Multimagnon absorption of antiferromagnetic $RbMnF_3$ in the optical spectrum

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Optical electric-dipole absorption in antiferromagnetic $RbMnF_3$ is investigated experimentally in the region of the $^6A_{1g}(^6S) \rightarrow {}^4E_g(^4G)$ transition in strong magnetic fields up to 300 kOe. The parameters of the previously investigated single-particle pure-exciton transition $^6A_{1g}(^6S) \rightarrow {}^4E_g(^4G)$ are used to calculate (within the framework of the exciton model for spin and optical excitations) the transition dipole moments and the absorption-peak frequencies for its two- and three-magnon satellites. Exciton dispersion for translationally equivalent ions and various exciton-magnon interactions is estimated in the nearest-neighbor approximation. It is shown that the major part (200 cm^{-1}) of the absorption band group under consideration is formed by one-, two-, and three-magnon satellites of the transition.

INTRODUCTION

It is well known that antiferromagnetic compounds of the iron group are characterized by intense electric-dipole absorption at frequencies of the spin- and parity-forbidden single-ion optical transitions; this typical behavior can be explained in terms of paired optical transitions. Among the competing mechanisms, the most universal is the variant of pair excitation of exchange-coupled magnetic ions, proposed by Tanabe, Moriya, and Sugano (TMS)[1]. The resultant effective electric-dipole moment of the transition provides, in particular, a natural explanation of the exciton-magnon absorption band, which it relates to the simultaneous formation of optical and spin excitations, i.e., of an exciton and magnon, on different sublattices.

One-magnon satellites of exciton lines, however, do not account by far for the entire fine structure of the observed spectra, which may receive a contribution not only from phonons but possibly also from transitions in which a larger number of magnons take part. The role of multimagnon absorption (in optical spectra) was never discussed seriously before, in spite of some identification attempts[2-5]. One of us[6] has indicated that multimagnon absorption electric-dipole transitions are due to interactions between excited ions. The corresponding operators of the effective moments of the transitions are obtained with the aid of the operator of TMS two-particle transitions with account taken of interactions that do not conserve the number of spin excitations. From this it follows immediately that multimagnon transitions, which actually occur in higher order of the approximation in the exchange interaction, should be much weaker than the single-magnon transitions. Their identification, however, should not be regarded as hopeless, in view of the appreciative intensity of exciton-single-magnon transitions and of the following circumstances:

First, the higher order in the exchange interaction means a stronger dependence of their intensity on the magnetic field that influences the relative spin orientation of the exchange-coupled magnetic ions. This makes the multimagnon-absorption bands quite distinct.

Second, most importantly, there is hope for determining the frequencies of the absorption maxima of the simplest types of multimagnon satellites. The point is that a direct comparison of the frequency intervals observed in the optical spectra with the spin-wave dispersion parameters (for example, with the magnon energy at the edge of the Brillouin zone) is meaningless. It is now clearly understood that the pair transitions are determined not only by the dispersion laws of the corresponding independent quasiparticles. It is necessary to take into account their interaction and the wave-vector-dependent probability of production of the considered pairs of excitations. In the usual phenomenological approach, allowance for the indicated interactions would call for the introduction of a number of underrated parameters for the description of various types of transitions. However, since spin excitation of the ion does not change its orbital part, the parameters of the interactions of interest to us should in final analysis be expressed in terms of exciton parameters determined from an analysis of the Davydov splitting of the single-particle transition. Indeed, within the framework of the unified exciton model of optical and spin excitations of an antiferromagnetic dielectric[7], the description of all types of magnon satellites of an excitonic transition is obtained without introducing any new parameters at all. All the necessary quantities, particularly the various exciton-magnon interactions, are expressed in terms of the single-particle transition parameters. This affords a possibility of an unambiguous experimental estimate (within the framework of the model) of the excitonic dispersion, and makes it possible in principle to predict the positions of the multimagnon satellites in the optical spectrum.

From this point of view, the investigation of multimagnon satellites becomes of interest regardless of their absolute contribution to the optical absorption. The foregoing considerations are quite general in character, but for their particular application it is necessary to know the parameters of the single-particle optical excitation of the considered state.

The only experimental data that are sufficiently complete for this purpose are those (see[8,9]) for the purely excitonic $^6A_{1g}(^6S) \rightarrow {}^4E_g(^4G)$ transition of antiferromagnetic $RbMnF_3$. The magnetic Davydov splitting typical of antiferromagnets has been hardly investigated, with the exception of a few substances, among which $RbMnF_3$ is the most suitable. This is the only two-sublattice collinear antiferromagnet that is strictly cubic in the ordered state ($T_N = 82.6°K$)[10]. The weak magnetic field ($H_{cr} \sim 3$ kOe) smoothly rotates the sublattice spins to a plane perpendicular to the field, while a strong field ($\sim 10^5$ kOe) noticeably disturbs the collinearity, causing the sublattices to be symmetrically canted in the field direction[11,12]. The spin-wave spec-

FIG. 1. Density patterns of the absorption spectrum of $RbMnF_3$ in the region of the $^6A_{1g}(^6S) \rightarrow {}^4E_g, {}^4A_{1g}(^4G)$ transition at T = 20.4°K: a—sample thickness 1.08 mm, b—normally exposed spectrum of sample 0.03 mm thick, which gives a qualitative idea of the intensity ratio of the fundamental absorption bands. Certain distinctly appearing bands are marked in accordance with the system adopted in Fig. 3 below.

