

FINE STRUCTURE OF EXCITON-MAGNON ABSORPTION OF LIGHT IN KMnF_3

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Some peculiarities of the structure of pure exciton and exciton-magnon light absorption bands in the region of the ${}^6A_{1g}({}^6S) \rightarrow {}^4A_{1g}{}^4E_g({}^4G)$ transition in weakly ferromagnetic KMnF_3 are observed and analyzed. It is shown that the splitting of pure exciton lines and the variation of the magnon frequencies at points W and U in the Brillouin zone make an additive contribution to the change of the exciton-magnon absorption band in a magnetic field. This may be regarded as proof of the weakness of the exciton-magnon interaction for the ${}^6A_{1g}({}^6S) \rightarrow {}^4A_{1g}{}^4E_g({}^4G)$ transition in KMnF_3 . The conclusions are valid for a similar transition in RbMnF_3 .

EXCITON-magnon bands have in recent years been observed in the absorption spectra of light of most investigated antiferromagnetic crystals. The problem of the fine structure of these bands has practically not been touched upon in the literature. It should, however, be noted that the nature of the fine structure of the exciton-magnon absorption attests to the general nature of its origin in a series of magnetically ordered crystals in which it has been observed (RbMnF_3 , KMnF_3 , and CsMnF_3). Therefore the study of the nature and features of this structure for one of these crystals would facilitate the understanding of its origin for a whole series of compounds.

The present paper is devoted to a detailed investigation of the fine structure of pure exciton and exciton-magnon absorption using the example of the ${}^6A_{1g}({}^6S) \rightarrow {}^4A_{1g}{}^4E_g({}^4G)$ transition (the C group of bands) in the spectrum of antiferromagnetic KMnF_3 which is weakly ferromagnetic. Because of the relatively weak magnetic anisotropy, it is possible to vary the magnetic structure of this crystal and the magnon spectrum by not too strong a magnetic field. It is precisely this circumstance which made it possible to understand the nature of the fine structure of the exciton-magnon bands and its behavior in an external magnetic field.

The method of the low-temperature investigation of crystals in a magnetic field using polarized light has been discussed previously.^[1] The absorption spectrum was recorded with a DFS-8 diffraction spectrograph having a linear dispersion of 3 Å/mm at $T = 4.2^\circ\text{K}$. The magnetic field was varied between 0 and 25 kOe with the aid of an SP-47 electromagnet. An Ahrens polarizer was used to polarize the light.

The particulars of growing the KMnF_3 single crystal and the results of an x-ray structure analysis of the investigated sample have been presented in^[2].

1. THE CRYSTAL AND MAGNETIC STRUCTURE OF KMnF_3

At room temperature KMnF_3 has a perovskite type structure which it retains down to $T = 184^\circ\text{K}$ (space group O_h^1).^[3] A series of tetragonal lattice distortions takes place as the temperature is further decreased. At $T < 81^\circ\text{K}$ the crystal has a distorted perovskite struc-

ture with the space group D_{2h}^{16} . The distortions manifest themselves in weak tetragonality ($c/a = 0.9985$) along with a tilting and rotation of the octahedra surrounding the Mn^{2+} ions (Fig. 1). The octahedra for the magnetic ions 1 and 3 and 2 and 4 are respectively tilted in opposite directions along the x axis. However, all octahedra in one column (along z) are rotated to the same side. Thus there are four types of pseudocells in the crystal. The octahedra surrounding, for example, the ions 2 and 4 are tilted to one side but rotated about the z axis in opposite directions.

In the temperature range of $88\text{--}81^\circ\text{K}$ KMnF_3 is a collinear antiferromagnet.^[3,4] At lower temperatures the collinearity of the magnetic moments of the sublattices is destroyed and a phase transition takes place to the state with weak ferromagnetism. Regardless of the fact that the space group permits four sublattices, Heeger et al.^[4] have provided evidence for a two-sublattice model for the weakly ferromagnetic KMnF_3 (for more detail, see Sec. 3 of this article). At $T = 4.2^\circ\text{K}$ the magnetic structure belongs to the so-called G-type configuration in which the two sublattices form two interpenetrating face-centered sublattices with opposing spins, producing a magnetic lattice with a NaCl type structure. The magnetic primitive unit cell contains two structural units of KMnF_3 .

