Physics of the Solid State, Vol. 45, No. 8, 2003, pp. 1500–1503. Translated from Fizika Tverdogo Tela, Vol. 45, No. 8, 2003, pp. 1429–1431.

MAGNETISM AND FERROELECTRICITY

Magnon Satellite Bands in the Optical Spectrum of Antiferromagnetic Rb_2MnCl_4

E. A. Popov* and S. G. Ovchinnikov**

** Siberian State Aerospace University, Krasnoyarsk, 660014 Russia*

*** Kirensky Institute of Physics, Siberian Division, Russian Academy of Sciences, Akademgorodok, Krasnoyarsk, 660036 Russia*

Received January 8, 2003

Abstract—The evolution of optical absorption in a two-dimensional antiferromagnet is investigated in the range of the transition ${}^6A_{1g} \longrightarrow {}^4A_{1g}, {}^4E_g({}^4G)$ observed in manganese ions in an external magnetic field inducing noncollinearity of the magnetic structure. It is revealed that hot and cold satellites of the exciton–magnon bands appear in the optical absorption spectrum and then increase in intensity. The shapes of the magnon satellite bands corresponding to a two-dimensional magnetic structure are calculated. It is demonstrated that magnons at the inner points of the Brillouin zone appreciably contribute to the absorption. The zero-point magnetic oscillations play a decisive role in the absorption associated with the magnon decay at low temperatures.

1. INTRODUCTION

As a rule, optical spectra of antiferromagnets containing ions with an open 3*d* shell are related to transitions forbidden with respect to the spin projection in the single-ion approximation. Consequently, in the optical spectra of these crystals, the intensive electrodipole absorption bands are caused by the excitation of groups of interacting ions, specifically by pairs of exchange-coupled ions belonging to different magnetic sublattices. The existence of single-magnon satellites of the exciton bands is characteristic of the absorption spectra of collinear antiferromagnets. In the case when the noncollinearity of the magnetic sublattices is induced by an external magnetic field, the optical transitions involving an even number of magnons become allowed [1]. The mechanism responsible for the formation of the light absorption bands can be elucidated by analyzing the dependences of the intensity and energy location of the absorption peak of the multimagnon satellites on the angle of canting of the magnetic sublattices. For example, the optical spectra of $RbMnCl_3$ and $CoCO_3$ compounds exhibit narrow isolated magnon satellite bands that correspond to the maxima observed in the density of exciton–magnon states at singular points of the Brillouin zone due to the participation of several magnons in light absorption [2] with the specific behavior in the magnetic field. In the present paper, we report results on the measurement of the optical absorption spectrum of an Rb_2MnCl_4 antiferromagnet in the frequency range of the transition ${}^6A_{1g} \longrightarrow {}^4A_{1g}, {}^4E_g({}^4G)$. The evolution of the absorption spectrum in the magnetic field was explained in terms of the two-dimensional magnetic structure responsible for the specific features in the density of states of quasiparticle combinations involved in optical excitations.

Crystals of Rb_2MnCl_4 at room temperature have a tetragonal structure with D_{4h}^{17} symmetry. At temperatures below the Néel point $T_N = 57$ K, the Rb_2MnCl_4 crystal is characterized by an antiferromagnetic order with easy-axis anisotropy. The magnetic moments are directed parallel to the C_4 symmetry axis of the crystal [3]. The exchange field is determined to be $H_E \approx 800$ kOe. The spin-flop transition occurs in the field $H_{SF} = 56$ kOe. The Mn^{2+} ions are located in layers perpendicular to the C_4 symmetry axis at sites of a plane square lattice of the crystal. The interlayer distance considerably exceeds the distance between the nearest neighbor Mn^{2+} ions located in the same layer. As a result, the intralayer exchange interaction between the Mn^{2+} ions is two orders of magnitude stronger than the interlayer exchange interaction, which causes the two-dimensional (2D) behavior of the magnetic system of the crystal.

2. EXPERIMENTAL TECHNIQUE

The optical absorption spectra of Rb_2MnCl_4 crystals were measured on a spectrometer with a resolution of 3 Å/mm. Magnetic fields with a strength up to 230 kOe were generated in a pulsed solenoid with a pulse duration of 20 ms. The spectra were recorded on photographic film.