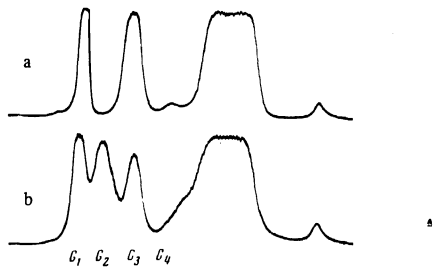

FIG. 2. Density patterns of the considered region of the $RbMnF_3$ spectrum at T = 20.4°K. Sample thickness 0.8 mm. The spectra were obtained in unpolarized light with a certain prior exposition in order to reveal the details more clearly. a) H = 0, b) H = 300 kOe.

trum of $RbMnF_3$ is simple and is well known[13]. The electric-dipole light-absorption spectrum of $RbMnF_3$ in the region of the $^6A_{1g}(^6S) \rightarrow {}^4E_g, {}^4A_{1g}(^4G)$ transition has a developed fine structure (Fig. 1). Its origin, however, is not quite clear, and there is still no convincing interpretation of most of the observed absorption bands, in spite of the efforts made in this direction[14-18]. The only exception is the most intense and asymmetrical band C_1 (Fig. 1), which is interpreted as a single-magnon satellite of the pure excitonic $^6A_{1g}(^6S) \rightarrow {}^4E_g(^4G)$ transition[14,18]. We are interested in the spectral region directly adjacent to the band C_1 on the high-frequency side, where multimagnon-absorption bands of the same excitonic transition may be located.

Proceeding now to a description of the results, we note immediately that we do not concern ourselves with the problem of the shapes of the multimagnon absorption bands, which in the case of a three-dimensional antiferromagnet calls for computer calculation even in the simplest case of a single-magnon satellite.

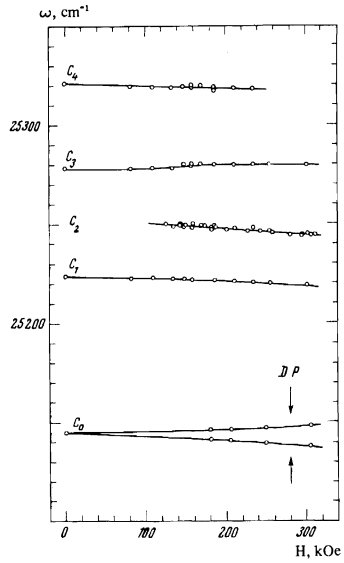

FIG. 3. Frequencies of $RbMnF_3$ absorption bands in the region of the $^6A_{1g}(^6S) \rightarrow {}^4E_g(^4G)$ transition in an external magnetic field H ∥ [110] at T = 20.4°K.

METHODOLOGICAL REMARKS

All the spectra were obtained with a DFS-13 diffraction spectrograph with dispersion 2 Å/mm and with resolution 144,000. The frequencies were determined by comparison with the spectrum of an iron arc, using an IZA-2 comparator, and from the density patterns (in the case of broad lines) with accuracy not worse than 1 cm^{-1}. The spectra in the magnetic field were obtained in the geometry of the longitudinal Zeeman effect, using a miniature pulsed solenoid cooled with liquid hydrogen. The samples were oriented by x-ray diffraction with accuracy 1—2°. The intensity estimates are only approximate.

EXPERIMENTAL RESULTS

Unlike the previously investigated[8,9] excitonic magnetic-dipole absorption C_0, the electric-dipole spectrum of the $^6A_{1g}(^6S) \rightarrow {}^4E_g, {}^4A_{1g}(^4G)$ transition, with the exception of the band C_1, turned out to be isotropic and practically unpolarized.

The most interesting feature of the influence of the strong magnetic field is the abrupt change in the intensities of a large number of absorption bands (Fig. 2). Particularly significant changes occur in the region of the band C_1—C_4, which form a distinctly isolated group in a strong field. A band C_2 is produced and is rapidly amplified, and the intensity of C_3 and C_4 decreases at no less a rapid rate. The exciton-magnon band C_1 seems to weaken a bit, but this effect is much less significant and can hardly be observed on the density patterns of Fig. 2, which pertain to a sample of considerable thickness.

The doublet Bethe splitting of the exciton band C_0, which is observed under circumstances (see[8,9]), gives rise to an identical splitting of C_1, which is of interest only for the polarization of the single-magnon bands corresponding to different components of C_0. The polarization direction turns out to be connected with the direction of the magnetic ordering, and the observed degree of polarization depends on the population of the possible equivalent directions of the ordering and their positions relative to the electric vector of the light. This multidomain structure, which is typical of $RbMnF_3$, is eliminated only by external uniaxial compression and is preserved in a magnetic field[8], leading to a noticeable polarization that masks, (under unfavorable conditions), the very fact of the Bethe splitting of the exciton-magnon band.

Particular interest attaches therefore to the behavior of the electric-dipole spectrum in a magnetic field applied along a twofold order and causing no noticeable Bethe splitting of C_0. As seen from Fig. 3, the appreciable Davydov splitting of the exciton band C_0, which is observed in this case[8], is not accompanied by a splitting of its single-magnon satellite C_1, despite certain predictions[18]. This fact, in contrast to the already mentioned Bethe splitting of C_1, is not trivial and calls for an explanation.

We note also that in strong magnetic fields most absorption bands have negative frequency shifts that do

TABLE I

Band	Experimental frequencies, cm^{-1}			Calculation results		
	$H=0$	H = 300 kOe	Shift	Identification	$\omega(H=0)$	Shift
C_0	25144.5	25141.5 *	−3		—	—
C_1	25223.5	25218.5	−5	ω^{I}_{max}	25223.5	−2
C_2	25253 **	25245	−8	$\omega^{II}_{2\,max}$	25252.5	−3.3
$C_{3'}$	25278	25280	+2	$\omega^{III}_{1\,max}$	25278.5	+1
C_4	25321	25317	−4	$\omega^{III}_{2\,max}$	25321.5	+1.5

*Position of center of gravity.
**Extrapolation.

not coincide with the shift of the center of gravity of the Davydov doublet C_0 (Table I).

