Temperature dependence of the optical exciton-magnon absorption lines in MnF_2 crystals

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The temperature dependence of the exciton and magnon sideband absorption has been investigated in MnF_2 crystals. A good agreement of the experimental data with Shinagawa and Tanabe's theoretical calculation has been obtained for each intensity of the cold and hot magnon sidebands. A general expression of the temperature dependence for the hot sidebands, which occur in one-, two-, and three-dimensional spin-coupling systems, is presented.

I. INTRODUCTION

MnF_2 has a rutile-type crystal structure and shows a three-dimensional (3D) antiferromagnetic spin coupling below 67.3 K ($=T_N$). Several broad absorption bands are observed in the vis-uv region below 550 nm as seen in Fig. 1. They are due to the $d^5 \rightarrow d^5$ transition in Mn^{2+} ions.[1] The integrated intensity of each band has a tendency to increase with increasing temperature. As shown in the inset of Fig. 1, the temperature dependence is different among the absorption bands: unlike the absorption band due to the ${}^6A_{1g} \rightarrow {}^4T_{1g}({}^4G)$ transition in Mn^{2+} ions (called the A band), the ${}^6A_{1g} \rightarrow {}^4T_{2g}({}^4G)$ band (B band) slightly increases at high temperatures. No theoretical work has been reported to explain such a different temperature dependence. The reason is that each band is composed of many lines due to excitons (i.e., zero-magnon, zero-phonon lines), excitons coupled with one and/or multiple magnons (i.e., exciton-magnon lines or magnon sidebands),[1] excitons coupled with phonons (i.e., exciton-phonon lines), and excitons coupled with both magnons and phonons (i.e., exciton-magnon-phonon lines), and the relative number of these components is different among the Mn^{2+} absorption bands. Tanabe and his collaborators, however, have derived the general formula for the magnon sidebands theoretically and the temperature dependence of the intensity, peak position, and half-width for the sidebands of MnF_2 and $RbMnF_3$.[2-4]

There are two kinds of magnon sidebands: one is a cold sideband, which decreases with increasing temperature, the other is a hot sideband, which grows with increasing temperature. So far, the temperature dependences of the peak position and half-width of the cold sideband observed in MnF_2 have been compared with the sublattice magnetization or with magnetic energy.[5-7] Although an agreement of the experimental data with the magnetization or magnetic energy was obtained, the experimental data should be compared with the theoretical calculation[3] which is made using three-spin and four-spin

