Observation of spontaneous canting of the spontaneous sublattices in the hexagonal antiferromagnet $CsMnF_3$

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It is observed that the ratio of the intensities of certain electrodipole absorption bands in the optical spectrum of $CsMnF_3$ have hexagonal anisotropy along the direction of the external field **H** in the basal plane. The results offer evidence of the appearance of a spontaneous canting of the magnetic sublattices if the magnetic moments are oriented along the crystallographic direction [100].

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Examination of the magnetic properties of hexagonal antiferromagnets shows that in the expansion of the thermodynamic potential in powers of **m** and **l** higher than the second, terms appear that admit of the existence of a weak ferromagnetic moment directed along a sixfold axis.[1] Recent reports of observation of weak ferromagnetism in antiferromagnetic $RbMnCl_3$ (space group D_{6h}^4)[2] served as the first experimental confirmation of this theoretical conclusion. In this paper we report results of magneto-optical investigations of the crystal $CsMnF_3$, which has the same symmetry; the results can be attributed to the existence of a spontaneous weak canting of the magnetic moments of the sublattices.

We have previously[3] discussed in detail the features of the optical absorption spectrum of antiferromagnetic $CsMnF_3$ in a magnetic field. It was shown, in particular, that the behavior of the frequencies and of the intensities of most absorption bands connected with the $^6A_{1g} \rightarrow {}^4A_{1g}$, ^4E_g transition (the so called C group of bands) makes it possible to evaluate the canting of the magnetic moments of each of the sublattices by an external field. The results needed for this work consist in the following. In the C group of the absorption bands, the intensity of the electric dipole bands C_1 and C_2, which are due to optical excitation of the noncentrosymmetric ions[1)] Mn_2, depends essentially on the mutual orientation of the magnetic moments of the ions and the electric vector of the line: the C_1 band is observed at $\mathbf{E} \perp \mathbf{M}$, while C_2 is observed at $\mathbf{E} \parallel \mathbf{M}$. Application of an external field to the sample leads to the appearance, at the same frequencies, of two new bands (we shall call them for convenience C_1' and C_2'), the intensities of which are determined by the magnitude of the cant of the magnetic moments of the sublattices of the Mn_2 ions.

In the present study we have investigated the dependence of the intensities of the absorption bands C_1, C_2, C_1', and C_2' on the intensity and the direction of the stationary magnetic field relative to the crystallographic axes. The field 0–40 kOe was produced by a superconducting solenoid of Helmholtz construction. The absorption spectrum was registered with the aid of a DFS-13 spectrograph, at the exit from which was placed either a photographic film or a slit scanning at

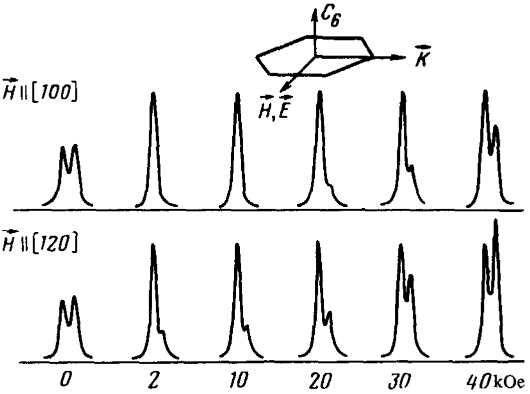

FIG. 1. Dependence of the intensities of the absorption bands in the σ spectrum on the magnitude and direction of the external magnetic field. Microphotograms.

a frequency 5 Hz and a photomultiplier. In the former case the photographs of the spectrum were investigated with the aid of an MF-2 microphotometer adapted for use with an automatic recorder of the spectrograms, and in the latter case the scanned section of the spectrum was fed directly to a long-persistence oscilloscope S8-2. The measurements were performed on samples of thickness 0.5 mm (δ spectra) and 1.7 mm (α spectra) in an optical cryostat at a temperature 4.2 K.