DISCUSSION

1. For a theoretical analysis of the absorption of light by the magnon satellites of the spin-forbidden exciton transition of antiferromagnetic crystal it is necessary to know the dipole moments of the corresponding pair transition. In the general noncollinear case, the operator P_{eff} of the transition dipole moment, which describes the excitation of a pair of ions into optical and spin excited states, was obtained in[19]. At $H > H_{cr}$ and when the sublattice spins are symmetrical about the magnetic field, we have

$$P_{eff}=\sum_{n\alpha,m\beta}\Pi_{n\alpha,m\beta}B_{n\alpha}^{+}(f)\,b_{m\beta}^{+},\quad \Pi_{n\alpha,m\beta}=\sin^2\vartheta(1-\delta_{\alpha\beta})P_{n\alpha,m\beta}, \tag{1}$$

where $P_{n\alpha,m\beta}$ is the dipole moment of a transition in a pair of magnetic ions $n\alpha$ and $m\beta$ from opposite sublattices α and β ($\alpha, \beta = 1, 2$) of a collinear antiferromagnet; $B^{+}_{n\alpha}(f)$ and $b^{+}_{m\beta}$ are the production operators of the f-th optical excitation and the spin excitation of the magnetic ion, and ϑ is the angle between the directions of the spins and the magnetic field.

As shown in[6], in addition to the dipole moment of the transition (1), additional dipole transitions are possible and characterize excitation of a pair of ions into optical states and doubly-spin-excited states:

$$D^{(1)}_{eff}=\sum_{n\alpha,m\beta}D^{(1)}_{n\alpha,m\beta}B_{n\alpha}^{+}(f)\,b_{n\alpha}^{+}b_{m\beta}^{+},\quad D^{(2)}_{eff}=\sum_{n\alpha,m\beta}D^{(2)}_{n\alpha,m\beta}B_{n\alpha}^{+}(f)\,b_{m\beta}^{+}b_{m\beta}^{+} \tag{2}$$

and optical and triply-spin-excited states:

$$R_{eff}=\sum_{n\alpha,m\beta}R_{n\alpha,m\beta}B_{n\alpha}^{+}(f)\,b_{n\alpha}^{+}b_{m\beta}^{+}b_{m\beta}^{+}. \tag{3}$$

The operators $D^{(1)}_{eff}$, $D^{(2)}_{eff}$, and R_{eff} are obtained with the aid of the operator P_{eff} with account taken, in first-order perturbation theory, of the terms of the interaction of the optically and spin-excited ions, which do not conserve the number of spin excitation. However, the expressions given in[6] did not take into account the terms for the interaction of spin-excited ions with one another. To take full account of the interaction terms there is no need to expand the Hamiltonian $\mathcal{H}$ of the antiferromagnetic crystal (an expression for $\mathcal{H}$ in terms of electronic operators is given in[7]), as was done in[6]. It suffices to use the definition of the dipole transition. Thus, for example,

$$D^{(1)}_{n\alpha,m\beta}=\sum_{p\gamma,r\delta}{}'\frac{\langle\varphi_{n\alpha}^{fs};\varphi_{m\beta}^{s}|\mathcal{H}|\varphi_{p\gamma}^{f};\varphi_{r\delta}^{s}\rangle\Pi_{p\gamma,r\delta}}{\Delta E_{\alpha}^{fs}+\Delta E_{\beta}^{s}-\Delta E_{\gamma}^{f}-\Delta E_{\delta}^{s}}, \tag{4}$$

where $|\varphi^{s}_{n\alpha}\rangle$, $|\varphi^{f}_{n\alpha}\rangle$, and $|\varphi^{fs}_{n\alpha}\rangle$ are the wave functions of the spin, f-th optical, and spin-optical excitations of the $n\alpha$-th magnetic ion, and are given in[7]; ΔE^{s}_{α}, ΔE^{f}_{α}, and ΔE^{fs}_{α} are the corresponding excitation energies, with

$$\Delta E_{\alpha}^{s}=\Delta\varepsilon_{\alpha}^{s}+D_{\alpha}^{s},\ \Delta E_{\alpha}^{f}=\Delta\varepsilon_{\alpha}^{f}+D_{\alpha}^{f},\ \Delta E_{\alpha}^{fs}=\Delta E_{\alpha}^{s}+\Delta E_{\alpha}^{f}+V_{n\alpha}^{fs} \tag{5}$$

(all the quantities in (5) are defined in[7]). Using the explicit expressions given in[7] for the wave functions, the Hamiltonian $\mathcal{H}$, and the quantities (5), we get in place of (4)

$$\begin{aligned}D^{(1)}_{n\alpha,m\beta}=\sin^3\vartheta\cos\vartheta P_{n\alpha,m\beta}\Big\{&\frac{2}{\sqrt{2(s-1)}}\Big(sJ_{n\alpha,m\beta}-\frac{1}{s}J^{f}_{n\alpha,m\beta}\Big)\\&+\frac{2sz}{\sqrt{2(s-1)}}\Big(I+\frac{1}{s}I^{f}\Big)-\frac{2}{\sqrt{2s}}\Big(\frac{s-1}{s}\Big)^{1/2}M^{f}_{n\alpha,m\beta}\Big\}\\&\times\Big\{szI-\frac{sz}{s-1}\Big(I+\frac{1}{s}I^{f}\Big)(2\cos^2\vartheta-1)\Big\}^{-1}.\end{aligned} \tag{6}$$

