes are at 18370.5 cm^{-1} and 18380.5 cm^{-1}, see Erratum, to be published).

3. No two-magnon mode has yet been observed by Rousseau et al. in our sample.

4. R. E. Dietz, A. E. Meixner, H. J. Guggenheim, and A. Misetich, Phys. Rev. Lett. 21, 1067 (1968).

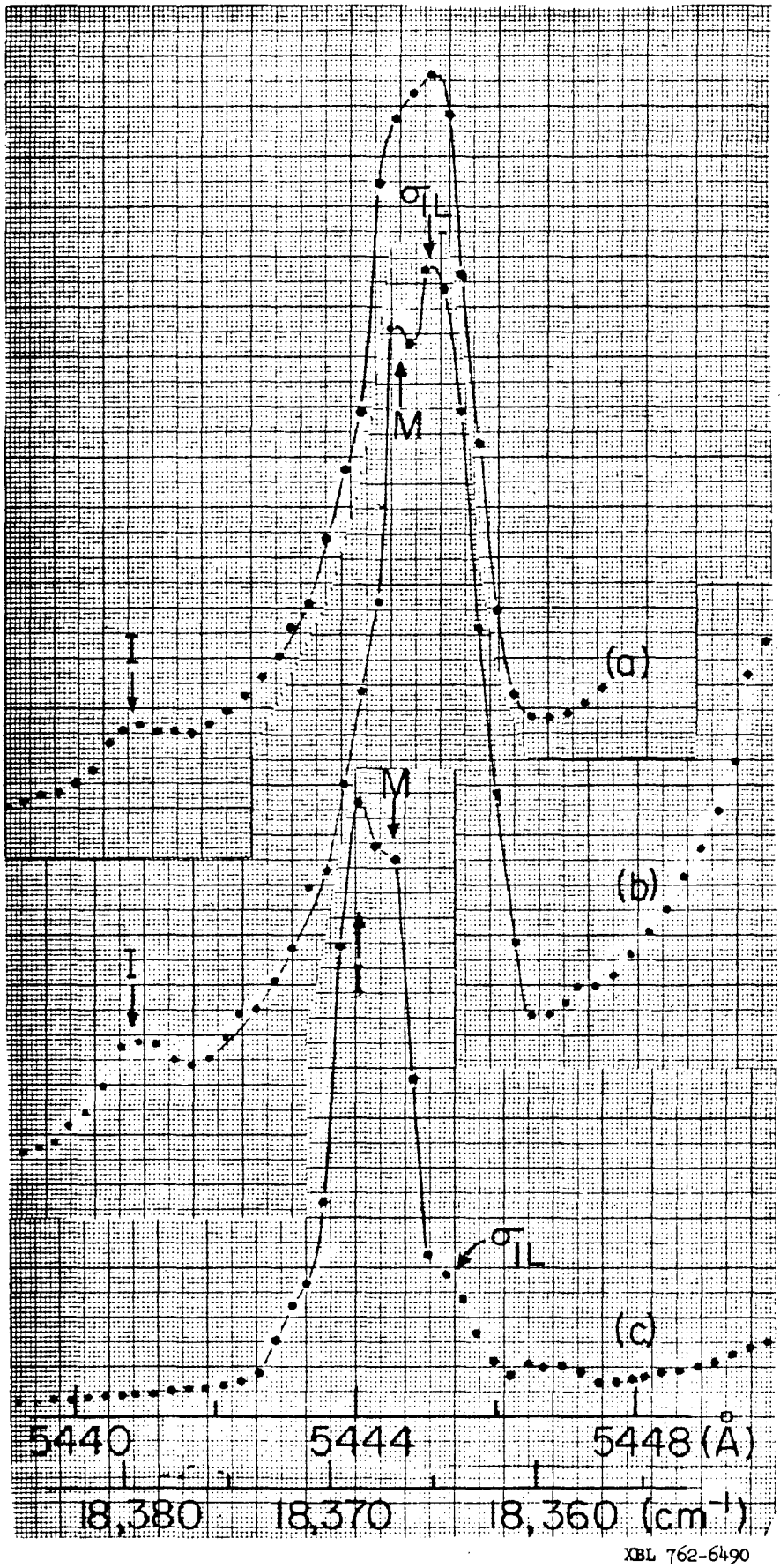
5. R. E. Dietz, A. E. Meixner, H. J. Guggenheim, and A. Misetich, J. Lumminesc. 1, 2, 279 (1970).

