Magnetic circular dichroism in antiferromagnetic cobalt fluoride

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Results are presented of an experimental investigation of magnetic circular dichroism (MCD) of antiferromagnetic cobalt fluoride, for different orientations of the magnetic field: $\mathbf{H}\|[001]$, $\mathbf{H}\|[110]$, and $\mathbf{H}\|[11h]$. MCD that is even in the magnetic field intensity was observed, and it is shown that its sign depends on the antiferromagnetic state of the sample. It is shown that the spectra of the MCD linear and quadratic in the magnetic field strength, obtained in Faraday and Voigt experimental geometries, carry independent information on the electronic transitions. In both geometries, the MCD is due mainly to lifting of the sublattice degeneracy of the energy levels of the cobalt ions. The observed singularities of the dispersion dependence of the MCD in the region of the exciton-magnon band, with maximum absorption at the frequency 22769 cm^{-1}, are discussed.

1. INTRODUCTION

In antiferromagnetic (AFM) crystals such as CoF_2, whose symmetry is described by a magnetic point group $\underline{4}/mm\underline{m}$ containing the odd operations 4_z^- and 2_d^-, a magnetic field applied parallel to the $[110]\|X'$, axis leads to the appearance of a magnetic-moment component directed along the tetragonal axis $C_4\|Z$. The possibility of appearance of a Z component of the magnetization at $\mathbf{H}\perp Z$ follows from the presence of an invariant of the type $fM_zM_xM_yL_z$ in the thermodynamic potential of a two-sublatice tetragonal AFM characterized by an AFM-ordering symmetry $4_z^-2_d^-$ (Refs. 1–3). It is seen also from a symmetry analysis of the the magnetization, quadratic in $\mathbf{H}$, of antiferromagnets that have no anti-inversion centers.[4] One of the mechanisms that produces magnetization along the [001] axis in a field $\mathbf{H}\|[110]$ can be discerned by considering the crystal structure of CoF_2. Its unit cell (see Fig. 1) contains two magnetic Co^{2+} ions surrounded by distorted octahedra of F^- ions, whose symmetry is described by the *mmm* group. The elongated axes of the two ligand octahedra that surround the cobalt ions from different magnetic sublattices (in Fig 1—along X' and Y' for sublattices 1 and 2 respectively) are mutually rotated trough 90° around the tetragonal axis of the crystal. The low symmetry of the ligand surround and its orientational nonequivalence for the spins of the ions $Co^{2+}1$ and $Co^{2+}2$ causes different deflections of the magnetic moments of the sublattices $\mathbf{M}_1$ and $\mathbf{M}_2$ from the $C_4\|Z$ axis under the influence of a field $\mathbf{H}\|[110]$. As a result, there is no cancellation of the Z components of the moments, $M_{1z} \neq M_{2z}$, and the magneic moment has a resultant Z component proportional to the square of the field strength. A certain contribution is made also by the changes in the spins S_1 and S_2 as a result of the nonequivalent mixing of the quantum-mechanical states of the ions $Co^{2+}1$ and $Co^{2+}2$ under a magnetostrictive lattice deformation that is quadratic in $\mathbf{H}$ and under the direct action of the magnetic field.[5,6] The sign of $M_z = M_{z1} - M_{z2}$ depends on the spin orientations of the ions of the first and second sublattices. When the sublattice magnetization is reversed, the sign of M_z sould change. Magnetizations M_z induced in CoF_2 by a field $\mathbf{H}\|[110]$ were observed in Ref. 7.

The appearance of a Z component of magnetization should lead to magnetic gyrotropic properties—magnetic rotation of the polarization plane and magnetic circular dichroism, quadratic in the field strength. A dependence, quadratic in the field, of the antisymmetric components of the dielectric tensor ε_{ij}^a, which describe the magnetic gyrotropic properties, is allowed also by the Onsager symmetry restrictions [8,9] generalized to include magnetically ordered media. A necessary condition for the appearance of quadratic magnetic rotation and circular dichroism is the absence of an anti-inversion center, which takes place in all pizeomagnets, including the AFM CoF_2 ($T_N = 37.8$ K).