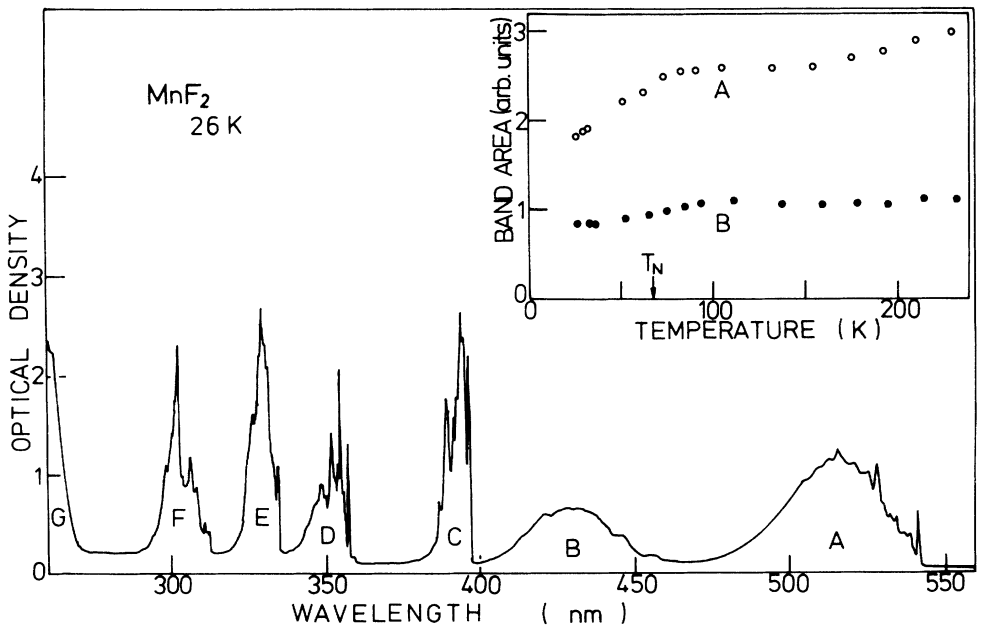

FIG. 1. Absorption spectrum of a MnF_2 crystal at 26 K which was taken using σ-polarized light. In the inset is shown the temperature dependence of the integrated intensity for each of the A and B Mn^{2+} absorption bands.

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correlation functions in addition to two-spin correlations. Recently Kleemann and Uhlig studied the intensity and half-width of an absorption band composed of the cold and hot sidebands in FeF_2.[8] The obtained temperature dependence is similar to the theoretical one expected for MnF_2 and $RbMnF_3$, but the exact comparison of the experimental data with the theoretical results is not possible because no calculation has been done for the temperature dependence of magnon sidebands in FeF_2.

Although a reliable theoretical calculation has only been made for MnF_2 and $RbMnF_3$,[2-4] the comparison of the experimental data with the theoretical prediction has not been made for each of the cold and hot sidebands in these crystals, except a rough comparison by one (T.T.) of the present authors[9] for the hot sideband of MnF_2. The rough comparison was inevitable, because the detailed temperature dependence for each of the cold and hot sidebands had not been measured. In this paper we examine the temperature dependence of the cold and hot sidebands in MnF_2 experimentally and compare it with the theoretical calculation.

II. EXPERIMENTAL RESULTS

Figure 2 shows the temperature dependence of fine structure appeared at the low-energy tail of the σ-polarized A band. Two weak lines, named $M1\sigma$ and $M2\sigma$, appear at 542.95 and 542.45 nm at 26 K, respectively, and two intense lines, $E1\sigma$ and $E2\sigma$, appear at 541.30 and 541.10 nm, in agreement with previous measurements.[5,10] The $M1\sigma$, $M2\sigma$, $E1\sigma$, and $E2\sigma$ lines were named $E1$, $E2$, $\sigma1$, and $\sigma2$, respectively, by Sell, Greene, and White,[5] but we follow the notation by Meltzer, Lowe, and McClure.[10] The letters M, E, and H which appear in this paper indicate the magnetic-dipole line, electric-dipole cold sideband, and electric-dipole hot sideband, respectively.

As temperature is increased, it is observed that the $M1$ and $M2$ lines vanish, and the $E1$ and $E2$ lines broaden and shift towards lower energies. The $E1$ and $E2$ lines are observed to show a large shift at low temperatures below 75 K but a small shift above 75 K. No anomaly is observed at T_N within experimental error. Unlike the case of $E1$ and $E2$ lines, the $M1$ and $M2$ lines do not shift as expected for the zero-magnon, zero-phonon exciton lines. When temperature is increased, a line named $H1\sigma$ is observed to appear at 544.40 nm and grow with temperature (see Fig. 2).

In Fig. 3, the total intensity (area) of the $E1\sigma$ and $E2\sigma$ lines are plotted against temperature, together with the integrated intensity of the $H1\sigma$ line. The $E1$ and $E1$ lines are observed to decrease with increasing temperature without showing any anomaly at T_N, while the $H1$ line is observed to grow with temperature below T_N but not above T_N. In the π-polarized absorption spectra, we also observed the cold sideband $E1\pi$ at 541.80 nm at 26 K and the hot sideband $H1\pi$ at 544.1 nm, in agreement with the previous result.[10] The thermal properties of the $E1\pi$ and $H1\pi$ lines were quite similar to those of the $E1\sigma$ (or $E2\sigma$) and $H1\sigma$ lines, respectively.