Analogously, we get

$$D^{(2)}_{n\alpha,m\beta}=\sin^3\vartheta\cos\vartheta P_{n\alpha,m\beta}\frac{(2s-1)J^{f}_{n\alpha,m\beta}+M^{f}_{n\alpha,m\beta}}{s(2s-1)^{1/2}szI}, \tag{7}$$

$$\begin{aligned}R_{n\alpha,m\beta}=\sin^4\vartheta P_{n\alpha,m\beta}&\Big\{\Big(\frac{2s-1}{2(s-1)}\Big)^{1/2}\Big(sJ_{n\alpha,m\beta}-\frac{1}{s}J^{f}_{n\alpha,m\beta}\Big)\\&+\Big(\frac{s-1}{2(2s-1)}\Big)^{1/2}\frac{1}{s}M^{f}_{n\alpha,m\beta}\Big\}\Big\{2szI-\frac{sz}{s-1}\Big(I+\frac{1}{s}I^{f}\Big)(2\cos^2\vartheta-1)\Big\}^{-1}.\end{aligned} \tag{8}$$

All the dipole moments are expressed in terms of the parameters $J_{n\alpha,m\beta}$, $J^{f}_{n\alpha,m\beta}$, and $M^{f}_{n\alpha,m\beta}$ defined in[7] and characterizing respectively the interaction of the unexcited ions, the interaction of the optically excited ion with an unexcited one, and the interaction that leads to the transfer of optical excitations to an unexcited ion. The values of these parameters for the nearest neighboring ions ($I = -J_{n1,n2}$, $I^{f} = J^{f}_{n1,n2}$, and $M^{f} = M^{f}_{n1,n2}$) are determined from the experimental data on antiferromagnetic resonance and on the Davydov splitting of the f-th optical excitation in a strong magnetic field. For $RbMnF_3$, according to the data of[13], we have $sIz = 72\ cm^{-1}$ and for excitation to the $^4E_g(^4G)$ state[8,9] we have $zI^{f} = 126\ cm^{-1}$ and $zM^{f} = 200\ cm^{-1}$, where $s = 5/2$ is the spin of the Mn^{2+} ion and $z = 6$ is the number of nearest neighbors from the opposite sublattice. The dipole transitions (2) and (3) together with (6)—(8) characterize the probability that the light will excite two-magnon and three-magnon satellites of the exciton transition.

2. We present now the necessary Hamiltonians. In the case of light absorption by a one-magnon satellite we have

$$\mathcal{H}^{f,s}=\mathcal{H}_0^{f}+\mathcal{H}_0^{s}+\mathcal{H}^{f-s}_{f-s},\quad \mathcal{H}^{f,s}_{cr-\gamma}(t)=EP_{eff}e^{-i\omega t}+\text{H.c.}, \tag{9}$$

where $\mathcal{H}^{f}_{0}$ and $\mathcal{H}^{s}_{0}$ are the Hamiltonians of the optical and spin excitations of the ferromagnet; $\mathcal{H}^{f-s}_{f-s}$ is the Hamiltonian of the interaction of optically- and spin-excited ions, explicit expressions for which in the band representation are given in[7]; $\mathcal{H}^{f,s}_{cr-\gamma}(t)$ is the operator of the interaction of the antiferromagnet with light (E is the electric vector of the light wave) in the region of the frequencies of the optical excitation and one spin excitation.

For two-magnon absorption we have

$$\begin{aligned}&\mathcal{H}^{f,2s}=\mathcal{H}_0^{f}+\mathcal{H}_0^{s}+\mathcal{H}^{fs-s}_{fs-s}+\mathcal{H}^{f-2s}_{f-2s}+\mathcal{H}^{fs-s}_{f-2s},\\&\mathcal{H}^{f,2s}_{cr-\gamma}(t)=E(D^{(1)}_{eff}+D^{(2)}_{eff})e^{-i\omega t}+\text{H.c.},\end{aligned} \tag{10}$$

where $\mathcal{H}^{fs-s}_{fs-s}$, $\mathcal{H}^{f-2s}_{f-2s}$, and $\mathcal{H}^{fs-s}_{f-2s}$ are the Hamiltonians of the exciton-two-magnon interactional $\mathcal{H}^{fs-s}_{fs-s}$ characterizes the interaction of pairs of magnetic ions, one of

which is optically- and spin-excited, and the other only spin-excited; $\mathcal{H}^{f-2s}_{f-2s}$ takes into account the interaction between the two considered types of excitations. In the Hartree-Fock approximation, these Hamiltonians take the form

$$\mathcal{H}^{fs-s}_{fs-s}=\sum_{n\alpha,m\beta}\{\Phi^{fs-s}_{1n\alpha,m\beta}B_{n\alpha}^{+}(f)\,b_{n\alpha}^{+}b_{m\beta}+B_{n\alpha}(f)\,b_{n\alpha}b_{m\beta}$$
$$+\Phi^{fs-s}_{2n\alpha,m\beta}B_{n\alpha}^{+}(f)\,b_{n\alpha}^{+}b_{m\beta}^{+}B_{m\beta}(f)\,b_{m\beta}b_{n\alpha}\},$$
$$\mathcal{H}^{f-2s}_{f-2s}=-\frac{1}{2}\sum_{n\alpha,m\beta}\{\Phi^{f-2s}_{1n\alpha,m\beta}B_{n\alpha}^{+}(f)\,b_{m\beta}^{+}b_{m\beta}^{+}B_{n\alpha}(f)\,b_{m\beta}b_{m\beta}\qquad(11)$$
$$+\Phi^{f-2s}_{2n\alpha,m\beta}B_{n\alpha}^{+}(f)\,b_{m\beta}^{+}b_{m\beta}^{+}B_{m\beta}(f)\,b_{n\alpha}b_{n\alpha}\},$$
$$\mathcal{H}^{fs-s}_{f-2s}=-\frac{1}{\sqrt{2}}\sum_{n\alpha,m\beta}\{\chi_{n\alpha,m\beta}B_{n\alpha}^{+}(f)\,b_{n\alpha}^{+}b_{m\beta}^{+}B_{m\beta}(f)\,b_{n\alpha}b_{n\alpha}+\text{H.c.}\}.$$