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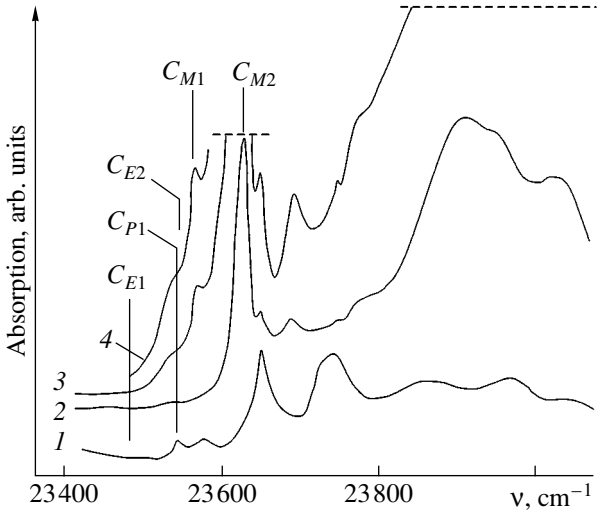

Fig. 1. Polarized spectra of Rb_2MnCl_4 crystals in the range of the transition ${}^6A_{1g} \longrightarrow {}^4A_{1g}, {}^4E_g({}^4G)$ at $T = 4.2$ K: (*1*) π polarization, $h = 2.2$ mm; (*2*) α polarization, $h = 0.14$ mm; (*3*) α polarization, $h = 0.5$ mm; and (*4*) σ polarization, $h = 1.51$ mm.

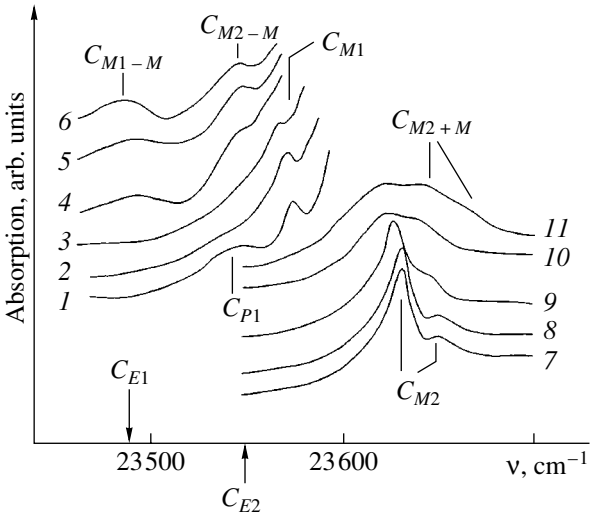

Fig. 2. Absorption spectra of Rb_2MnCl_4 crystals for the α polarization in magnetic fields *H*: (*1*, *7*) 0, (*2*, *8*) 46, (*3*) 70, (*4*, *9*) 135, (*5*, *10*) 160, and (*6*, *11*) 230 kOe. Sample thickness *h*: (*1*–*6*) 1.65 and (*7*–*11*) 0.08 mm. $T = 4.3$ K, $\mathbf{H} \parallel C_4$.

3. RESULTS AND DISCUSSION

The polarized absorption spectra of crystal samples with different thicknesses *h* at the temperature $T = 4.2$ K are shown in Fig. 1. According to the selection rules, the magnetodipole exciton bands in the optical spectra of the Rb_2MnCl_4 crystal should be observed for the polarizations α (the wave vector **k** is aligned parallel to the C_4 axis of the crystal, and the electric-field vector **E** is perpendicular to the C_4 axis) and π ($\mathbf{k} \perp C_4$, $\mathbf{E} \parallel C_4$), whereas the electrodipole single-magnon satellites of the exciton bands should manifest themselves for the polarizations α and σ ($\mathbf{k} \perp C_4$, $\mathbf{E} \perp C_4$). We failed to observe exciton bands in the spectra of crystal samples of the specified thickness. Vervoitte *et al.* [4] measured the spectrum of magnetic circular dichroism and revealed two dichroic magnetodipole bands (23489 and 23547 cm^{-1}). The locations of these bands are indicated in Fig. 1, and the bands themselves are designated as C_{E1} and C_{E2}, respectively, and are interpreted as exciton bands. In [4], the bands C_{P1} and C_{M1} were identified as photon and magnon satellites of the exciton band C_{E1}, respectively. The band C_{M1} is located at a distance of ~80 cm^{-1}, which corresponds to the magnon frequency at the boundary of the Brillouin zone.

We measured the absorption spectra of Rb_2MnCl_4 crystals in different magnetic fields and at temperatures ranging from 1.8 to 100 K. It turned out that the temperature dependences of the spectral characteristics do not provide useful information. As the temperature increases to the Néel point T_N, the half-width of the bands increases; consequently, it becomes impossible to determine the location of most of them to sufficient accuracy. The only isolated band, namely, C_{M2}, which can be clearly observed up to the temperature T_N and higher, does not undergo a noticeable shift at temperatures $T < T_N$. This is a typical manifestation of the two-dimensional magnetic structure of the crystal [5].