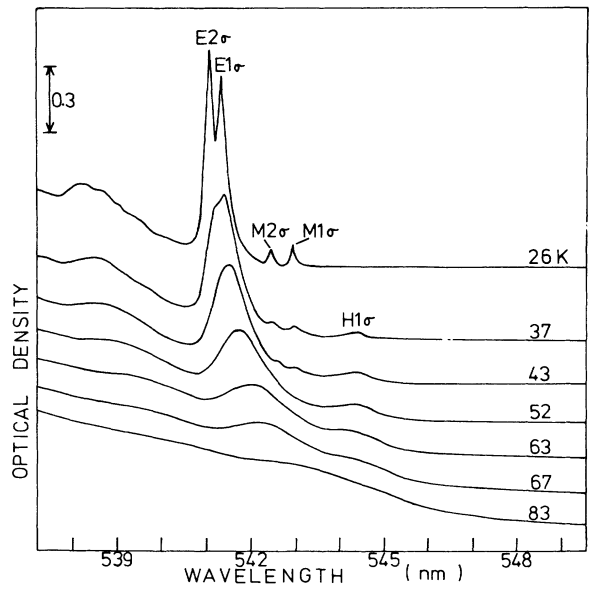

FIG. 2. Temperature variation of the fine structure which appears at the low-energy tail of the A band due to the ${}^6A_{1g} \rightarrow {}^4T_{1g}({}^4G)$ transition in Mn^{2+} ions. The spectra were obtained using σ-polarized light.

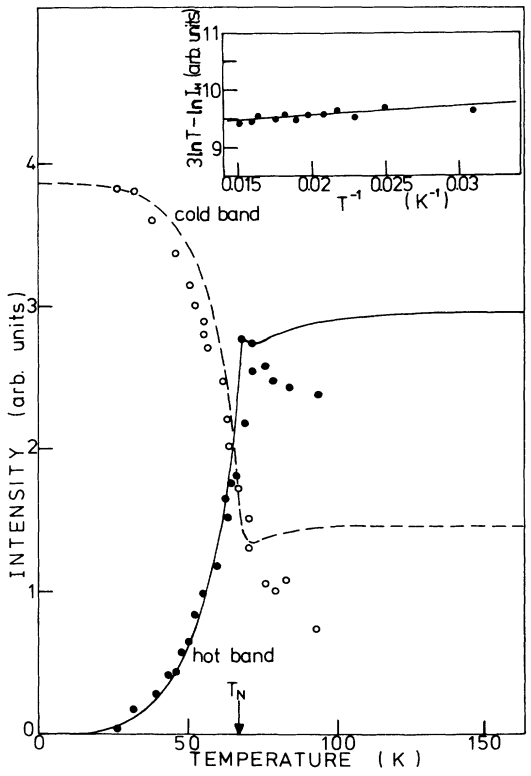

FIG. 3. Temperature dependence of the total intensity of the σ-polarized $E1$ and $E2$ cold sidebands (open circle) and of the σ-polarized $H1$ hot sideband (closed circle). Solid curves are obtained from the calculation by Shinagawa and Tanabe (Ref. 3) (see text). In the inset is shown $(3\ln T - \ln I_H)$ vs T^{-1}, where I_H is the hot-sideband intensity.

III. DISCUSSION

Shinagawa and Tanabe have calculated the temperature variation of the integrated intensity for each of the cold and hot sidebands in MnF_2.[3] We compare the temperature dependence of the experimentally obtained intensity for each of the hot and cold sidebands in MnF_2 with the theoretically obtained one. As seen in Fig. 3, a good agreement is found for both the hot and cold sidebands although disagreement is found at the high-temperature region above T_N. The temperature variation of our cold sideband is similar to the magnetic energy which has been obtained from magnetic linear birefringence,[11] while the calculated intensity reaches a minimum value near T_N and takes a constant value at high temperatures.[3] The discrepancy above T_N seems to be due to a difficulty in estimating the intensity exactly because of extreme broadening above T_N.