2. PECULIARITIES OF THE BEHAVIOR OF THE FINE STRUCTURE OF EXCITON-MAGNON BANDS IN A MAGNETIC FIELD

The absorption spectrum of KMnF_3 in the region of the C group of bands is shown in Fig. 2. The results of previous work^[5-7] have made it possible to connect the weak magnetic dipole lines C_0 and C_3 with the pure exciton transitions ${}^6A_{1g}({}^6S) \rightarrow {}^4E_g({}^4G)$ and ${}^6A_{1g}({}^6S) \rightarrow {}^4A_{1g}({}^4G)$ in Mn^{2+} respectively. The shape and the temperature dependence of the frequencies of the electric dipole bands C_1 and C_4 made it possible to consider them exciton-magnon satellites of the bands C_0 and C_3 respectively, differing from the latter by the magnitude of the maximum of the magnon frequency at the Brillouin zone boundary $\nu_m \sim 80\text{ cm}^{-1}$.

The features of the C_2 band did not contradict the assumption that it is a two-magnon satellite of the C_0 band.

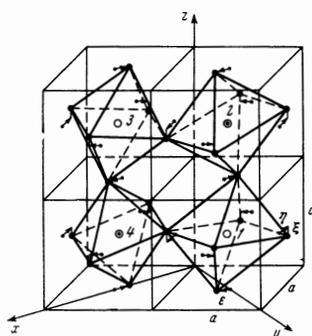


FIG. 1. Distortions of the fluorine octahedra for four neighboring Mn^{2+} ions in the $KMnF_3$ lattice; \circ and \ominus — Mn^{2+} ions with spin orientations — and + respectively; \bullet — F^- ions; $\eta = 0.060a$, $\epsilon = 0.05a$, $\xi = 0.06a$.^[3]

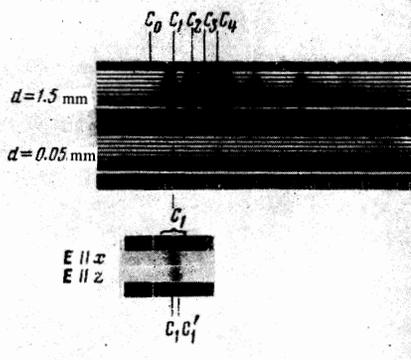


FIG. 2. Absorption spectrum of light of $KMnF_3$ in the region of the C group of bands for crystals of various thicknesses d in unpolarized light, $T = 4.2^\circ K$. The doublet C_1/C_1' is shown with polarized light, on a magnified scale (E is the electric vector of the linearly polarized light).

The separation between these bands, 140 cm^{-1} , is less than $2\nu_m \sim 160\text{ cm}^{-1}$; this can be related to a distortion of the maximum magnon frequency due to the strong exciton-magnon interaction in this complex three-particle process.

These conclusions were drawn on the basis of investigations of the spectrum of $KMnF_3$ with unpolarized light. The investigation of the spectrum with polarized light, undertaken for the first time in this work, has shown that the C_1 band is a doublet of sharply polarized bands C_1/C_1' , the separation between which is $\Delta = 2.5\text{ cm}^{-1}$. Figure 3a shows the behavior of this separation as a function of the intensity and orientation of an external magnetic field relative to the entire symmetry of the crystal. The frequencies of the C_1/C_1' doublet do not react to the external magnetic field parallel to the z axis of the crystal up to $H = 20\text{ kOe}$; on further increasing the field, the separation Δ decreases somewhat. A complicated dependence of the separation Δ on the intensity of the external magnetic field is observed in the case when $H \parallel x$.

The behavior of the frequency of the magnetic dipole band C_0 in a magnetic field is considerably simpler (Fig. 3b). In the absence of a magnetic field one observes a single unpolarized band C_0 ($\nu = 25134.3\text{ cm}^{-1}$) which does not react to an increase of the external magnetic field parallel to the z axis. For the case $H \parallel x$ this band splits into a doublet of unpolarized lines, the maximum interval between which is reached in a field $H \sim 6\text{ kOe}$, and does not change on further increasing the field.