In the case considered, the magnetic field $\mathbf{H}\|[110]$ shifts in different ways the energy levels of the ions $Co^{2+}1$ and $Co^{2+}2$ and mixes in different ways their quantum-mechanical states, making the sublattices noneqivalent—the AFM CoF_2 acquires ferrimagnetic properties. The partial contributions of the sublattices to the magnetooptic properties, normalized to unity magnetic moment, can become unequal. In this situation, the magnetic gyrotropic properties for light propagating along the Z axis are allowed also at $M_Z = 0$. This property is reflected in the expansion of ε_{XX}^a in terms the antiferromagnetic and ferromagnetic vectors. The expansions contain terms proportional to $L_XL_YL_Z$ and

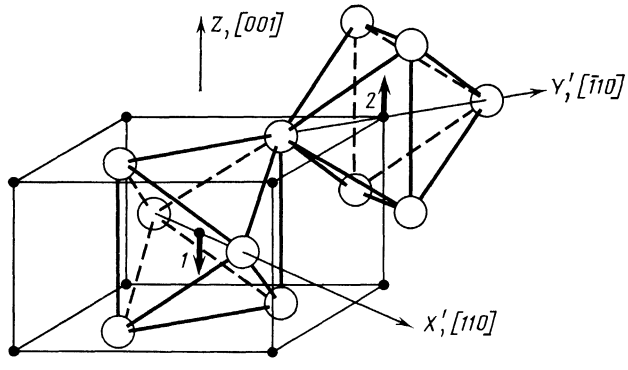

FIG. 1. Orientational nonequivalence of the ligand surround of the ions $Co^{2+}1$ and $Co^{2+}2$ in a CoF_2 crystal. The elongated axes of the octahedra are directed along X' and Y' for sublattices 1 and 2, respectively.

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$M_X M_Y L_Z$, which are transformed in the same way as the Z components of an axial magnetic vector. It is clear from this alone that for light with a wave vector $\mathbf{k}\|[001]$ the magnetic rotations and the circular dichroism, due to the field $\mathbf{H}\|[001]$ and the field $\mathbf{H}\|[110]$ can qualitatively differ even if they produce equal Z components of the magnetization. The quadratic and linear magnetic rotations of the light-polarization plane, measured in the transparency region at $\lambda = 6328$ Å and normalized to the magnetization M_Z, turned out to be close.[10] In the region of separate optical transitions, however, the difference between the two effects, both in rotation and in dichroism, can be substantial.

We present here the results of an investigation of magnetic circular dichroism for light propagating in the AFM CoF_2 along the C_4 axis at different field orientations: $\mathbf{H}\|[110]$, $\mathbf{H}\|[001]$, $\mathbf{H}\|[11h]$. The measurements were made in the region of absorption bands connected with $3d$ excitations of Co^{2+} ions in the visible region of the spectrum.

2. FEATURES OF EXPERIMENTAL PROCEDURE

As noted earlier, the effects of quadratic magnetic gyration reverse sign when the directions of the AFM sublattice moments are reversed. We had therefore to perform the experiments with homogeneously ordered samples. To make them single-domain and to reverse their magnetization we used the previousy mentioned effect of quadratic magnetization.[4,7,11] The magnetic field was applied in the (110) plane. The field component $H_{[110]}$ induced a moment

$$M_z = |C_{zxy}| H_x H_y \operatorname{sign} L_z,$$

and the component H_Z made the AFM^+ and AFM^- states, which differ in the sign of L_Z, energywise nonequivalent. The oblique field was produced by two superconducting solenoids whose axes were oriented along [110] and [001]. The field strength of each solenoid could be varied independently from 0 to 25 kOe. The homogeneity of the AFM state of the sample was monitored visually with the aid of the linear magnetooptic effect.[12]

