# ZEEMAN EFFECT IN THE OPTICAL SPECTRUM OF ANTIFERROMAGNETIC CRYSTALS OF $MnF_2$

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The influence of strong magnetic fields (up to $2 \times 10^5$ Oe) on the structure of the optical absorption spectrum of antiferromagnetic $MnF_2$ crystals ($T_N = 68°$ K) was investigated at T = 20.4° K. In the region of the optical transition ${}^6S_{5/2} \rightarrow {}^4D$, frequency shifts and splitting of the bands were observed in magnetic fields exceeding $H_c = \sqrt{2H_A H_E} = 95$ kOe, which is the field for the magnetic sublattice reversal in $MnF_2$.

THE energy level degeneracy in magnetically ordered crystals is lifted completely by the exchange field $H_E$. However, we may expect that in an external field the absorption bands are split due to the lifting of the sublattice degeneracy, since magnetic sublattices of an antiferromagnet become nonequivalent. Such splitting has been observed for $Cr_2O_3$,[1] and its anomalous behavior has been detected near the field for the magnetic sublattice reversal ($H_c = \sqrt{2H_A H_E}$, where $H_A$ is the anisotropy field).

In the present work, we investigated the influence of a magnetic field on the optical spectrum of $MnF_2$ crystals at 20° K. The measurements were carried out with the magnetic field oriented along the $C_4$ axis of tetragonal $MnF_2$ crystals, as well as at right-angles to this axis. Below the temperature $T_N = 68°$ K, manganese fluoride becomes antiferromagnetic with the ordering directed along the $C_4$ axis.[2] An estimate of $H_E$ gives a value of $5.5 \times 10^5$ Oe,[3] while $H_c$ amounts to about 95 kOe, both according to the antiferromagnetic resonance data[4] and according to the results on the magnetic susceptibility at 20° K.[5] Since the intensity of the most interesting magnetic fields is quite high, we used the method of investigating the magneto-optical effects in crystals[6] similar to the Kapitza pulse method[7]. The pulse nature of the method did not introduce any special features since the duration of application of the field was about $5 \times 10^{-3}$ sec, and the measurements of the magnetic susceptibility, illustrating the effect of the magnetic sublattice reversal in $MnF_2$, were carried out in fields of much shorter duration.[5]

The spectra were recorded using a type DFS-13 diffraction spectrograph with a linear dispersion of about 3 Å/mm.