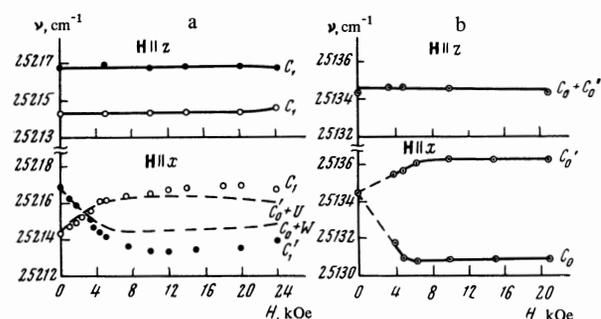


FIG. 3. Dependence of the frequencies of the C-group bands on the intensity of the external magnetic field for two orientations relative to the symmetry axes of the $KMnF_3$ crystal: a — for the doublets of the electric dipole bands C_1/C_1' ; b — for the magnetic dipole band C_0 . The points show the experimental results for $T = 1.35^\circ K$: \bullet — the electric vector E of linearly polarized light is parallel to the z axis, \circ — $E \parallel x$. The dashes show the calculated curves; one of these is a sum of the frequencies of the C_0 band and of the magnon frequency at the point W, corresponding to a given external field, the other is the sum of the frequencies of the C_0 band and of the magnon frequency at the point U (see Sec. 3).

3. DISCUSSION

In discussing the experimental results we start out from the previously accepted identification of the bands: C_0 is a pure exciton transition and the C_1/C_1' doublet corresponds to exciton-magnon transitions. Knowing the local symmetry group of the ion absorbing the light and the factor group of the magnetically ordered crystal, we can find by Loudon's scheme^[8] the fundamental properties of the pure exciton and exciton-magnon absorption bands. Before we carry out a group-theory analysis for the case of interest, we shall dwell in more detail on the problem of the possible spin configurations of weakly ferromagnetic $KMnF_3$.

The appearance of a weak ferromagnetic moment connected with the destruction of the collinearity of the magnetic moments of the sublattices can be due either to the single-ion anisotropy or to the antisymmetric spin interaction (Dzyaloshinskii interaction).^[9] The microscopic theory of the destruction of the collinearity of the magnetic moments as an illustration of the case of single-ion anisotropy was developed for the case of $KMnF_3$ from the following considerations.^[4,10] The low symmetry of $KMnF_3$ connected with distortions of the octahedra produced by the fluorine ions leads to the appearance of single-ion anisotropy. It is readily shown that the potential of the crystalline field coincides for ions 1 and 3 but differs from the potential for ions 2 and 4 (Fig. 1). From general considerations of the spin Hamiltonian the energy of the single-ion anisotropy is an expansion in powers of spin components and transforms in the same way as the potential of the crystalline field. In this connection the anisotropy energy will be the same for the pair of ions 1 and 3 but will differ from the analogous energy of the pair 2 and 4. This entitled Heeger, Beckman, and Portis^[4] to consider $KMnF_3$ as a two-sublattice antiferromagnet with different single-ion anisotropy energies for the two sublattices. The latter leads to a destruction of the collinearity of the magnetic moments of the sublattices due to the fact that the easy directions of their spins do not coincide.

The Hamiltonian for two-sublattice KMnF_3 with account of the symmetry of the positions of the fluorine ions can be written as follows:

$$\mathcal{H} = - \sum_{l \neq l'} J_{11} S_l S_{l'} - \sum_{m \neq m'} J_{11} S_m S_{m'} + \sum_{l \neq m} J_{12} S_l S_m - A_1 \left(\sum_l S_{lx}^2 + \sum_m S_{mx}^2 \right) - A_2 \left(\sum_l S_{lx} S_{lz} - \sum_m S_{mx} S_{mz} \right) - g \mu \mathbf{H} \left(\sum_l S_l + \sum_m S_m \right), \quad (1)$$

where the first three terms describe the intra and inter exchange interaction between ions of the l and m sublattices (all exchange integrals are positive), the fourth term gives the usual single-ion anisotropy, the nature of which has been discussed in detail in^[11], the fifth term is connected with the difference in the single-ion anisotropy of the ions of the two sublattices, and the last term is the Zeeman energy.

The problem of the determination of the fundamental states and spectra of the spin waves for KMnF_3 in an external magnetic field has been presented in full in^[12] using the usual methods of quantum mechanics.^[13] In this paper we present only the basic results of the calculation.