The quantities $\Phi_{jn\alpha,m\beta}$ and $\chi_{n\alpha,m\beta}$ in (11) are obtained by the method developed in[7] and are given by

$$\Phi^{fs-s}_{1n\alpha,m\beta}=-(2\cos^2\vartheta-1)\frac{1}{s-1}\left\{sJ_{n\alpha,m\beta}+\frac{s-2}{s}J^{f}_{n\alpha,m\beta}+sIz+zI^{f}\right\},$$
$$\Phi^{fs-s}_{2n\alpha,m\beta}=-\cos^2\vartheta\,\frac{s-1}{s}M^{f}_{n\alpha,m\beta},\qquad\Phi^{f-2s}_{1n\alpha,m\beta}=-(2\cos^2\vartheta-1)\frac{2}{s}J^{f}_{n\alpha,m\beta},$$
$$\Phi^{f-2s}_{2n\alpha,m\beta}=-\cos^2\vartheta\,\frac{1}{2(2s-1)}M^{f}_{n\alpha,m\beta},\qquad(12)$$
$$\chi_{n\alpha,m\beta}=-(2\cos^2\vartheta-1)\frac{2}{s}\left(\frac{s-1}{2s-1}\right)^{1/2}M^{f}_{n\alpha,m\beta}.$$

Finally, to investigate three-magnon absorption of light we need the Hamiltonians

$$\mathcal{H}^{f,3s}=\mathcal{H}_0{}^{f}+\mathcal{H}_0{}^{s}+\mathcal{H}^{fs-2s}_{fs-2s},$$
$$\mathcal{H}^{f,3s}_{cr-\gamma}(t)=\mathbf{E}R_{eff}e^{-i\omega t}+\text{H. c.}\qquad(13)$$

Here $\mathcal{H}^{fs-2s}_{fs-2s}$ describes the interaction of the spin-optically- and doubly-spin-excited ions and is given in the Hartree-Fock approximation by

$$\mathcal{H}^{fs-2s}_{fs-2s}=\frac{1}{2}\sum_{n\alpha,m\beta}\{W^{fs-2s}_{1n\alpha,m\beta}B_{n\alpha}^{+}(f)\,b_{n\alpha}^{+}b_{m\beta}^{+}b_{m\beta}^{+}B_{n\alpha}(f)\,b_{n\alpha}b_{m\beta}b_{m\beta}\qquad(14)$$
$$+W^{fs-2s}_{2n\alpha,m\beta}B_{n\alpha}^{+}(f)\,b_{n\alpha}^{+}b_{m\beta}^{+}b_{m\beta}^{+}B_{m\beta}(f)\,b_{m\beta}b_{n\alpha}b_{n\alpha}\},$$

where

$$W^{fs-2s}_{1n\alpha,m\beta}=-(2\cos^2\vartheta-1)\frac{2}{s-1}\left\{sJ_{n\alpha,m\beta}+\frac{s-2}{s}J^{f}_{n\alpha,m\beta}+\frac{1}{2}(sIz+zI^{f})\right\},\qquad(15)$$
$$W^{fs-2s}_{2n\alpha,m\beta}=-(2\cos^2\vartheta-1)\frac{4}{s}\,\frac{s-1}{2s-1}M^{f}_{n\alpha,m\beta}.$$

We emphasize that all the coefficients of the presented Hamiltonians are expressed in terms of the same system of parameters $J_{n\alpha,m\beta}$, $J^{f}_{n\alpha,m\beta}$, and $M^{f}_{n\alpha,m\beta}$.

3. The absorption coefficient $K_\nu(\omega)$ of light of frequency ω and polarization ν is expressed, according to[20], by the formula

$$K_\nu(\omega)=-\frac{4\pi\omega}{cV\eta}\operatorname{Im}\lim_{\delta\to0}G(T^{\nu}_{eff}|T^{\nu}_{eff})_{\omega+i\delta},\qquad(16)$$

where c is the speed of light, η is the reflection coefficient, V is the volume of the crystal, T_{eff} is the operator of the corresponding dipole moment of the transition, and $G(T^{\nu}_{eff}|T^{\nu}_{eff})_\omega$ is the Fourier transform of the retarded Green's function

$$G(T^{\nu}_{eff}(t)|T^{\nu}_{eff})\equiv-i\Theta(t)\ll T^{\nu}_{eff}(t)|T^{\nu}_{eff}(0)\gg.$$

For convenience in the calculation of $K_\nu(\omega)$, we use the representation of quasiparticles, excitons, and magnons, which are introduced by the transformations

$$B_{n\alpha}(f)=\frac{1}{\sqrt{N}}\sum_{k\mu}e^{ikn\alpha}u_{\alpha\mu}(\mathbf{k},f)B_{\mu}(\mathbf{k},f),$$

$$b_{n\alpha}=\frac{1}{\sqrt{N}}\sum_{k\mu}\{e^{ikn\alpha}u_{\alpha\mu}(\mathbf{k})\,b_{\mu}(\mathbf{k})+e^{-ikn\alpha}v_{\alpha\mu}(\mathbf{k})\,b_{\mu}^{+}(\mathbf{k})\},$$

where

$$u_{\alpha\mu}(\mathbf{k},f)=\frac{1}{\sqrt{2}}\begin{pmatrix}1&1\\1&-1\end{pmatrix},$$

and the coefficients of the u-v transformation that diagonalizes the spin Hamiltonian $\mathcal{H}^{S}_{0}$ can be easily found (see[21]) for the concrete case considered by us, that of a cubic two-sublattice antiferromagnet in the nearest-neighbor approximation. The equations for the determination of the Green's functions are solved in the same approximation[1)].