We investigated the strongest, most clearly ob-

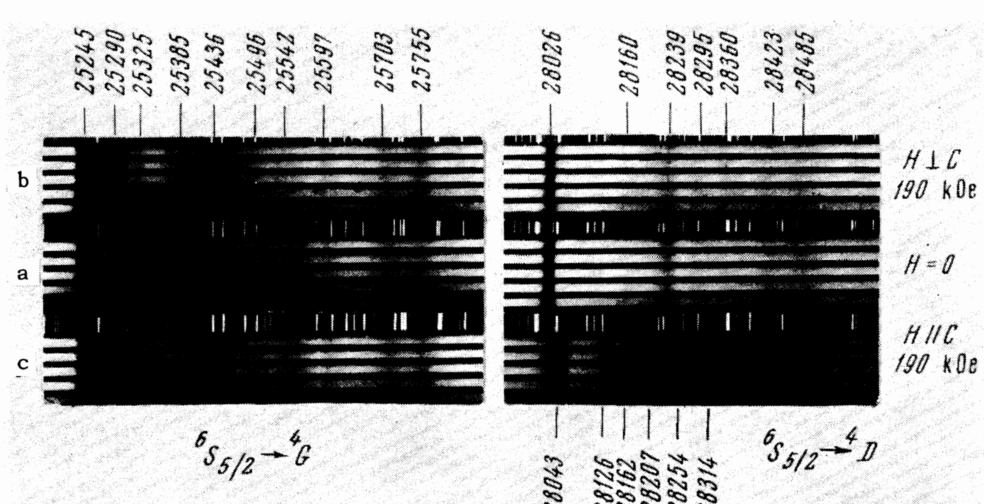


FIG. 1

served groups of absorption bands due to the transitions ${}^6S_{5/2} \rightarrow {}^4G$ and ${}^6S_{5/2} \rightarrow {}^4D$ in the $Mn^{2+}$ ion and lying, respectively, in the region of 3900 and 3500 Å.[8-11] Figure 1a shows the absorption spectrum of a $MnF_2$ crystal, 2 mm thick, at 20°K. The spectrum of this crystal has, apart from the intense bands observed earlier for thinner samples,[8-11] a group of weak bands both in the ${}^6S_{5/2} \rightarrow {}^4G$ region and in the ${}^6S_{5/2} \rightarrow {}^4D$ region. At present there is no way of interpreting with certainty the weak bands but the following should be noted: a) these bands are observed only at low temperatures $T < T_N$; b) there is some similarity between the weak-band positions in the ${}^6S \rightarrow {}^4G$ and ${}^6S \rightarrow {}^4D$ regions; in particular, this applies to the doublet 25703/25755 $cm^{-1}$ ($\Delta\nu = 52 \pm 5$ $cm^{-1}$) and the doublet 28403/28485 $cm^{-1}$ ($\Delta\nu = 62 \pm 5$ $cm^{-1}$). These bands may be due to optical transitions with the simultaneous excitation of the magnetic moment oscillations, by analogy with the additional bands observed earlier for other antiferromagnetic crystals.[11,12] Although the frequency intervals between the bands of the ${}^6S_{5/2} \rightarrow {}^4G$ group, as well as the ${}^6S_{5/2} \rightarrow {}^4D$ group, exceed considerably the antiferromagnetic resonance frequency of $MnF_2$ ($\approx 9$ $cm^{-1}$),[13,14] they are close to the values of the exchange splitting of the levels of the excited states ${}^4G$ and ${}^4D$.[8-10]

A magnetic field up to H = 190 kOe oriented at right-angles to the tetragonal axis does not change either in the ${}^6S \rightarrow {}^4G$ region or the ${}^6S \rightarrow {}^4D$ region (Fig. 1b). However, such a strong field, if oriented along the $C_4$ axis causes considerable changes in the region of the ${}^6S \rightarrow {}^4D$ transition (Fig. 1c): 1) an intense band at 28026 $cm^{-1}$ is displaced toward short wavelengths by 18—20 $cm^{-1}$; 2) the 28239 $cm^{-1}$ band is split by the 190 kOe field into a 28207/28254 $cm^{-1}$ doublet with its center of gravity displaced by 10 $cm^{-1}$ toward long wavelengths and the long-wavelength component considerably stronger; 3) a weak band at 28160 $cm^{-1}$ also splits into a 28126/28162 $cm^{-1}$ doublet with both components considerably more intense than the original band; the center of gravity is again displaced toward long wavelengths by about 13 $cm^{-1}$; 4) the 28295 $cm^{-1}$ band is displaced toward short wavelengths by 17 $cm^{-1}$; 5) weak short-wavelength satellites become strongly broadened so that their frequencies cannot be measured.

The influence of a 190 kOe field $\mathbf{H} \parallel C_4$ on the structure of the ${}^6S \rightarrow {}^4D$ transition is illustrated schematically in Fig. 2.

Measurements carried out in fields of lower intensity show that the effect described appears quite clearly in fields of about 90—100 kOe. Figure 3 illustrates the field dependences of the band frequencies observed in the region of the ${}^6S_{5/2} \rightarrow {}^4D$ transition in $MnF_2$. The strong change can be seen particularly clearly in the 90—100 kOe region, which coincides with the value of the field $H_c$ producing the magnetic sublattice reversal in $MnF_2$[5]. This, and the fact that the magneto-optical effect is only observed for $H \parallel C_4$, the only case when sublattice reversal can be expected,[5] allows us to conclude that both effects are closely related.

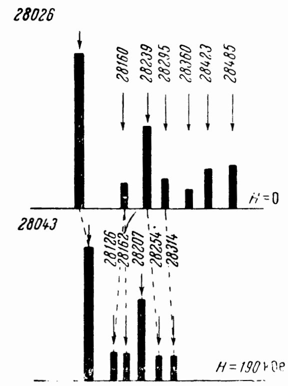

FIG. 2

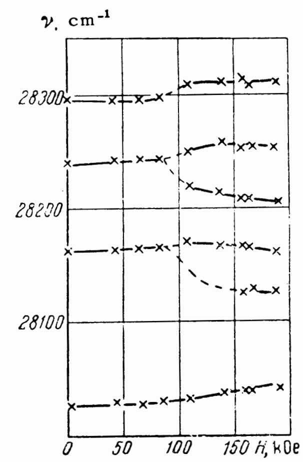

FIG. 3

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