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STRESS INDUCED LINEAR DICHROISM OF ANTIFERROMAGNETIC PEROVSKITES : KNiF₃, KCoF₃ AND RbCoF₃

I. — MAGNETIC ORIGIN AND EVIDENCE FOR DOMAIN TWIN-WALLS MOVEMENTS

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Résumé. — Nous considérons le dichroïsme linéaire (DL) sous contrainte de KNiF₃, KCoF₃ et RbCoF₃, au-dessus et au-dessous de la température de Néel. Au-dessus de T_N , le dichroïsme sous une contrainte S parallèle à $\langle 001 \rangle$ varie linéairement en fonction de S tandis qu'au-dessous de T_N on observe deux comportements différents de part et d'autre de S_{cr} . En particulier, sous une faible contrainte ($S < S_{cr}$), la variation du DL est rapide et réversible. Nous interprétons nos résultats expérimentaux par le déplacement aisé des parois de domaines (parois de macle). Ce comportement est confirmé par la faible valeur de l'énergie nécessaire à la formation d'un monodomaine comparée à l'énergie d'un magnon à $\mathbf{k} = 0$ déduite d'expériences de diffusion Raman. La valeur de cette énergie est en accord avec nos récentes études sur le dichroïsme circulaire magnétique. Pour une contrainte $S \parallel \langle 001 \rangle$ supérieure à S_{cr} , dans tous les échantillons, les spins s'alignent suivant la direction $\langle 001 \rangle$. Par contre, une contrainte parallèle à $\langle 011 \rangle$ ne donne aucun DL pour KCoF₃ et RbCoF₃. Ceci est en accord avec nos calculs, car le coefficient de magnétostriction λ_{111} est très faible. En présence d'une forte contrainte ($S > S_{cr}$), immédiatement au-dessous de T_N , le DL augmente rapidement si la température décroît. Nous concluons que le DL a une origine purement magnétique liée aux séparations énergétiques de l'état fondamental dues à l'interaction d'échange. Le rôle de la contrainte est d'aligner les spins suivant une direction déterminée.

Abstract. — We report here on the stress induced linear dichroism (LD) of KNiF₃, KCoF₃ and RbCoF₃ above and below their Néel temperature. Above T_N we obtain a linear variation of the dichroism with a $\langle 001 \rangle$ applied stress S , but below T_N we find two different parts for this curve when S increases. Under a small stress ($S < S_{cr}$) the LD increases rapidly and is reversible with S . From these experiments, we infer that there is an easy reversible movement of the domain twin-walls. This is confirmed by the small energy required to obtain a single domain in comparison to the $\mathbf{k} = 0$ magnon energy deduced from Raman experiments. The value of this energy agrees with our recent magnetic circular dichroism studies. For all samples above S_{cr} ($S \parallel \langle 001 \rangle$) we obtain an untwinned crystal where the spins align along the $\langle 001 \rangle$ direction. In agreement with our calculations, no effect is observed for KCoF₃ and RbCoF₃ under a $\langle 011 \rangle$ stress since they have a small λ_{111} magnetostrictive coefficient. Under a high stress ($S > S_{cr}$), the LD increases rapidly with decreasing temperature just below T_N . We attribute this dichroism to a pure magnetic origin which is related to the exchange splitting in the ground state, the stress being only used to align the spins along a definite direction.

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1. Introduction. — Many authors have tried to explain the origin of the birefringence or the linear dichroism (LD) in antiferromagnets [1-3] or in ferromagnets [4, 5]. However the question is still open as to how much of this birefringence has a purely magnetic origin [1, 3, 4] and how much is due to magnetostrictive deformation [2]. In an effort to help clarify this point, we reported in earlier papers [1, 6] measurements of the linear dichroism induced by an applied uniaxial stress. These measurements were made on cubic antiferromagnets (KNiF₃, KCoF₃ and RbCoF₃) in which the magnetostrictive deformations are weak. The dominant effect should be that due to the alignment of spins along the direction of the applied stress.