Figure 1 shows microphotograms of a section of the spectrum in the frequency interval C_1—C_2 for the following experimental geometry: $\mathbf{k}\perp C_6$, $\mathbf{E}\perp C_6$, $\mathbf{H}\perp C_6$, and $\mathbf{H}\perp\mathbf{k}$ (σ spectrum). It is seen that whereas in a zero field the spectrum contains both bands C_1 and C_2 (the magnetic moments in the basal plane have arbitrary directions, and the average values of the projections of **M** along and across **E** are equal), when the field is applied along the [100] axis, total reorientation of the magnetic moments into a single-domain structure $\mathbf{M}\perp\mathbf{H}$ takes place even at a low field intensity, and all that is left of the doublet of

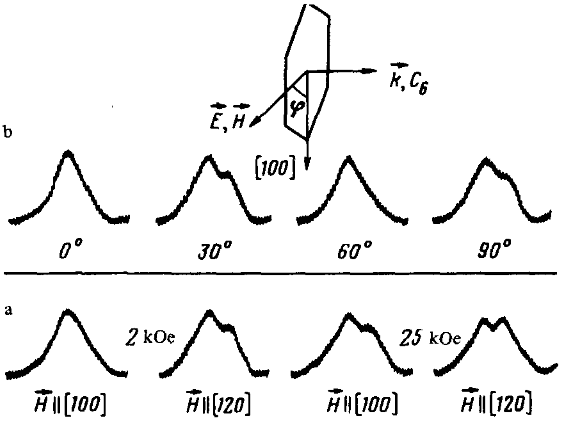

FIG. 2. (a) Comparison of the ratio of the intensities of the bands C_1 and C_2' in the α spectrum at different directions of **H** in the basal plane.(b) Variation of the form of the spectrum as a function of the angle ϕ between the direction of **H** and the chosen [100] axis at H = 4 kOe. Oscillograms.

bands C_1 and C_2 is C_1(E ⊥ H), the intensity of which, naturally, increases. With increasing field intensity, when the cant angle of the moments becomes noticable (3.6° at 20 kOe and 7.2° at 40 kOe for the Mn_2 sublattices),[3] the band C_2' appears and its intensity increases. On the other hand, if the field is directed along the [120] axis, the band C_2' is present in the spectrum starting with the minimal field. With increasing field intensity, the intensity of this band increases at a faster rate than in the case of H ∥ [100]. As seen from Fig. 1, the initial intensity of C_2' at H = 2 kOe and H ∥ [120] is the same as in a field H ≈ 25 kOe and H ∥ [100]. Figure 2 shows oscillograms of the investigated section of the spectrum at k ∥ C_6 (α spectrum). It is seen that the ratios of the band intensities follow the same regularity as in the σ spectrum. The field at which the intensity C_2' at H ∥ [100] reaches its initial value at H ∥ [120] is also 25 kOe. When the sample is rotated in the field, the form of the spectrum repeats itself with a period 60° (Fig. 2b). We note that on going to the geometry E ⊥ H, meaning that therefore E ∥ M, the picture of the transformation in the spectrum changes symmetrically, namely, the C_2 band remains in a weak field, and C_1' appears when the intensity is increased. As to the polarization features of the bands C_1' and C_2' we can draw the following conclusion: in analogy with the case C_1 and C_2, they turn out to be sensitive to the mutual orientation of the vectors E and M, with C_1' observed at E ∥ M and C_2' observed at E ∥ M and C_2' at E ⊥ M. This agrees with the results of our measurements in strong fields (up to 300 kOe).

The presented experimental data make it possible, in our opinion, to draw the following conclusion: when the magnetic moments are located in the basal plane along the [100] directions, a spontaneous cant of the magnetic moments of the sublattices takes place in the crystal, and its value, for the sublattices of the noncentrosymmetric ions Mn_2, is the same as in the external field 25 kOe, if the latter aligns the moments along the crystallographic axis [120]. As already noted, the theory admits of a possible canting, which should be directed along a sixfold axis. It might seem that the last statement and our experimental results allow us to speak of a Dzyaloshinskiĭ field in the crystal amounting to $H_D \approx 25$ kOe. We shall refrain, however, from making this quantitative estimate, which is valid only if it is certain that the effective fields H_D are equal at the sublattices of the ions Mn_1 and Mn_2. At present there are no such data. Furthermore, the face that magnetistatic and resonance measurements do not reveal a ferromagnetic moment in this crystal[4,5] suggests that these quantities are essentially not equal. Calculation with the data of[6] shows, e.g., that the canting of the magnetic moments of the Mn_2 sublattices turns out to be the same as in an external field 25 kOe, and that the net moment over all sublattices is zero if the effective fields at the ions Mn_1 and Mn_2 are set equal to 52 and 14.5 kOe, respectively.

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