The two possible configurations with weak ferromagnetism in the absence of an external magnetic field are shown in Figs. 4a and b. The state of type b has the lowest energy. Therefore, in the absence of an external magnetic field the state should be realized in which the magnetic moments of the sublattices orient themselves close to the x axis and deviate from it in such a way that the weak ferromagnetic moment is parallel to the z axis. This is in agreement with the experimental data.^[4] The state a, which is close to state b in its energy, is readily realized with the aid of an external magnetic field $H_{\text{cr}} = 6400$ Oe.^[12] In this connection we shall analyze the main properties of the exciton and exciton-magnon absorption bands of KMnF_3 at special points of the Brillouin zone both for the magnetic configuration a as well as for b¹⁾.

The Brillouin zone of paramagnetic KMnF_3 is a cube with sides $2\pi/a$. In the ordered state when the unit cell doubles the volume of the Brillouin zone is conserved.

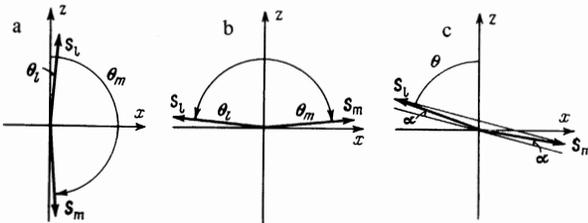


FIG. 4. Possible configurations with a weak magnetic moment for KMnF_3 in the absence of an external magnetic field: a - $\theta_l + \theta_m = \pi \tan(\theta_l - \theta_m) = A_2/(12J_{12} + A_1)$; b - $\theta_l + \theta_m = 0$, $\tan(\theta_l - \theta_m) = -A_2/(12J_{12} - A_1)$. Case c - rotation of the magnetic moments of the sublattices from the configuration b in an external magnetic field parallel to the x axis.

¹⁾The complete group-theoretical analysis of exciton and exciton-magnon processes in weakly ferromagnetic KMnF_3 is given in^[14]. The space groups of the crystal and the local groups of the Mn^{2+} ions are presented for the cases in which the spins are oriented parallel to the z and x axes. The types of exciton and exciton-magnon transitions, as well as the selection rules for their observation at all special points of the magnetic Brillouin zone, are determined.

Its shape is close to that of a truncated octahedron and is practically identical with the Brillouin zone of a face-centered cubic structure whose symmetry has been discussed in^[15] (the small deviation from a regular octahedron is connected with the fact that a is somewhat larger than c).

The local group of the Mn^{2+} ion for the configurations a and b is C_i ; the factor group of the crystal $D_{2h}(C_{2h})$ also coincides for both configurations. The wave functions of the Mn^{2+} ion transform like the Γ_2^+ representation of the group C_i . The process of the production of excitons from single-ion excitations for the two Mn^{2+} ions at the $\Gamma(0, 0, 0)$ point of the Brillouin zone ($\mathbf{k} = 0$) is shown schematically in Fig. 5. The excitons and magnons are transformed by the same representations of the factor group of the crystal. Since none of the representations of the factor group $D_{2h}(C_{2h})$ has any additional degeneracy connected with a time-reversal operation, all the excitons are nondegenerate and are components of the Davydov splitting. Both excitons are active in the magnetic dipole process: Γ_1^+ in the case when the magnetic vector \mathbf{h} of linearly polarized light is parallel to the x axis of the crystal, and Γ_2^+ —when $\mathbf{h} \parallel z$ and y .

Experimentally one observes for $H = 0$ a single, somewhat diffuse, unpolarized magnetic dipole line which practically does not react to a switching on of a magnetic field of intensity up to 25 kOe along the z axis and splits into a doublet when the external field is oriented along the x axis (Fig. 3b). Here we shall not discuss in detail the splitting of the pure exciton band C_0 . We merely note that its behavior when the KMnF_3 crystal is placed in an external magnetic field is analogous to the behavior of the 25143.5 cm^{-1} magnetic dipole line in the RbMnF_3 spectrum.^[16] On the other hand, we use the obtained experimental results concerning the behavior of the C_0 band in an external magnetic field in discussing the behavior of the fine structure of the C_1/C_1' doublet.

The irreducible representations of excitons and magnons for special points of the Brillouin zone consistent with the corresponding representations at the point $\Gamma(0, 0, 0)$ for the magnetic configuration b are shown in Fig. 6. The selection rules for double transitions in

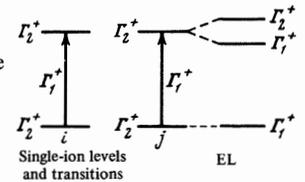


FIG. 5. Exciton production scheme from single-ion excitations for Mn^{2+} in KMnF_3 ; EL - exciton levels for $\mathbf{k} = 0$.