The experimental results were obtained with an automated facility based on the universal spectral computing assembly KSVU-6. The facility can record, by a modulation procedure, the spectra of the circular and linear dichroism and of the absorption at temperatures above 6 K in a longitudinal field of strength up to 75 kOe, or else in an oblique field with components H_Z along the direction of the light propagation and $H_\perp$ perpendicular to it. The two components could be varied independently from 0 to 25 kOe. The facility permits also a reduction of the spectra with the "Elektronika DZ-28" computer. A piezooptic polarization modulator operating at 18 kHz was used. The modulation amplitude was set at 2.41 rad automatically for each wavelength.[13] Owing to the presence of feedback of the average photo-receiver current, the lock-in amplifier UPI-1 records and transmits to the computer a signal proportional to the dichroism

$$D(\nu) = \frac{I^+(\nu) - I^-(\nu)}{I^+(\nu) + I^-(\nu)},$$

where $I^+(\nu)$ and $I^-(\nu)$ are the intensities of the right- and left-polarized light passing through the sample. The sensitivity of the dichroism-measurement channel depends strongly on the intensity of the received light, and reaches 10^{-4} under "stringent" conditions [narrow mochromator spectral slits (about 50 μm), appreciable absorption in the sample ($I/I_0 \approx 0.2$), small single-domain observation section (about 0.1×0.1 mm)]. The dichroism and absorption spectra were recorded simultaneously. The reduced widths of the slits in the described experiments were 1, 2 and 4 cm^{-1} in the frequency regions 13 000, 23 000, and 25 000 cm^{-1}, respectively.

Particular attention in the circular dichroism measurements was paid to prevention of false signals and of distortion of the true form of the spectral curves, which can result either from possible inhomogeneities in the optical-channel elements, or from the presence of an induced linear anisotropy in the sample itself. The main feature of the appratus, from this standpoint, is the use a cryostat without cold windows. The sample was placed in a vacuum cavity inside the cryostat and was secured to the cold finger by a special paste that remained elastic when cooled and prevented mechanical stresses. Other precautionary measures will be described below in the discussion of the nontrivial dispersive dependence of the magnetic circular dichroism near the 22 769 cm^{-1} exciton-magnon band.

3. RESULTS AND DISCUSSION

In a perpendicular experimental geometry $\mathbf{H}\perp\mathbf{k}$ with $\mathbf{H}\|[110]$ and $\mathbf{k}\|[001]$ the magnetic circular dichroism (MCD) is distinctly observed. As expected, its sign remains unchanged when the field direction is reversed, but changes following AFM magnetization reversal of the sample. The change of the sign of the dichroism when the magnetic moments of the sublattices become reoriented in the sample is illustrated in Fig.2, which shows, the dichroism spectra obtained in succession for the two AFM state of the sample, AFM^+ and AFM^-.

The spectra of the MCD quadratic in the magnetic field, obtained in a transverse experimental geometry (Voigt geometry), are compared in Figs. 3–5 with the spectra of the MCD linear in the magnetic field, obtained in longitudinal (Faraday) geometry. The figures demonstrate also the change of the MCD spectra in an oblique field oriented in a plane such as (110), with increase of the Z component of the magnetic field. As seen from these figures, there are pronounced differences between the spectra of the quadratic and linear MCD: the dichroism is weakly pronounced in the

