ZEEMAN EFFECT ON EXCITON-MAGNON BANDS IN ANTIFERROMAGNETIC MnF_2 CRYSTALS

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The appearance of additional satellite bands at $T < T_N$ was recently predicted theoretically [1] and demonstrated experimentally [2] with the ${}^6A_{1g}({}^6S) \to {}^4T_{1g}({}^4G)$ transition in MnF_2 (T_N = 68°K) as an example. The form of these bands and their spectral position agree with the assumption that a magnetic excitation (spin wave with maximum quasimomentum $\vec{K} = \pi/a$) is produced simultaneously with the electronic excitation (exciton). However, an experimental confirmation of the realization of such exciton-magnon transitions can hardly be regarded as convincing, since we are dealing here with one single band at ν = 18477 cm^{-1} (T = 2.2°K) among a large number of other bands capable of distorting the form of the investigated band and make it seem asymmetrical (the neighboring intense band ν = 18485 cm^{-1} is especially effective in this respect); in addition, an interval close to the maximum magnon frequency ($\omega_M \approx 55$ cm^{-1}) can occur among the large number of bands.

It is necessary in this connection to search for additional more direct and more unambiguous experimental proof of the realization of the simultaneous excitation of an exciton and a magnon. Most promising in this respect is a study of the response of the optical absorption spectrum to magnetic fields that are sufficiently strong to change the magnetic structure of an antiferromagnetic crystal. For MnF_2 this field is equal to 90 kOe ($H_z \parallel C_4$-axis). Magneto-optical investigations in this range of magnetic fields have become possible through the use of a pulse technique [3].

We have previously observed the influence of the strong magnetic field, sufficient to flop the spin structure of MnF_2 (spin flopping), on the structure of the optical transition ${}^6A_{1g}({}^6S) \to {}^4T_{2g}({}^4D)$ [4]. Magneto-optical effects in the region of these transitions are so large that they cannot be set in correspondence directly with the changes of the magnetic spectrum of the MnF_2 crystal during the flopping. Therefore, to observe more subtle effects it is necessary to turn to other parts of the light-absorption spectrum of MnF_2 where, as shown by earlier investigations [4,5], there are no abrupt changes in the frequencies and intensities of the bands.

We have investigated the narrow absorption bands due to the optical transitions ${}^6A_{1g} \to {}^4T_{1g}({}^4G)$ and ${}^6A_{1g} \to {}^4T_{1g}({}^4P)$. Their frequencies are listed in the table. The doublet 18473/18478 cm^{-1} must be associated with the two intense bands among the group of narrow absorption bands of the ${}^6A_{1g} \to {}^4T_{1g}({}^4G)$ transition of MnF_2 [2]. One of these bands, marked by

No	ν, cm^{-1}	Transition	T, °K
1	18473*	$^6A_{1g}(^6S) \rightarrow {}^4T_{1g}(^4G)$	4.2
2	18478	"	"
1	31942.7*	$^6A_{1g}(^6S) \rightarrow {}^4T_{1g}(^4P)$	20.4
2	32009	"	"
3	32065*	"	"
4	32095	"	"
5	32214.2	"	"
6	32481	"	"
7	32723	"	"

* Bands tentatively identified with exciton-magnon transitions

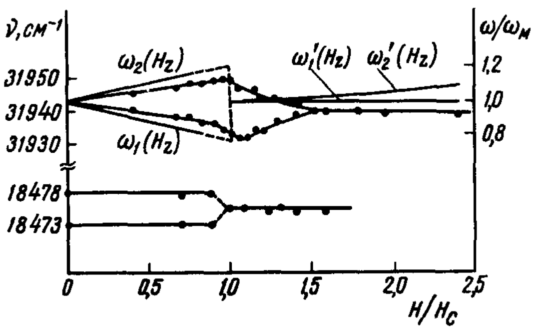

Fig. 1. Calculated and measured influence of a magnetic field on the light-absorption bands in antiferromagnetic MnF_2. $\omega_M \approx 55$ cm^{-1}, $H_c \simeq 9 \times 10^4$ Oe.