4. Confining ourselves for concreteness to the case of polarization along the fourfold axis x of the crystal, we obtain the following expression for the absorption of light by a one-magnon satellite:

$$K_x(\omega)=-\frac{4\pi\omega}{cV\eta}\sin^4\vartheta|P_x|^2\operatorname{Im}G(\omega),\qquad(17)$$

where

$$G(\omega)=2N\frac{\Lambda(\omega)}{1-(V_1-V_2)\Lambda(\omega)},\quad\Lambda(\omega)=\frac{2}{N}\sum_{k\mu}\frac{u_\mu^2(\mathbf{k})\sin^2ak_x}{\omega-\varepsilon_\mu(\mathbf{k})-E_\mu(\mathbf{k},f)}$$

Here N is the number of magnetic cells of the crystal; V_1 and V_2 are the quantities $V^{fs}_{1n\alpha,m\beta}$ and $V^{fs}_{2n\alpha,m\beta}$ defined in[7] for the nearest neighbors, and characterize respectively the contribution made to the exciton-magnon interaction by the "static" interaction of the optically- and spin-excited nearest magnetic ions, and the "dynamic" interaction due to the fact that the optical and spin excitations exchange places. Then

$$V_1-V_2=(2\cos^2\vartheta-1)(M^{f}-I^{f})\frac{1}{s}\approx(2\cos^2\vartheta-1)\cdot22\ \text{cm}^{-1}.\qquad(18)$$

Further, the quantities $u_\mu(k)$ denote the coefficients of the u-v transformation $u_1(k)=u_{11}(k)=u_{21}(k)$, $u_2(k)=u_{12}(k)=-u_{22}(k)$, $\epsilon_\mu(k)$ and $E_\mu(k,f)$ are respectively the energies of the magnon (band index $\mu=1,2$; k is the wave vector) and of the exciton of the f-th optical excitation, the explicit expressions for which[7,5,13,22] in the noncollinear case ($H>H_{cr}$) take in the nearest-neighbor approximation the form

$$\varepsilon_\mu(k)=sIz\{1-\gamma^2(k)+2\cos^2\vartheta\gamma(k)[\gamma(k)-(-1)^{\mu}]\}^{1/2},$$
$$E_\mu(k,f)=\Delta\varepsilon^{f}+A^{f}\cos^2\vartheta+\tilde{z}\tilde{M}^{f}\bar{\gamma}(k)-(-1)^{\mu}zM^{f}\gamma(k)\cos^2\vartheta,\qquad(19)$$
$$\gamma(k)=\tfrac{1}{3}(\cos ak_x+\cos ak_y+\cos ak_z),$$
$$\bar{\gamma}(k)=\tfrac{1}{3}(\cos ak_x\cos ak_y+\cos ak_x\cos ak_z+\cos ak_y\cos ak_z),\qquad(20)$$
$$A^{f}=2sIz+2zI^{f},\ \cos\vartheta=H/2H_E;$$

$\Delta\epsilon^{f}$ is a constant for the given state; M^{f} and $\tilde{M}^{f}$ are the values of the parameter $M^{f}_{n\alpha,m\beta}$ for the first and second nearest magnetic neighbors, the number of which is z and $\tilde{z}$, respectively; H_E is the exchange field, 0.89 $\times 10^6$ kOe for $RbMnF_3$.

Let us determine the position of the maximum of the absorption by a one-magnon satellite. It can correspond to the points k_0 of the Brillouin zone (see Fig. 4 and

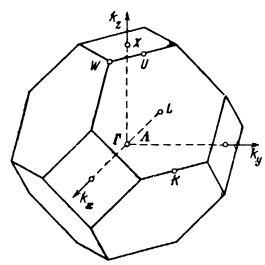

FIG. 4. Brillouin zone of antiferromagnetic $RbMnF_3$.

TABLE II. Values of $\gamma(\mathbf{k})$ and $\tilde{\gamma}(\mathbf{k})$ for the most characteristic points of the Brillouin zone (Fig. 4)

	$\gamma(\mathbf{k})$	$\tilde{\gamma}(\mathbf{k})$
Γ	1	1
X	$1/3$	$-1/3$
L	0	0
W	0	$-1/3$
K	$(1-2^{1/3})/3 \approx -0.14$	$(1-2\cdot 2^{1/3})/6 \approx -0.30$
U	$-(1-2^{1/3})/3 \approx 0.14$	$(1-2\cdot 2^{1/3})/6 \approx -0.30$

Table II), which are singular points of the density of states of the exciton-magnon band:

$$\rho_\mu(\omega)=\frac{1}{N}\sum_{\mathbf{k}}\delta(\omega-\varepsilon_\mu(\mathbf{k})-E_\mu(\mathbf{k},f)), \tag{21}$$

in which the factor $u_\mu^2(\mathbf{k})\sin^2 ak_x$ in (17) does not vanish. The frequency of the absorption maximum is determined in this case by the expression

$$\omega^{\mathrm{I}}_{max}(f,H)=E_\mu(\mathbf{k}_0,f)+\varepsilon_\mu(\mathbf{k}_0)+V_1-V_{2a}. \tag{22}$$

Using the explicit expressions for the energies of the exciton (20) and of the magnon (19), we can show that the singular points of the exciton-magnon density of states (21), at which the group velocity vanishes

$$\mathbf{v}_\mu^{\mathrm{rp}}(\mathbf{k})=\frac{\partial}{\partial \mathbf{k}}\{\varepsilon_\mu(\mathbf{k})+E_\mu(\mathbf{k},f)\}$$

are the points $\Gamma = (\pi/a)(0,0,0)$, $X = (\pi/a)(1,0,0)$ and the points of the Γ-L lines, determined by the expressions

$$\cos ak_{0x}=\cos ak_{0y}=\cos ak_{0z}=\frac{(-1)^\mu\cos^2\vartheta[zM'+(szI)^2/\varepsilon_\mu(\mathbf{k}_0)]}{(2\cos^2\vartheta-1)(szI)^2/\varepsilon_\mu(\mathbf{k}_0)+2\tilde{z}\tilde{M}'}. \tag{23}$$

It is the latter which determine the position of the absorption maximum, since $\sin^2 ak_x = 0$ at the points X and Γ.