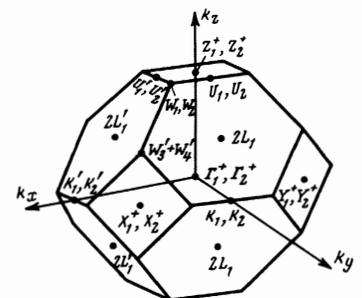


FIG. 6. Irreducible representations of excitons and magnons for symmetric points of the Brillouin zone of KMnF_3 , consistent with the corresponding representations at the point $\Gamma(0,0,0)$ for the case when the magnetic moment is parallel to the x axis of the crystal.

which excitations at points with wave vectors \mathbf{k} and $-\mathbf{k}$ at the boundary of the Brillouin zone participate simultaneously (see the table) were obtained by the method of Lax and Hopfield.^[17] It follows from the table that the contribution to the electric dipole absorption can only be due to the points $W(\pi/2a, 0, \pi/c)$, $K(3\pi/4a, 3\pi/4a, 0)$, and $U(\pi/4a, \pi/4a, \pi/c)$ of the Brillouin zone. Only these points turn out to be active also in the case of the magnetic configuration a.^[14]

In order to explain the nature of the separation Δ between the components of the C_1/C'_1 doublet we shall attempt to estimate the energy of the spin waves at the points W, K, and U for various directions of the magnetic moments of the sublattices. The two branches of the spin waves for the basic spin configuration b for $H = 0$ are of the form^[12]

$$\begin{aligned} (E_1/g\mu)^2 &= [\gamma_{11}(\mathbf{k}) + \gamma_{12}(\mathbf{k})][\gamma_{11}(\mathbf{k}) - \gamma_{12}(\mathbf{k}) \cos 2\theta_i] \\ &\quad + \gamma_{11}(\mathbf{k})(H_E + 2H_{A_1} + 5H_{A_2} \sin \theta_i) \\ &\quad + \gamma_{12}(\mathbf{k})(-H_E \cos 2\theta_i + 2H_{A_1} + 3H_{A_2} \sin \theta_i) \\ &\quad + (2H_E H_{A_1} + 4H_{A_2}^2)[1 + (H_{A_1}/H_E)^2], \\ (E_2/g\mu)^2 &= [\gamma_{11}(\mathbf{k}) - \gamma_{12}(\mathbf{k})][\gamma_{11}(\mathbf{k}) + \gamma_{12}(\mathbf{k}) \cos 2\theta_i] \\ &\quad + \gamma_{11}(\mathbf{k})(H_E + 2H_{A_1} + 3H_{A_2} \sin \theta_i) \\ &\quad + \gamma_{12}(\mathbf{k})(-H_E - H_{A_2} \sin \theta_i + 2H_{A_1} \cos 2\theta_i) \\ &\quad + (2H_E H_{A_1} + H_{A_2}^2)[1 + 2(H_{A_2}/H_E)^2], \end{aligned} \quad (2)$$

where

$$\begin{aligned} \gamma_{11}(\mathbf{k}) &= \frac{8J_{11}S}{g\mu}[3 - Y_{11}(\mathbf{k})], \quad \gamma_{12}(\mathbf{k}) = \frac{4J_{12}S}{g\mu}[Y_{12}(\mathbf{k}) - 3], \\ Y_{11}(\mathbf{k}) &= \cos k_x a \cos k_y a + \cos k_x c + \cos k_y c, \\ Y_{12}(\mathbf{k}) &= \cos k_x a + \cos k_y a + \cos k_z c; \\ H_E &= \frac{24J_{12}S}{g\mu} = 1,6 \cdot 10^6 \text{ Oe}, \quad H_{A_1} = \frac{A_1 S}{g\mu} = 2 \text{ \AA}, \quad H_{A_2} = \frac{A_2 S}{g\mu} = 1300 \text{ \AA}. \end{aligned} \quad (3)$$

The last terms in both expressions (2) give the anti-ferromagnetic resonance frequency ($\mathbf{k} = 0$).