In the present paper, we discuss in more detail the variation of this linear dichroism (LD) with stress and temperature in these crystals. We show that the characteristic behaviour observed can be understood in terms of the movement of domain twin-walls under stress to produce a monodomain. A similar process has been previously proposed by us [7] to interpret the magnetic circular dichroism under a magnetic field in KNiF₃. The dispersion of the LD with frequency was also studied and is reported in a second paper [8].

LD is defined as the difference in absorbance for light polarized parallel and perpendicular to the stress direction respectively. The main advantage of the LD over magnetic birefringence is that the experiment is carried out in absorption regions where essentially one transition is involved, its origin being usually known. Experiments have been performed in the spectral range 13 500-35 000 cm⁻¹ on single crystals of KNiF₃ ($T_N = 246$ K) KCoF₃ ($T_N = 117$ K) and RbCoF₃ ($T_N = 101$ K) grown by the Bridgman-Stockbarger method. Crystallographic measurements on cobalt fluoperovskites show the existence of a quadratic distortion below T_N [9]; in KNiF₃ the deformation does not exceed 10⁻⁵ [9].

2. Experimental results. — The apparatus has been described previously [10]. In the experiments 0.5 × 5 × 5 mm³ samples are used and the cross-section of the light beam is about 4 mm². In this paper we refer mainly to the ³A_{2g} → ¹T_{2g} transition for

KNiF₃ and to the ⁴T_{1g} → ²T_{1g} transition for KCoF₃ and RbCoF₃ because of their reasonable absorbance for a 0.5 mm thick crystal. However the same temperature and stress dependence can be observed in all the bands located between 13 000 and 25 000 cm⁻¹.

In the region of magnetic ordering, we note :

— A sudden increase of the LD and a modification of the shape of the spectrum at T_N (Fig. 1 and ref. [1, 6]) for all samples when $S \parallel \langle 001 \rangle$. For $S \parallel \langle 011 \rangle$ such a variation was observed only for KNiF₃. For KCoF₃ and RbCoF₃ we only detect a small stray signal related to the misorientation or to stress inhomogeneities; this prevents the estimate of the true signal when $T < T_N$ for cobalt fluoperovskites.

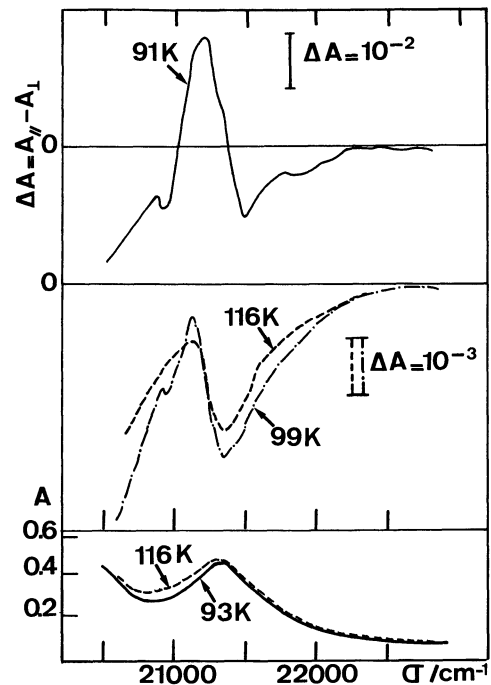


FIG. 1. — Absorption (bottom), LD around T_N for $S = 62 \text{ kgp.cm}^{-2}$ (middle) and LD below T_N for $S = 34 \text{ kgp.cm}^{-2}$ (top) ($S \parallel \langle 001 \rangle$) for RbCoF₃.