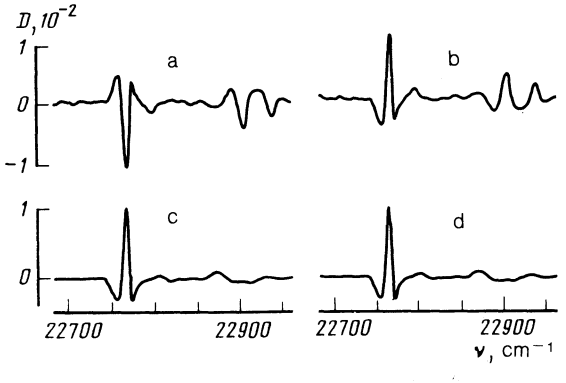

FIG. 2. Dependence of the sign of the magnetic circular dichroism on the antiferromagnetic state of the crystal and on the field orientation: a, b— $\mathbf{H}\perp C_4$, $H = 20$ kOe a) $A\Phi M^+$, M_Z^+; b) $A\Phi M^-$, M_Z^-; c, d— $\mathbf{H}\|C_4$, $H = 5$ kOe; c) $A\Phi M^+$, M_Z^+; d) $A\Phi M^-$, M_Z^+.

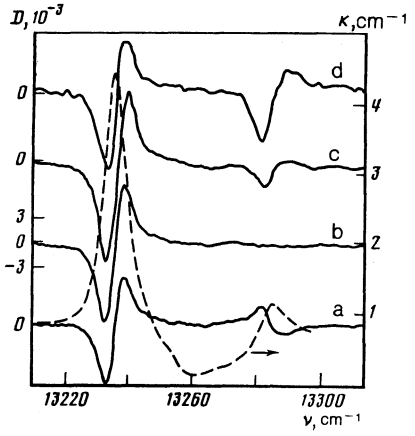

FIG. 3. Comparison of MCD spectra in magnetic fields of different direction in the region of the transitions ${}^4\Gamma_4^+ ({}^4F) \rightarrow {}^4\Gamma_2^+ ({}^4F)$: a) $H_Z = 0$, $H_{X'} = 24$ kOe; b) $H_Z = 6$ kOe, $H_{X'} = 24$ kOe; c) $H_Z = 12$ kOe, $H_{X'} = 24$ kOe; d) $H_Z = 20$ kOe; $H_{X'} = 0$. Dashed line—absorption spectrum.

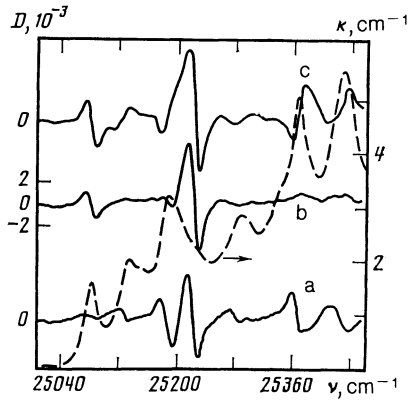

FIG. 5. MCD spectra in oblique magnetic field in the region of the transitions ${}^4\Gamma_4^+ ({}^4F) \rightarrow \Gamma_5 ({}^2D)$: a) $H_z = 0$, $H_{X'} = 24$ kOe; b) $H_Z = 6.5$ kOe, $H_{X'} = 24$ kOe; c) $H_z = 13$ kOe, $H_{X'} = 24$ kOe. Dashed line—absorption spectrum.

longitudinal geometry and well manifested in the transverse (group of bands in the interval 22 880–22 950 cm^{-1}); the dichroism is better manifested in longitudinal than in transverse geometry (25 083 cm^{-1} band).[1)] The signs of the linear and quadratic MCD are different for one and the same sign of the Z component of the magnetic moment and for one and the same AFM state of the sample (the 13 283, 22 769, 23 047, 25 129, 25 191, 25 366, 25 428 cm^{-1} bands).

The invariance of the AFM state and the constancy of the magnetic-moment Z-component sign in the experiment were ensured by the fact that the sign of the component H_X in MCD measurements in transverse geometry and the sign of H_Z in MCD measurements in longitudinal geometry of the experiment remained the same as when the sample was made single-domain. The invariance of the AFM state of the sample was monitored visually via the linear magnetooptic effect.[12] The signs of the MCD in two experimental geometries turned out to be the same only in certain bands (13 236 and 25 221 cm^{-1}).