In the absence of an external magnetic field ($\cos\vartheta = 0$) the points $\mathbf{k}_0$ obviously coincide with the points $L = (\pi/a)(1/2, 1/2, 1/2)$. Recalling that the pure exciton absorption corresponds to the point Γ of the Brillouin zone, we obtain the frequency difference between the maximum of the exciton-magnon satellite and line of pure exciton absorption (C_1 and C_0 in Table I) at $H = 0$:

$$\omega^{\mathrm{I}}_{max}-E[\Gamma,{}^4E_g({}^4G)]=szI-\tilde{z}\tilde{M}'-\frac{1}{s}(M'-I'). \tag{24}$$

The numerical values of all the parameters in this expression, except $\tilde{M}^f$ (which characterizes the exciton dispersion for the translationally-equivalent ions), are known. We therefore find from Table I and from (24) that $\tilde{z}\tilde{M}^f = -29\ \mathrm{cm}^{-1}$. We emphasize that the determination of this dispersive term of the exciton energy (20) is an important step in the determination of the frequencies of the maxima of two- and three-magnon absorption, which are determined, generally speaking, by other points of the Brillouin zone.

It follows from (23) that in fields up to 300 kOe the points $\mathbf{k}_0$ shift little away from the points L, and therefore the magnon and exciton energies in (22) are practically independent of the exciton-band index μ. This explains indeed the experimentally observed (Fig. 3) absence of splitting of the exciton-magnon satellite C_1, in spite of the appreciable Davydov splitting of the exciton line C_0.

5. For two- and three-magnon absorption, the calculations are quite similar, the expressions obtained for the absorption coefficients are similar to (17), it being obvious from (1) and (6)—(8) that $\sin^4\vartheta$ must be replaced by $\sin^6\vartheta\cos^2\vartheta$ and $\sin^8\vartheta$ respectively for the two- and three-magnon absorptions. To find the frequencies of the absorption maxima we use the same ideas concerning the singularities and densities of state of the excitons and magnons, making additional allowance for the possible decay of the spin-optical excitation of the magnetic ion, which occurs in the transitions described by $D^{(1)}_{\mathrm{eff}}$ and R_{eff}. With the ion spin projection changing by two units and the spin itself changing by one unit, the spin-optical excitation cannot migrate spontaneously over the crystal, owing to the strong spin forbiddenness, and this leads to the need for taking into account its decay into optical and spin excitations of two ions, excitations capable of migrating.

In the case of two-magnon absorption, we obtain two principal absorption maxima, located at the frequencies

$$\begin{aligned}\omega^{\mathrm{II}}_{1,2\,max}(H)=&{}^1/_2(E_1(\mathbf{k}_0,f)+E_2(\Gamma,f)+3\varepsilon_1(\mathbf{k}_0)+\varepsilon_2(X)+\Phi_1+\Phi_2+\tilde{\Phi}_1+\tilde{\Phi}_2\\ &\pm([E_1(\mathbf{k}_0,f)-E_2(\Gamma,f)+\varepsilon_2(X)-\varepsilon_1(\mathbf{k}_0)+\Phi_1+\Phi_2-\tilde{\Phi}_1-\tilde{\Phi}_2]^2+4\chi^2)^{1/2}),\end{aligned} \tag{25}$$

where the quantities

$$\begin{aligned}&\Phi_1\approx(2\cos^2\vartheta-1)\cdot 48\ \mathrm{cm}^{-1},\\ &\Phi_2\approx-\cos^2\vartheta\cdot 20\ \mathrm{cm}^{-1}\quad \tilde{\Phi}_1\approx(2\cos^2\vartheta-1)\cdot 16\,\mathrm{cm}^{-1},\\ &\tilde{\Phi}_2\approx-4\cos^2\vartheta\cdot 4\ \mathrm{cm}^{-1},\quad \chi\approx-(2\cos^2\vartheta-1)\cdot 16\,\mathrm{cm}^{-1}\end{aligned}$$

are respectively the values of $\Phi^{\mathrm{fs\text{-}s}}_{1n\alpha,m\beta}$, $\Phi^{\mathrm{fs\text{-}s}}_{2n\alpha,m\beta}$, $\Phi^{\mathrm{f\text{-}2s}}_{1n\alpha,m\beta}$, and $\Phi^{\mathrm{f\text{-}2s}}_{2n\alpha,m\beta}$, $\chi_{n\alpha,m\beta}$ for the nearest magnetic ions; the points $\mathbf{k}_0$ practically coincide with L in the experimental interval of the magnetic fields, and $\epsilon_2(X) \approx (1 - \frac{1}{4}\cos^2\vartheta)\times 68\ \mathrm{cm}^{-1}$. Using expressions (19) and (20) and taking the foregoing estimate of $\tilde{z}\tilde{M}^f$ into account, we obtain

$$\omega^{\mathrm{II}}_{1\,max}(H)=\Delta\varepsilon'+111-220\cos^2\vartheta,\quad \omega^{\mathrm{II}}_{2\,max}(H)=\Delta\varepsilon'+79-110\cos^2\vartheta. \tag{26}$$