— A change of the slope of the LD amplitude versus S (Fig. 2 and ref. [1, 6]) for all samples if $S \parallel \langle 001 \rangle$ at a small value of the stress S_{cr} (Table I) whatever the light frequency is. The shape of the LD spectra does

TABLE I

Experimental values of S_{cr} and K for KNiF₃, KCoF₃ and RbCoF₃ below T_N

| | λ_{100} [9] | $S_{cr} (S \parallel \langle 001 \rangle) / \text{kgp.cm}^{-2}$ | $S_{cr} (S \parallel \langle 011 \rangle) / \text{kgp.cm}^{-2}$ | $K (\text{LD}) / \text{erg.cm}^{-2}$ | $k = 0$ magnon frequency (Raman) (2 K) | $K (\text{Raman}) / \text{erg.cm}^{-2}$ |
|--------------------|--------------------------------|---|---|--------------------------------------|---|---|
| KNiF ₃ | $< 10^{-5}$ (4 K) | 120 (242 K) 70 (150 K) | 50 (242 K or 150 K) | 10^3 (150 K) | 5 cm ⁻¹ [11] | 10^5 |
| KCoF ₃ | 1.9×10^{-3} (95 K) | 20 (95 K) | | | 38.5 cm ⁻¹ [12] | |
| RbCoF ₃ | 1.9×10^{-3} (83 K) | 20 (83 K) | | | 31 cm ⁻¹ [13] | |

not depend on the stress value. Then, at a given temperature we have estimated the LD amplitude for the ${}^4T_{1g}^a \rightarrow {}^2T_{1g}$ transition on an extremum whose frequency depends of the magnetic order (Fig. 1 and 2). We found a similar behaviour for $S \parallel \langle 001 \rangle$ for KNiF_3 . We have observed that the S_{cr} value may vary by a factor two with the sample, depending on its thickness and its origin.

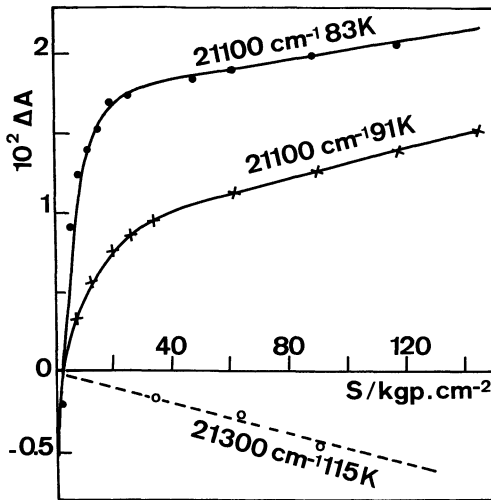


FIG. 2. — Stress dependence of LD in paramagnetic and antiferromagnetic region for RbCoF_3 ($S \parallel \langle 001 \rangle$). We give the results for the transition ${}^4T_{1g}^a \rightarrow {}^2T_{1g}$, as an example.

— The reversibility of the LD versus the stress up to 250 kgp.cm^{-2} for KNiF_3 and RbCoF_3 when we have cooled the sample under or without an applied stress. However, for KCoF_3 , after a cooling under stress followed by a rapid removal of the stress, a signal remains. A slight but rapid temperature variation destroys this metastable equilibrium and the LD then decreases to a low value.

— For $S = 0$ there remains a random residual signal due to internal strains.

— The LD varies approximately as the square of the sublattice magnetization M^2 .

A LD momentum analysis cannot be achieved because of the too large thickness of the studied sample. For this case, the LD amplitude at a given frequency does not follow exactly M^2 .

3. Discussion. — **3.1 ORIGIN OF LD UNDER STRESS** IN KNiF_3 , KCoF_3 , RbCoF_3 . — For these compounds above T_N the shape of the LD is similar to the absorption for each individual spin-orbit component [1]. This behaviour cannot be attributed to a splitting of the excited electronic levels of the Ni^{2+} ion. The LD under stress above T_N can only arise from either the splitting of the infrared t_{1u} modes of vibration which assist the transition [1] or to a mixing of the spin orbit excited states via the stress.