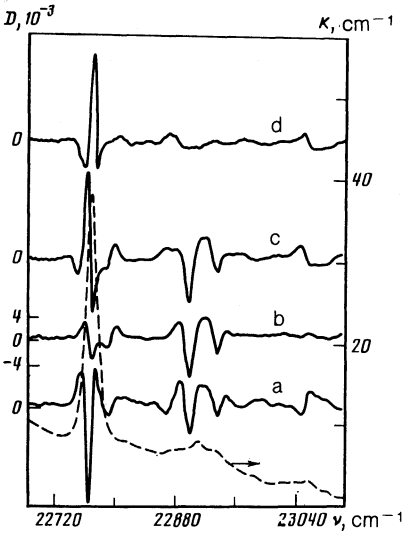

FIG. 4. Comparison of MCD spectra in magnetic fields of different direction in the region of the transitions ${}^4\Gamma_4^+ ({}^4F) \rightarrow {}^2\Gamma_1^+ ({}^2G)$, ${}^2\Gamma_4^+ ({}^2P)$: a) $H_Z = 0$, $H_{X'} = 24$ kOe; b) $H_Z = 5$ kOe, $H_{X'} = 24$ kOe; c) $H_Z = 10$ kOe, $H_{X'} = 24$ kOe; d) $H_Z = 5$ kOe, $H_{X'} = 0$. Dashed line—absorption spectrum.

It must be noted first of all that circular dichroism is observed in transverse geometry of the experiment in all the bands of the inestigated sections of the absorption spectrum. This indicates that all the observed absorption bands are connected with electronic transitions in the magnetically ordered subsystem of the crystal ions. In contrast to the case of longitudinal geometry, in transverse geometry the MCD spectrum cannot show optical transitions due to impurity ions that do not interact with the AFM subsystem of the Co^{2+} ions. Another well pronounced common feature of the MCD spectra is that all the isolated bands (the 22 769 cm^{-1} band calls for a separate examination) have an S-like dispersion, while the dispersion of the MCD of overlapping bands can be represented as a superposition of S-shaped curves. This means that the MCD is due to splitting of each band into two components with right- and left-elliptic polarization. Obviously, the cause of the splitting is the lifting of the sublattice degeneracy of the Co^{2+}-ion energy levels.

The degeneracy is lifted by the difference in the orientations of the ligand octahedra surrounding the cobalt ions in the first and second sublattices relative to the [110] direction. Taking into account the orthorhombic symmetry of the local magnetic anisotropy, we can find the shifts of the energy levels of the cobalt ions from the first and second sublattices in the field $\mathbf{H} \| [110]$. From the results of Ref. 5, in which the magnetic susceptibility $\chi_{X',X'}$ of CoF_2 was calculated with allowance for the nearby Kramers doublet in the spectrum of the Co^{2+} ion, we can obtain an expression for the difference between the energies of the ground states of $Co^{2+}1$ and $Co^{2+}2$ ions from different sublattices in a field $\mathbf{H} \| [110]$:

$$\Delta E_0 = \frac{(f_1 g_{xx}^2 - f_2 g_{YY}^2)\,\mu_B^2 H^2}{2[1 - f_1 f_2 (z_2 J_2)^2]}. \qquad (1)$$

Here $g_{xx} = 2.75$ *and* $g_{yy} = 2.08$ are g-factors, $z_2 J_z = 35$ cm^{-1} is the parameter of exchange interaction of the Co^{2+} ion with its nearest neighbors from the other sublattice, and $f_1 = 0.0480$ and $f_2 = 0.0085$ are determined by the axial and rhombic anisotropy parameters and by the intersublattice exchange interaction. The exchange interaction between ions of the same sublattice is assumed to be zero. An estimate yields $\Delta E_0 / H_{X'}^2 \approx 7 \cdot 10^{-10}$ $cm^{-1} \cdot Oe^{-2}$. In a field $H_{X'} = 27$ kOe we get for ΔE_0 a value 0.5 cm^{-1}. The spectroscopic