Figure 2 illustrates the evolution of the red edge of the α-polarized absorption spectra of thick and thin crystal samples in different magnetic fields at $T = 4.3$ K. In the case when the magnetic field exceeds the field of the spin-flop transition and the magnetic moments of the sublattices exhibit angularity, the spectrum undergoes a considerable transformation. It can be seen that an intense broad band, namely, C_{M2+M}, appears in the spectrum, increases in intensity, and overlaps with the C_{M2} band whose intensity decreases. As the magnetic field increases above 100 kOe, there appear two new bands, namely, C_{M1-M} and C_{M2-M}, at frequencies corresponding to pure exciton transitions and their intensities increase. Judging from the field dependences of the aforementioned bands, the C_{M2+M} band can be identified as an exciton–two-magnon band (a magnon satellite of the exciton–magnon band C_{M2}), whereas the C_{M1-M} and C_{M2-M} bands can be treated as hot satellites of the exciton–magnon bands. The integrated intensity of these bands should be proportional to $\sin^2 2\theta \sin^4\theta$, where $\cos\theta = H/2H_E$ and θ is half the angle between the sublattices. However, in the former case, the band is unusually broad. In the latter case, the intensities of the bands should be proportional to the magnon population [6]: $n = [\exp(\varepsilon_m/kT) - 1]^{-1}$, which is small at the temperature of the experiment; hence, these bands should not be observed. In order to explain the above facts, the specific features of the magnetic structure of the crystal must be taken into account.

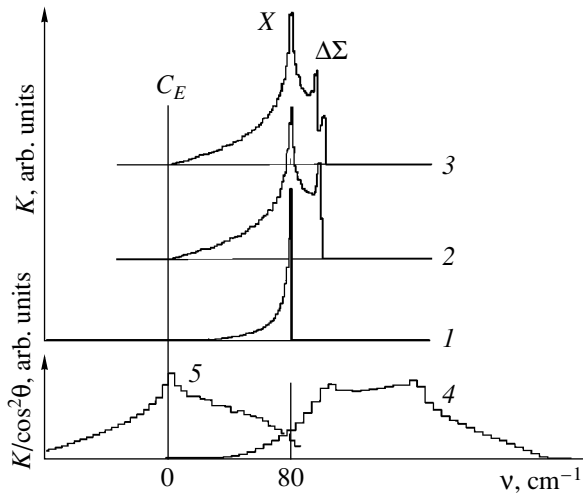

Fig. 3. Calculated band shapes: (*1*) the exciton–magnon band at $K_1 = 0$, $K_2 = 0$; (*2*) the exciton–magnon band at $K_1 = 0$, $K_2 = 2.5$; (*3*) the exciton–magnon band at $K_1 = 0.1$, $K_2 = 2.5$; (*4*) the cold magnon satellite of the exciton–magnon band at $K_1 = 0$, $K_2 = 2.5$; and (*5*) the hot magnon satellite of the exciton–magnon band at $K_1 = 0$, $K_2 = 2.5$.

The shapes of the absorption bands were calculated taking into account the results obtained in [6]. The absorption coefficients at the frequency ω were calculated for the exciton–magnon band from the relationship

$$K^{e+m}(\omega) \propto \sum_{\mu,\,\mathbf{k}} |\mathbf{P}(\mathbf{k})|^2 \Phi_\mu^{e+m}(\mathbf{k})\delta(\omega - E_\mu(\mathbf{k}) - \varepsilon_\mu(-\mathbf{k})),$$

for the cold magnon satellite of the exciton–magnon band from the expression

$$K^{e+m+m}(\omega) \propto \sum_{\mu,\,\mathbf{k}} |\mathbf{P}(\mathbf{k})|^2 \Phi_\mu^{e+m+m}(\mathbf{k})\delta$$
$$\times\left(\omega - E_\mu(\mathbf{k}) - \sum_{\mathbf{k}_1 - \mathbf{q} = -\mathbf{k}}\left(\varepsilon_\mu(\mathbf{k}_1) + \sum_{\mathbf{q}}\varepsilon_\mu(\mathbf{q})\right)\right),$$

and for the hot magnon satellite of the exciton–magnon band from the formula

$$K^{e+m-m}(\omega) \propto \sum_{\mu,\,\mathbf{k}} |\mathbf{P}(\mathbf{k})|^2 \Phi_\mu^{e+m-m}(\mathbf{k})\delta$$
$$\times\left(\omega - E_\mu(\mathbf{k}) - \sum_{\mathbf{k}_1 - \mathbf{q} = -\mathbf{k}}\left(\varepsilon_\mu(\mathbf{k}_1) - \sum_{\mathbf{q}}\varepsilon_\mu(\mathbf{q})\right)\right).$$