From Eqs. (2) one can readily show with the aid of the known parameters (3) that for $H = 0$ the frequencies

Product of representations of points on the Brillouin zone boundary with wave vectors \mathbf{k} and $-\mathbf{k}$ (magnetic configuration b)	Expansions in representations of the factor group $D_{2h}(C_{2h})$	Polarization*	
		MD process	ED process
$X_i^+ X_i^+ = Y_i^+ Y_i^+ = Z_i^+ Z_i^+ (i=1, 2)$	Γ_1^+	$\mathbf{h} \parallel z$	
$Z_1^+ Z_2^+ = X_1^+ X_2^+ = Y_1^+ Y_2^+$	Γ_2^+	$\mathbf{h} \parallel x, y$	
$W_i W_i (i=1, 2)$	Γ_1^+	$\mathbf{h} \parallel z$	
$W_1 W_2$	Γ_2^+	$\mathbf{h} \parallel x, y$	$\mathbf{E} \parallel z$
$W_i' W_i' (i=3, 4)$	Γ_1^+	$\mathbf{h} \parallel z$	
$W_3' W_4'$	Γ_2^+	$\mathbf{h} \parallel x, y$	$\mathbf{E} \parallel x, y$
$L_1 L_1 = L_1' L_1'$	Γ_1^+	$\mathbf{h} \parallel z$	
$L_1 L_1 = L_1' L_1'$	Γ_2^+	$\mathbf{h} \parallel x, y$	$\mathbf{E} \parallel z$
$K_i K_i = K_i' K_i' = U_i U_i = U_i' U_i' (i=1, 2)$	Γ_1^+	$\mathbf{h} \parallel z$	
$K_i K_i = K_i' K_i' = U_i U_i = U_i' U_i' (i=1, 2)$	Γ_2^+	$\mathbf{h} \parallel x, y$	$\mathbf{E} \parallel x, y$
$K_1 K_2 = K_1' K_2' = U_1 U_2 = U_1' U_2'$	Γ_1^+	$\mathbf{h} \parallel z$	
$K_1 K_2 = K_1' K_2' = U_1 U_2 = U_1' U_2'$	Γ_2^+	$\mathbf{h} \parallel x, y$	$\mathbf{E} \parallel z$

*Here MD and ED are magnetic dipole and electric dipole processes respectively; \mathbf{E} and \mathbf{h} are the electric and magnetic vectors of the linearly polarized light; $x, y,$ and z are the symmetry axes of the crystal (Fig. 1).

of the branches E_1 and E_2 coincide at any point on the boundary of the Brillouin zone with an accuracy to within 10^{-4} cm^{-1} . Therefore the observed separation $\Delta = 2.5 \pm 0.5 \text{ cm}^{-1}$ cannot be explained by the excitation of two branches of the spin-wave spectrum at a definite point of the Brillouin zone. However, the frequencies of these branches at different points of the Brillouin zone differ from one another as follows:

Point of the Brillouin zone	W	K	U
$E_1 = E_2, \text{ cm}^{-1}$	78.4	77.3	76.2

The frequency difference at the points W and U equal to $\delta = 2.2 \text{ cm}^{-1}$ gives a quantity close to the observed separation. Consequently, one could relate the C_1/C'_1 doublet with the excitation of magnons at the points W and U respectively. This assumption does not contradict the polarization properties of the components of the doublet (see the table)²⁾.

In order to confirm the above assumption, we shall consider the change of the magnetic configuration and the magnon frequency at the points W and U when the intensity of the external field along the x axis is increased. A calculation shows that the magnetic moments should then rotate (Fig. 4c) until at H_{cr} the total moment becomes parallel to the external field. The behavior of the system for $H \neq 0$ is described by the angles α and θ (Fig. 4c).

The dispersion law for the range of fields $0 < H < H_{\text{cr}}$ is of a complex form and the two branches can be represented as follows:

$$\begin{aligned} (E_1/g\mu)^2 &= 1/2[(a_1 + c + d_1 + b)(a_1 + c - d_1 - b) \\ &\quad + (a_2 + c + d_2 + b)(a_2 + c - d_2 - b)], \\ (E_2/g\mu)^2 &= 1/2[(a_1 - c + d_1 - b)(a_1 - c - d_1 + b) \\ &\quad + (a_2 - c + d_2 - b)(a_2 - c - d_2 + b)], \end{aligned} \quad (4)$$