splitting for the purely excitonic bands is determined by the difference between the level shifts of the ground and excited states in the two sublattices:

$$\Delta E_e = |\Delta E_0 - \Delta E^*|. \quad (2)$$

The splitting of the exciton-magnon band should be different because of the energy splitting of the magnon branch on the edge of the Brillouin zone in a field $\mathbf{H}\|[110]$. The splitting should be close to

$$\Delta E_{e\text{-}m} = \Delta E_0 + \Delta E^*. \quad (3)$$

Although we cannot know the splitting of the excited energy level, it is nonetheless of interest to compare ΔE_0 with the band splitting due to MCD. To estimate ΔE we choose an isolated band. A well pronounced symmetric S-shaped dispersion dependence is possessed by the 13 236 cm^{-1} band (see Fig. 3). Assuming that the polarization of the absorbed modes is circular and that the bands connected with both sublattices are not deformed when displaced in a field , we get $\Delta E \approx 0.6$ cm^{-1}. This value differs somewhat from the real splitting. To determine band splitting due to dichroism we must know the dispersion relation for dichroism corresponding to polarization of the absorbed optical modes. This spectrum can be reconstructed from the circular and linear dichroism spectra. It is not our aim here to determine rigorously the value of ΔE for the obsered bands. However, the proximity of the approximated ΔE to the difference between the shift of the ground energy levels of the $Co^{2+}1$ and $Co^{2+}2$ ions indicates only that the considered mechanism producing the quadratic MCD and connected with the lifting of the energy degenracy of the $Co^{2+}1$ and $Co^{2+}2$ ions is acceptable.

A change of the spin as a result of quantum-mechanical mixing of the states under the influence of a field $\mathbf{H}\|[110]$, which was estimated in Ref. 6, is not noticeably manifested in experiment. Were this mechanism the principal one, the dichroism would be due to a change of the probabilities of optical transitions for right- and left-hand polarizations to a greater degree than to a shift of the transition frequencies. In this case the dichroism curves should have an Λ-shaped dispersion. No such curves were observed in the investigated spectral regions.

Attention is called to the circumstance that the 22 769 cm^{-1} exciton-magnon band (see Fig. 4) has a dispersive MCD dependence which is not typical of a single band, and its dispersive dependences in the longitudinal and transverse geometries are close. It is known that measurement of circular dichroism in an anisotropic medium calls for special caution in view of the influence of the linear dichroism and of the linear birefringence on the observation result.[15-17] Both factors take place in our case also when the crystal is strictly oriented with $\mathbf{k}\|C_4$, owing to the linear magnetooptic effect for longitudinal geometry, and owing to the Voigt or the Cotton-Mouton effect for transverse geometry. To decrease the influence of linear birefringence and dichroism we used a thin sample ($d = 0.46$ mm) and a weak magnetic field, to ensure small phase shifts of the optical modes. In addition, the MCD was recorded for different combinations of the signs of the magnetic circular dichroism and of the linear birefringence and dicrhroism. The reproducibility of the spectral form of the MCD in these experiments is evidence that the observed shape of the curve is not the result of parasitic effects.

Such a dispersion of the dichroism can be possessed by two closely lying bands with approximately equal but opposite dichroism. Choosing the parameters of the assumed bands in the approximation of "hard" splitting, good agreement can be reached between the resultant dispersive MCD dependence and the observed one (Fig. 6a). The 22 769 cm^{-1} band exhibits a similar "anomalous" form of MCD dispersion in a longitudinal $\mathbf{k}\|\mathbf{H}[001]$ geometry (see Fig. 4). By choosing the components H_Z and H_X, it is possible to achieve almost complete compensation of only one of the suggested bands. It can be seen from Fig. 4 that the uncompensated part of the curve is S-shaped, and the position of its midpoint coincides with the position of the absorption maximum of the exciton-magnon band.