Here, $\mathbf{P}(\mathbf{k})$ is the dipole moment of the transition in the $\mathbf{k}$ space and $\Phi_\mu^{e+*}(\mathbf{k})$ are the functions dependent on the direction of the magnetic moments of the sublattices, the population of magnon states, and the structure of the crystal. The summation is carried out over all vectors $\mathbf{k}$ of the Brillouin zone and over the zones μ. The exciton energy $E_\mu(\mathbf{k})$ and magnon energy $\varepsilon_\mu(\mathbf{k})$ were calculated in the nearest neighbor approximation according to the expressions

$$E_\mu(\mathbf{k}) = \Delta\tilde{\varepsilon} + A\cos\theta\tilde{z}\tilde{M}\tilde{\gamma}(\mathbf{k}) - (-1)^\mu z|M|\gamma(\mathbf{k})\cos^2\theta,$$

$$\varepsilon_\mu(\mathbf{k})$$
$$= SI(0)\sqrt{1 - \gamma^2(\mathbf{k}) + 2\cos^2\theta\gamma(\mathbf{k})[\gamma(\mathbf{k}) - (-1)^\mu]},$$

$$\gamma(\mathbf{k}) = \cos\left(\frac{ak_x}{2}\right)\cos\left(\frac{ak_y}{2}\right),$$

$$\tilde{\gamma}(\mathbf{k}) = \frac{1}{2}(\cos ak_x + \cos ak_y),$$

where a is the lattice parameter. In contrast with the case considered in [6], we ignored the renormalization of the exciton energy due to the exciton–magnon interaction Δ and assumed that $|\Delta(\mathbf{k}, \mu)|^2 = \Delta\sin^2 2\theta$. The role of the exciton–magnon interaction, in our case, consists in resolving the magnon satellites of the exciton–magnon band. All the other designations correspond to those presented in [2]. In our calculations, we varied the parameters $K_1 = |M|\cos^2\theta$ and $K_2 = \tilde{M}$ determining the transfer of excitation inside and between the sublattices.

The calculated shapes of the exciton–magnon band and its cold single-magnon satellite having characteristic weakly pronounced peaks associated with the specific features of the density of states corresponds to the observed shape under the condition $K_2 = 2.5|J|$ (Fig. 3). Here, $|J|$ is the quantity characterizing the intralayer exchange between the nearest neighbors. The magnitude $|M|$ can be determined from the Davydov splitting of the exciton band. We failed to do this because no weak exciton band was observed for crystals of the specified thickness. For this reason, in our calculations, we chose small values of K_1 in units of $|J|$, which is specified by the range of the magnetic fields used. In this case, the shapes of the bands change insignificantly, whereas the maxima of the intensity should vary as $\sin^4\theta$ and $\sin^2 2\theta\sin^4\theta$ for the single-magnon and two-magnon satellites, respectively. Figure 3 displays the calculated shape of the two-magnon satellite for small values of K_1, which is described by a function $\sim K^{e+m+m}(\omega)/\cos^2\theta$, because when $H = 0$ and $\theta = \pi/2$ (and, correspondingly, $K_1 = 0$), we obtain $K^{e+m+m}(\omega) = 0$. An increase in the fraction of magnons with wave vectors at points of the general type, as compared to that at singular points in a 2D antiferromagnet, gives rise to a broad two-magnon band.

Unlike the cold magnon satellite of the exciton–magnon band, the hot magnon satellite of this band exhibits a sharp peak on the background of the wide dome at a frequency close to the frequency of the exci-

ton (Fig. 3) for $K_2 = 2.5$ and $K_1 = 0$. Consequently, the bands C_{M-M1} and C_{M-M2} are relatively narrow. They are characterized by a high intensity and a weak temperature dependence due to the two-dimensional magnetic structure. According to [6], we can write the relationship $\Phi_{\mu}^{e+m-m}(\mathbf{k}) \sim n_{\mu}(\mathbf{k})$. The decrease in the magnetization due to zero-point oscillations was calculated for a two-dimensional Heisenberg antiferromagnet in [7]. It was shown that this decrease at $T = 0$ reaches ~20% [7]. Therefore, at a low temperature, n takes on a value of no less than 0.2, which ensures a sufficient number of magnons for the participation in the hot absorption of light. The shape of the band was calculated under the assumption that magnons are uniformly distributed in the Brillouin zone.

Upon summing the band intensities with weights dependent on the sublattice angularity in the magnetic field, we obtain a spectrum similar to that observed in the experiment.

ACKNOWLEDGMENTS

This work was supported in part by the Russian Federal Program “Integration,” project no. 0017.

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Translated by O. Moskalev