The temperature-dependence curve derived by Shinagawa and Tanabe[3] is difficult to describe using a simple function of temperature T, because it was obtained by numerical calculation. In the following, we try to derive a simplified function describing the T dependence of the hot-sideband intensity to understand the property of the hot sideband intuitively.

In a 2D spin-coupling system, it is known that the hot sideband is proportional to $T^2\exp(-A/kT)$, where A is the energy gap in the spin-wave dispersion, i.e., the magnon gap energy at the zone center.[12,13] In a 1D system, the hot sideband is proportional to T at low temperatures.[9,14,15] From these facts, we assume that the hot sideband in a 3D system has an intensity proportional to $T^3\exp(-A/kT)$. The T^3 dependence is not so unreasonable because Yen, Imbusch, and Huber and Kleemann and Uhlig have also expected it.[6,8] Additionally it was shown, from the comparison with the experimental result, that such an assumption is not unreasonable.[9] To check the validity of our assumption quantitatively, we plot $3\ln T - \ln I_H(T)$ against T^{-1} as shown in the inset of Fig. 3 where $I_H(T)$ means the hot-sideband intensity at a temperature T. A straight line can be drawn from the experimental data below T_N (see inset of Fig. 3). From the slope of the straight line, we obtain $A=9.72\ \mathrm{cm}^{-1}$. This value is close to $8.72\ \mathrm{cm}^{-1}$ of the magnon gap energy at the zone center for MnF_2.[16] Therefore it is concluded that the intensity of the hot sideband in the magnets with d-dimensional ($d=1,2,3$) spin coupling is approximately described by the T^d dependence law at low temperatures below T_N.

ACKNOWLEDGMENT

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[1]See, e.g., Y. Tanabe and K. Aoyagi, *Excitons,* edited by E. I. Rashba and M. D. Sturge (North-Holland, Amsterdam, 1982), p. 603.

[2]T. Fujiwara, W. Gebhardt, K. Pedanides, and Y. Tanabe, J. Phys. Soc. Jpn. **33**, 39 (1972).

[3]K. Shinagawa and Y. Tanabe, J. Phys. Soc. Jpn. **30**, 1280 (1971).

[4]T. Fujiwara and Y. Tanabe, J. Phys. Soc. Jpn. **32**, 912 (1971).

[5]D. D. Sell, R. Greene, and R. M. White, Phys. Rev. **158**, 489 (1967).

[6]W. M. Yen, G. F. Imbusch, and D. L. Huber, in *Optical Properties of Ions in Crystals,* edited by H. M. Crosswhite and H. W. Moos (Interscience, New York, 1967), p. 301.

[7]M. S. Seehra and S. Abumansoor, Solid State Commun. **56**, 97 (1985).

[8]W. Kleemann and R. Uhlig, J. Phys. Condens. Matter **1**, 1653 (1989).

[9]T. Tsuboi, Phys. Lett. **102A**, 138 (1984).

[10]R. S. Meltzer, M. Lowe, and D. S. McClure, Phys. Rev. **180**, 561 (1969).

[11]I. R. Jahn, Phys. Status Solidi B **57**, 681 (1973).

[12]D. J. Robbin and P. Day, J. Phys. C **9**, 867 (1976).

[13]S. E. Schnatterly and M. Fontana, J. Phys. (Paris) **33**, 691 (1972).

[14]K. Ebara and Y. Tanabe, J. Phys. Soc. Jpn. **36**, 93 (1974).

[15]R. Laiho and T. Tsuboi, J. Magn. Magn. Mater. **54-57**, 1265 (1986).

[16]W. M. Yen, L. D. Rotter, W. M. Dennis, W. Grill, and J. Wesner, *Excited States of Transition Elements,* edited by B. J. Trzebiatowska (World Scientific, Singapore, 1989), p. 636.