where

$$\begin{aligned} a_1 - d_1 &= 2d_1 + f, \quad a_1 + d_1 = 4d_1 + f, \\ f &= \gamma_{11}(\mathbf{k}) + 1/2 H_E \cos 2\alpha + 2H_{A_1} + H(\sin \theta \sin \alpha + \cos \theta \cos \alpha), \\ a_2 - d_2 &= 2d_2 + f', \quad a_2 + d_2 = 4d_2 + f', \\ f' &= \gamma_{11}(\mathbf{k}) + 1/2 H_E \cos 2\alpha + 2H_{A_1} + H(\sin \theta \sin \alpha - \cos \theta \cos \alpha), \\ b + c &= -1/6 H_E Y_{12}(\mathbf{k}) \cos 2\alpha, \quad c - b = 1/6 H_E Y_{12}(\mathbf{k}), \\ d_1 &= 1/2 H_{A_1} (\sin 2\alpha \cos 2\theta - \sin 2\theta \cos 2\alpha), \\ d_2 &= 1/2 H_{A_1} (\sin 2\alpha \cos 2\theta + \sin 2\theta \cos 2\alpha). \end{aligned}$$

The two spin-wave branches for fields $H_{\text{cr}} < H \ll H_E$ are of the form

$$\begin{aligned} \left(\frac{E_1}{g\mu}\right)^2 &= [\gamma_{11}(\mathbf{k}) + \gamma_{12}(\mathbf{k})][\gamma_{11}(\mathbf{k}) - \gamma_{12}(\mathbf{k}) \cos 2\alpha] \\ &\quad + \gamma_{11}(\mathbf{k}) \left(H_E - \frac{6H_{A_1}H}{H_E} + \frac{5H_{A_2}^2}{H_E} - 2H_{A_1} + \frac{H^2}{H_E} \right) \\ &\quad - \gamma_{12}(\mathbf{k}) \left(H_E + 2H_{A_1} + \frac{4H_{A_2}H}{H_E} - \frac{3H_{A_2}^2}{H_E} - \frac{H^2}{H_E} \right) \\ &\quad + (H^2 - 5H_{A_1}H + 4H_{A_2}^2 - 2H_E H_{A_1}) \left(1 - \frac{H_{A_1}H}{H_E^2} + \frac{H_{A_2}^2}{H_E^2} \right), \\ \left(\frac{E_2}{g\mu}\right)^2 &= [\gamma_{11}(\mathbf{k}) - \gamma_{12}(\mathbf{k})][\gamma_{11}(\mathbf{k}) + \gamma_{12}(\mathbf{k}) \cos 2\alpha] \\ &\quad + \gamma_{11}(\mathbf{k}) \left(H_E - \frac{2H_{A_1}H}{H_E} + \frac{3H_{A_2}^2}{H_E} - \frac{H^2}{H_E} \right) \\ &\quad - \gamma_{12}(\mathbf{k}) \left(H_E + \frac{H_{A_2}^2}{H_E} - \frac{H^2}{H_E} \right) \end{aligned} \quad (5)$$

²⁾The fact that the separation between the C_1/C'_1 doublet and the pure exciton band C_0 exceeds the magnon frequency can be understood if it is assumed that the width of the exciton band is $\sim 4 \text{ cm}^{-1}$.

$$-H_{A_2}(H-H_{A_2}) \left[1 - \frac{(H+2H_{A_2})(H-H_{A_2})}{H_E^2} \right]$$

Using expressions (2), (4), and (5) with known parameters (3), one can obtain the dependence of the magnon frequencies ($E_1 = E_2$) at the special points W and U of the Brillouin zone on the intensity of the external magnetic field (Fig. 7). With allowance for the behavior of the pure exciton absorption line (Fig. 3b) these results make it possible to understand the behavior of the C_1/C'_1 doublet.

In fact, in the case when the external magnetic field is parallel to the z axis both the fundamental band C_0 as well as its magnon satellites C_1 and C'_1 connected with magnon excitation at the points U and W respectively, do not react to an increase of the intensity of the external magnetic field. This attests to the absence of any effect of the external field in this case either on the exciton or on the magnon bands of the crystal. The latter is not surprising since the applied fields are essentially small compared to the exchange field H_E .

The more complicated behavior of the C_1/C'_1 doublet in the case when $H \parallel x$ is also understood. It is seen from Fig. 3a that the experimentally observed behavior of the components C_1 and C'_1 is qualitatively, and for the case of fields $0 \leq H \leq 3$ kOe also quantitatively, quite satisfactorily described by the sum of the frequencies of the components C_0/C'_0 (Fig. 3b) and of the calculated magnon frequencies at the points W and U (Fig. 7).