In the case of a three-magnon satellite we also obtain two absorption maxima at the frequencies

$$\begin{aligned}\omega^{\mathrm{III}}_{1\,max}(H)=&{}^1/_2(E_1(\Gamma,f)+E_2(\Gamma,f))+2\varepsilon_1(\mathbf{k}_0)+\varepsilon_2(X)+W_1-W_2,\\ \omega^{\mathrm{III}}_{2\,max}(H)=&{}^1/_2(E_1(W,f)+E_2(W,f))+2\varepsilon_1(\mathbf{k}_0)+\varepsilon_2(\mathbf{k}_0)+W_1-W_2,\end{aligned} \tag{27}$$

where $W_1 \approx (2\cos^2\vartheta - 1)\cdot 58\ \mathrm{cm}^{-1}$ and $W_2 \approx -(2\cos^2\vartheta - 1)\cdot 20\ \mathrm{cm}^{-1}$ are respectively the values of $W^{\mathrm{fs\text{-}2s}}_{1n\alpha,m\beta}$ and $W^{\mathrm{fs\text{-}2s}}_{2n\alpha,m\beta}$ (15) for the nearest neighbors, and the points $\mathbf{k}_0$ also practically coincide with L. Using (19) and (20), we obtain finally

$$\omega^{\mathrm{III}}_{1\,max}(H)=\Delta\varepsilon'+105+31\cos^2\vartheta,\quad \omega^{\mathrm{III}}_{2\,max}(H)=\Delta\varepsilon'+148+48\cos^2\vartheta. \tag{28}$$

The value $\Delta\epsilon^f = 25\,173.5\ \mathrm{cm}^{-1}$ is obtained with the aid of (20) from the experimental data (Table I), recognizing that the exciton absorption (C_0) corresponds to the center of the Brillouin zone (the point Γ).

6. Thus an analysis of the absorption of light by magnon satellites of the purely excitonic ${}^6A_{1g}({}^6S) \rightarrow {}^4E_g({}^4G)$ transition, undertaken within the framework of the exciton model of spin and optical excitations using parameters determined from an experimental investigation of single-particle excitations, leads to the following results: The position of the maxima of the intensity of one-, two-, and three-magnon absorptions in a magnetic field $H > H_{cr}$ is determined by the expressions (cf. (22), (26), (28))

$$\begin{aligned}&\omega^{\mathrm{I}}_{max}=25223.5-64\cos^2\vartheta,\\ &\omega^{\mathrm{II}}_{1\,max}=25284.5-220\cos^2\vartheta,\quad \omega^{\mathrm{II}}_{2\,max}=25252.5-110\cos^2\vartheta,\\ &\omega^{\mathrm{III}}_{1\,max}=25278.5+31\cos^2\vartheta,\quad \omega^{\mathrm{III}}_{2\,max}=25321.5+48\cos^2\vartheta.\end{aligned} \tag{29}$$

The integral intensities of the one-, two-, and three-magnon absorptions are proportional to $\sin^4\vartheta$, $\sin^6\vartheta\cos^2\vartheta$, and $\sin^8\vartheta$, respectively. It follows therefore that the one-magnon and three-magnon absorptions are present also in the collinear case (H = 0), and become weaker with increasing magnetic field, the three-magnon absorption decreasing more abruptly. Two-magnon absorption appears only with violation of collinearity, and its intensity depends on the magnetic field in a more complicated manner, but in the experimental range of fields it is possible to obtain only a sharp increase of its intensity, leading to the appearance of the corresponding band in the optical spectrum.

We note now that one of the maxima of the two-magnon absorption (ω^{II}_{1max}) falls in the region of the three-magnon band, which has a maximum intensity in the absence of a magnetic field, and can hardly be observed. The remaining frequencies (29) actually correspond to the observed (Table I) absorption bands, and the magnetic-field induced changes of the integral intensities of these bands (Fig. 2) agree qualitatively with the predicted values.

CONCLUSION

Having at our disposal only qualitative data on the integral intensities of the bands of the investigated spectrum and on their behavior in a strong magnetic field, we did not attempt to obtain estimates of the relative intensities of magnon satellites of various types. To this end it is necessary to carry out a more rigorous analysis with a consistent application of the exciton scheme at all stages of the calculation. On the other hand, the qualitative aspect of the problem, particularly the character of the change of the integral intensities of the magnon-absorption band, is not affected by the assumed simplifications.

The frequency shifts of the intensity maxima of the magnon-absorption bands in a strong magnetic field, given by expressions (29), agree qualitatively with those observed (with the exception of C_4). We note that a more correct calculation calls for allowance for the renormalization of both the exciton and magnon energies and of the exciton-magnon interactions, thereby greatly complicating the problem. Inasmuch as the appearance of many terms in the renormalizations is due to violation of the collinearity in the magnetic field, the agreement between the calculated frequencies of the maxima of the multimagnon satellites with those observed in the absence of a magnetic field is all the more instructive.

Taking the foregoing into account, we conclude that the entire frontal part of the electric-dipole absorption spectrum in the region of the $^6A_{1g}(^6S) \rightarrow {}^4E_g, {}^4A_{1g}(^4G)$ transition of antiferromagnetic $RbMnF_3$ is due to magnon satellites of the $^6A_{1g}(^6S) \rightarrow {}^4E_g(^4G)$ exciton transition with participation of one, two, and three magnons. It has been shown experimentally and theoretically that the Davydov splitting of the exciton-single-magnon band is not observed even in a magnetic field sufficient for the appearance of such a splitting of a purely exciton band.

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[1)]The details of the calculations will be published elsewhere.

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