The MCD of the 22 769 cm^{-1} band in the $\mathbf{H}\|[001]\|\mathbf{k}$ geometry was investigated earlier in Ref. 18. However, the MCD dispersion curve in that reference is distinctly S-shaped, patently different from that obtained by us. On the other hand, our own earlier investigations of linear dichroism[19] have shown that the 22 769 cm^{-1} band shows an S-shaped dispersion of the magnetic linear dichroism in a field $\mathbf{H}\|[001]$. Attention is called, however, to the smaller amplitude of the low-frequency extremum and to the small asymmetry of the curve—a weakly pronounced shoulder is noted on the low-frequency wing (see Fig. 6b). Assuming the presence of a weak dichroic absorption coponent, we can simultaneously obtain the indicated ingularities on an initially symmetric curve, which follows under the condition of "hard" splitting of the observed absorption band (see Fig. 6b). Good agreement between the dispersion curves of the linear and circular magnetic dichroisms can be achieved only by assuming that the MCD and MLD of the "additional" weak band are close to each other, whereas the dichroisms of the principal band differ by an order of magnitude. The results of a joint computer reduction of the MCD and MLD spectra and of the absorption coefficient, shown in

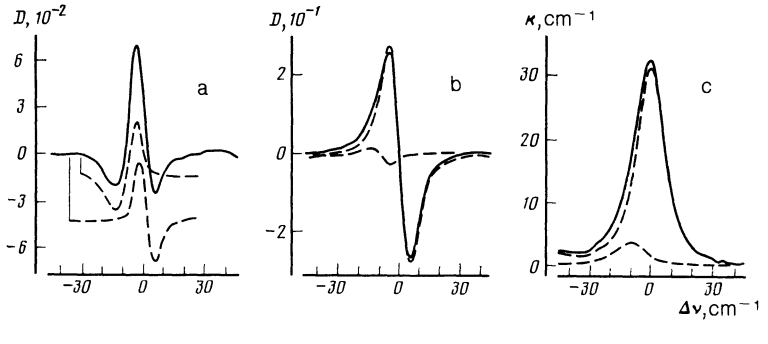

FIG. 6. Spectra of circular (a) and linear (b) magnetic dichroism and of the absorption (c) in the 22 769 cm^{-1} exciton-magnon absorption band, represented in the form of a superposition of two bands. Solid curves—experiment, dashed—reduction; $H_z = 40$ kOe.

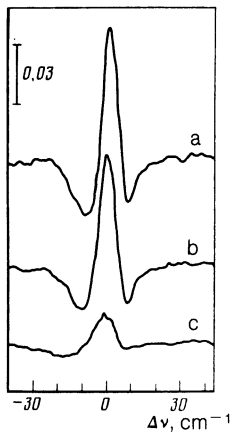

FIG. 7. Temperature dependences of the MCD spectrum in the 22 769 cm^{-1} exciton-magnon absorption band; $H_z = 40$ kOe; a) $T = 6.8$ K; b) $T = 18$ K; c) $T = 28$ K.

Fig. 6, were obtained under the assumption that both lines undergo the same splitting in a magnetic field. The distance between the absorption maxima of the bands turned out to be 9.6 cm^{-1}, the intensity ratio is close to 10, and the spectroscopic g factor is equal to $g_Z = 2.54$. The last value coincides with the data of Ref. 20. The temperature-induced changes of the MCD in the region of this exciton-magnon absorption are shown in Fig. 7, It is more likely that the dispersion curve varies as one whole.

The qualitative difference beteen the MCD spectra obtained in the region of the 22 769 cm^{-1} band in Ref. 18 and in the present paper is due, in our opinion, to the fact that in the MCD measurements of Ref. 18 no sufficient measures were taken to exclude the influence of the linear dichroism, the presence of which was not assumed in the Faraday experimental geometry with $\mathbf{k}\|C_4$ in Ref. 18.