Thus the attempt made in this paper to describe the behavior of the exciton-magnon bands C_1/C'_1 by a simple superposition of the frequency change of the exciton and of the magnon in a magnetic field has also turned out to be successful in the case of KMnF_3 . The obtained result can be viewed as an additional confirmation of the exciton-magnon nature of the C_1/C'_1 doublet in the C group of bands of KMnF_3 . It also means that in the case of the transition under consideration the exciton-magnon interaction is weak. This is in agreement with the fact that the interval $C_0 - C_1$ exceeds the frequency of the magnon at the boundary of the Brillouin zone by only 4 cm^{-1} .

Starting from the analogy in the structure of the exciton-magnon band of the transition ${}^6A_{1g}({}^6S) \rightarrow {}^4A_{1g}, {}^4E_g({}^4G)$ in the crystals KMnF_3 and RbMnF_3 , one can

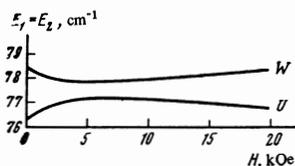


FIG. 7. Dependence of the frequencies of the spin-wave spectrum on the intensity of the external magnetic field $H \parallel x$ for the points W and U of the magnetic Brillouin zone of KMnF_3 .

infer that the conclusions regarding the weakness of the exciton-magnon interaction and the nature of the fine structure of the C_1/C'_1 doublet are valid for crystals of both compounds.

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¹A. I. Belyaeva, V. V. Eremenko, V. N. Pavlov, and A. V. Antonov, *Zh. Eksp. Teor. Fiz.* 53, 1879 (1967) [*Sov. Phys.-JETP* 26, 1069 (1968)].

²B. V. Beznosikov and N. V. Beznosikova, *Kristallografiya* 13, 188 (1968) [*Sov. Phys.-Crystallography* 13, 158 (1968)].

³O. Beckman and K. Knox, *Phys. Rev.* 121, 376 (1961).

⁴A. J. Heeger, O. Beckman, and A. M. Portis, *Phys. Rev.* 123, 1652 (1961).

⁵R. Stevenson, *Phys. Rev.* 152, 531 (1966).

⁶A. V. Antonov, A. I. Belyaeva, and V. V. Eremenko, *Fiz. Tverd. Tela* 8, 3397 (1966) [*Sov. Phys.-Solid State* 8, 2718 (1967)].

⁷J. Ferguson, H. J. Guggenheim, and Y. Tanabe, *J. Phys. Soc. Japan* 21, 692 (1966).

⁸R. Loudon, *Adv. Phys.* 17, 243 (1968).

⁹T. Moriya, *Magnetism*, v. 1, Acad. Press, N. Y.-L., 1963, p. 85.

¹⁰J. J. Pearson, *Phys. Rev.* 121, 695 (1961).

¹¹A. J. Heeger, A. M. Portis, D. T. Teaney, and G. Witt, *Phys. Rev. Letters* 7, 307 (1961).

¹²A. I. Belyaeva and V. S. Kuleshov, *Tr. Fiziko-tekhnicheskogo instituta nizkikh temperatur AN USSR (Proc. of the Physico-technical Institute for Low Temperatures, Academy of Sciences of the Ukrainian SSR)*, No. 3, 1969, Khar'kov.

¹³E. A. Turov, *Fizicheskie svoystva magnitouporyadchennykh kristallov (Physical Properties of Magnetically Ordered Crystals)*, Nauka, 1963, Suppl. A.

¹⁴N. A. Sergienko, V. V. Eremenko, and A. I. Belyaeva, *Tr. Fiziko-tekhnicheskogo instituta nizkikh temperatur AN USSR (Proc. of the Physico-technical Institute for Low Temperatures, Academy of Sciences of the Ukrainian SSR)* No. 3, 1969, Khar'kov.

¹⁵L. P. Bouckaert, R. Smoluchowski, and W. Wigner, *Phys. Rev.* 50, 58 (1936).

¹⁶V. V. Eremenko and A. I. Belyaeva, *Usp. Fiz. Nauk* 98, 27 (1969) [*Sov. Phys.-Uspekhi* 12, 320 (1969)].

¹⁷M. Lax and J. J. Hopfield, *Phys. Rev.* 124, 115 (1964).

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