6. R. Loudon, J. Phys. Radium 26, 677 (1965).

FIGURE CAPTIONS

Fig. 1. Raman and luminescence spectra obtained with the laser excitation close to the σ_1 absorption peak at 18475.8 cm^{-1} ± 0.5 cm^{-1}. (a) Sample A with some misalignment; the peak counting rate was 6.8K counts/sec; (b) Sample A with good alignment; the peak counting rate was 8.2K counts/sec; (c) Sample B with good alignment; the peak counting rate was 12K counts/sec. The spectral resolution was 1.5 cm^{-1} and the scan rate was 0.24Å/channel.

Fig. 2. Raman and luminescence spectra obtained with the laser excitation at 18478.5 ± 0.5 cm^{-1} in the valley between the σ_1 and σ_2 absorption bands. (a) Sample A with some misalignment; (b) Sample A with good alignment; the peak counting rate was 4.4K counts/sec; (c) Sample B with good alignment; the peak counting rate was 11K counts/sec. The spectral resolution was 1.5 cm^{-1} and the scan rate was 0.24Å/channel.



XBL 762-6490

Fig. (1)

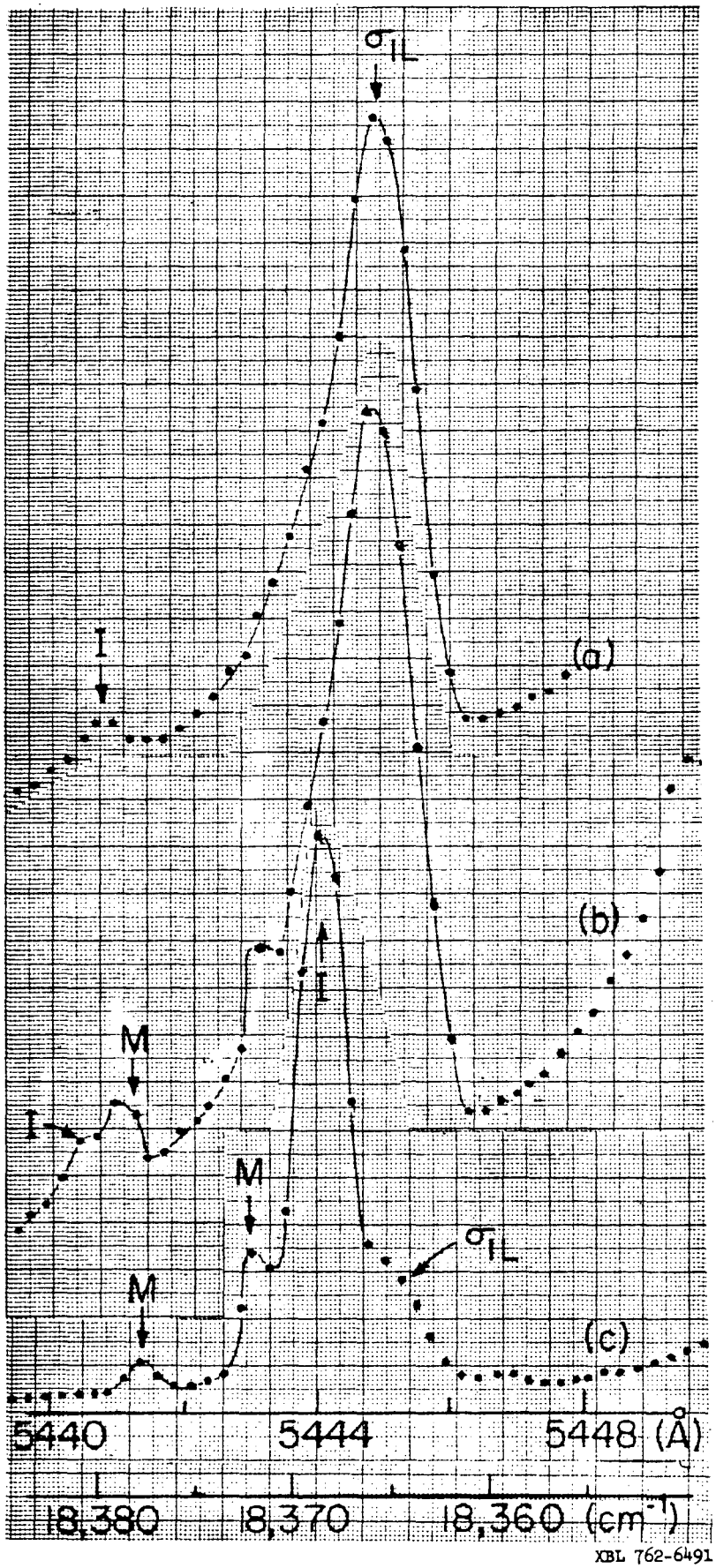

σIL
I
(a)
(b)
I
M
I
M
σIL
M
(c)
5440
5444
5448 (Å)
18,380
18,370
18,360 (cm⁻¹)
XBL 762-6491

Fig. (2)

TECHNICAL INFORMATION DIVISION
LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720