It is difficult to draw a definite conclusion concerning the osberved singularity of the MCD. It is possible that the distinctive form of the MCD dispersion is indeed connected with the presence of an additional absorption band that overlaps the exciton-magnon absorption. In this case it must be noted that in experiments on neutron diffraction in AFM CoF_2 (Ref. 21) there was observed, in addition to the phonon and magnon modes, also an unidentified excitation whose energy was close to the magnon energy at the boundary of the Brillouin zone. It is not excluded that the additional band that has appeared in the MCD spectrum is due to a two-particle transition in which this excitation participates. It is also possible, however, that the complicated form of the MCD dispersion is due to the character of the exciton-magnon absorption. In two-particle absorption, excitons and magnons having different momenta take part in the process. Their contributions to the MCD may differ, since transitions with excitation of excitons and magnons at different points of the Brillouin zone can be caused by light with different polarization. In this case the model of "hard" splitting of the band may turn out to be too crude for the description of the MCD. The 9.6 cm^{-1} interval singled out in the MCD spectrum should correspond then to the sum of the energy distances between the magnon and exciton Brillouin-zone points that are magnetooptically most active. Final conclusions concerning the nature of the MCD dispersion in this region of the spectrum can be drawn only after a thorough analysis of all the possibilities.

As to the other bands in the MCD spectrum, attention is called to bands that can be completely cancelled out in a magnetic field. These bands are apparently of a simpler nature. Attention is called to the 340 cm^{-1} interval between the pure exciton bands 23 070 cm^{-1} and 25 026 cm^{-1} (Ref 14), on the one hand, and the 23 047 cm^{-1} and 25 366 cm^{-1} bands of the same kind. This interval is close to the frequency of the M_5 phonon at the Z point of the Brillouin zone.[21] It is typical also that the MCD cancellation in bands belonging to one group of transitions takes place in practically in one and the same field. This circumstance favors the assumption that such bands are exciton-phonon satellites, whose properties in a magnetic field should be close to the properties of the excitonic transition.

Since the values of the MCD linear and quadratic in the field turned out to be comparable in easily attainable fields, it should be noted that for oblique orientation of the magnetic field the connection between the MCD and the component of the magnetization along the light propagation direction becomes more complicated than in the case of transverse or longitudinal orientations. The the dichroism and the component of the magnetization can be written for $\mathbf{k}\|Z$ in the form

$$D = aH_z + bH_{x'}^2, \tag{4a}$$

$$M_z = \chi_{zz}H_z + C_{zx'x'}H_{x'}^2. \tag{4b}$$

The different signs of the coefficients a and b for the same direction of M_Z, demonstrated in Figs. 3–5, is evidence that the coefficients in Eqs. (4a) and (4b) are not proportional: $a/\chi_{zz} \neq b/C_{zx'x'}$. As a result, the linear relation between the MCD and the magnetization M_Z is lost even while the MCD is still proportional to M_Z at the symmetric field orientations $\mathbf{H}\|C_4$ and $\mathbf{H}\|[110]$.

4. CONCLUSIONS

The experimental investigations of the linear and quadratic (in $\mathbf{H}$) magnetic circular dichroism show the following:

1. The MCD spectra obtained in Faraday ($\mathbf{H}\|k$) and Voigt ($\mathbf{H}\perp\mathbf{k}$) experimental geometries carry independent information and can be useful for the identification of the absorption bands of a crystal, and also to determine the connection between impurity ions and the magnetic subsystem of the matrix.

2. The form of the MCD dispersion curves offers evidence that in the investigated sections of the spectrum the MCD is due mainly to lifting of the sublattice degeneracy of the energy levels of the AFM CoF_2.

The features of the dispersion dependence of the MCD in the 22 769 cm^{-1} exciton-magnon absorption band show that the transition has a complicated structure, and necessitate a thorough additional analysis.

[1)]Since it was not our purpose to determine exactly the frequencies of the absorption-band maxima, we cite here and elsewhere the values from Ref. 14.

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