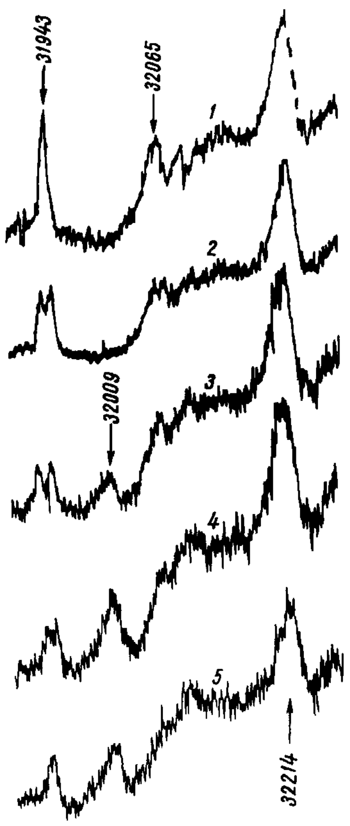

Fig. 2. Microphotographs of the portion of absorption spectrum of MnF_2 connected with the $^6A_{1g} \rightarrow {}^4T_{1g}(^4P)$ transition in the following magnetic fields: 1 - $H_z = 0$, 2 - $H_z = 61$ kOe, 3 - 95 kOe, 4 - 127 kOe, 5 - 217 kOe. T = 20.4°K.

an asterisk in the table, was interpreted as being an exciton-magnon band [2].

The influence of the magnetic field on the exciton-magnon light-absorption bands can be estimated qualitatively. It is necessary for this purpose to use the expression for the form and spectral position of the exciton-magnon band in the optical spectrum of an antiferromagnet [1,2] and the expression for the spin-wave spectrum in a magnetic field $H_z < H_c = \sqrt{2H_AH_B}$ and $H_z > H_c$ [6,7]. The results of such a calculation are illustrated graphically in Fig. 1.

The figure shows also the results of an experimental investigation of the influence of the strong magnetic field on the 18473/18478 cm^{-1} and 31943 cm^{-1} bands. In the latter case the ordinate scale is reduced 1/2.

The first striking fact is that for the 18473 cm^{-1} band, previously [2] identified with the exciton-magnon transition, there is not even qualitative similarity between the experimental behavior in the magnetic field and the calculation. This band is not split by an external field but, to the contrary, on reaching $H_z \simeq H_c$ it coalesces with its satellite at 18478 cm^{-1}, forming a single narrower and more intense band. These facts offer evidence that the interpretation of the 18473 cm^{-1} band as being an exciton-magnon transition is not the only one possible.

There is qualitative agreement between calculation and experiment in the behavior of the 31943 cm^{-1} band in a magnetic field. In weak magnetic fields, a clearly pronounced splitting into a doublet is observed (Figs. 1, 2), with the distance between components directly proportional to the magnetic field intensity H_z. The doublet splitting decreases when $H_z \approx H_c$ is reached, and disappears completely in a field $H_z \approx 1.5H_c$. It is in this non-monotonic dependence of the Zeeman splitting on the magnetic field intensity that we see a qualitative similarity between the experimental and calculated data. However, there is a noticeable disparity between the two: in the region $H > H_z$ the decrease in the splitting of the 31943 cm^{-1} band is smooth (from H_c to $1.5H_c$), and not abrupt as follows from calculation; the predicted splitting in the region $H_z \approx 2H_c$ is not observed. If we assume that this weak splitting is so small that it cannot be resolved by the spectral apparatus employed (DFS-13, linear dispersion 4 Å/mm), then we expect at least a shift of the band towards the shorter wavelengths, and not the longer ones as observed in the experiment. These facts demonstrate the need for a deeper theoretical analysis of the features of light absorption in antiferromagnetic crystals.

Worthy of special attention is the 32009 cm^{-1} band, which acquires a noticeable intensity only when $H_z > H_c$ is reached. It must be noted that the interval between the 32009 cm^{-1} band induced by the magnetic field and the 32065 cm^{-1} band is very close to the maximum magnon frequency $\omega_M = 54.5$ cm^{-1} in MnF_2 [8]. This circumstance, and also the results of magneto-optical investigations, make it possible to relate the 31943 and 32065 cm^{-1} bands to exciton-magnon transitions [1,2]. This statement is not contradicted by a careful analysis of the shape of these bands, which have noticeable asymmetry.

In conclusion it must be noted that the 32065 and 32214 cm^{-1} bands listed in the table apparently split in a magnetic field in a fashion similar to the 31943 cm^{-1} band, but the experimental accuracy is much lower here, in view of the considerable spread.

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