If we assume that the photoelastic contribution to the LD below T_N is of the same order of magnitude as in the paramagnetic region, we do not explain the

large amplitude change of the LD at T_N . Moreover this LD cannot be related to the magnetostriction in KNiF_3 because the corresponding deformation ($< 10^{-5}$) [9] remains small compared to the stress induced deformation (5×10^{-4} at 200 kgp.cm^2). We therefore conclude that the large LD ($\Delta A/A \sim 10^{-1}$) below T_N has a *pure magnetic origin*, i.e., is closely related to the exchange splitting in the ground state. The stress only orients the spins along a favoured direction, which may be compared to the effect of a magnetic field.

Below T_N , for KCoF_3 and RbCoF_3 [6] the conclusion about the origin of the LD is the same as in KNiF_3 except perhaps for the ${}^4T_{1g}^a \rightarrow {}^2T_{1g}$ transition (Fig. 1) where the S-shaped LD curve indicates that the exchange interaction in the excited state can be significant.

A comparison of the sign of the LD under stress and under a magnetic field in KNiF_3 and RbNiF_3 [14] for the ${}^3A_{2g} \rightarrow {}^1E_g$ transition, leads us to the conclusion that, below T_N , the spins in KNiF_3 align along the stress direction ($S \parallel \langle 001 \rangle$). This is in agreement with torque measurements [15]. Our results suggest that the spins also align along the stress direction for $S \parallel \langle 011 \rangle$. For KCoF_3 a $\langle 001 \rangle$ stress aligns the spins along its direction [15]. By LD under stress, we have verified that the same behaviour occurs in RbCoF_3 .

We now consider the temperature dependence of the LD. Below T_N the amplitude of the LD versus temperature varies approximately as M^2 . This will be demonstrated in section 2 of our paper [8]. Note that a similar variation has been observed recently in birefringence for the cubic antiferromagnet NiO [16]. From T_N up to higher temperatures, as for KNiF_3 [1], the magnetic part of the LD for KCoF_3 and RbCoF_3 reaches zero very rapidly within about two degrees. By comparison with heat capacity [9, 17] and neutron diffraction measurements on RbCoF_3 [18] where short range ordering is still efficient ten degrees above T_N , we conclude that the LD does not follow the spin pair correlation function. This agrees with the fact that the variation of the LD is related to the vanishing of domains.

3.2 DESCRIPTION OF THE SPIN-FLOPPING. — For a crystal containing a single magnetic domain, we study now the spin behaviour under stress by minimising the sum of the anisotropy, elastic and magneto-elastic energies. Since a stress preserves the antiparallel alignment of the spins we consider the general expression given for ferromagnets [19] where we take into account only the terms of the fourth power in the cosines of the angle of magnetization. The calculated critical values S_{cr} necessary to align the spins along the stress direction are summarised in table II using the λ_{100} and λ_{111} magnetostriction coefficients and the anisotropy constant K . If K is negative we should find two successive S_{cr} values (α) and (β), by increasing the

TABLE II

S_{cr} values for $\langle 001 \rangle$ or $\langle 011 \rangle$ stress directions. If $S = 0$, for the (α) case the spins lie along the $\langle 111 \rangle$ or $\langle \bar{1}\bar{1}\bar{1} \rangle$ axis; in the (β) case the spins directions are $\langle \bar{1}11 \rangle$ or $\langle 1\bar{1}\bar{1} \rangle$.

| $S \parallel$ | K | |
|-----------------------|-----------------------------|---|
| | > 0 | < 0 |
| $\langle 001 \rangle$ | $\frac{2K}{3\lambda_{100}}$ | $\frac{-2K}{3\lambda_{100}}$ |
| $\langle 011 \rangle$ | $\frac{K}{3\lambda_{111}}$ | (α) $\frac{-2K}{\lambda_{111}}$ (β) $\frac{-K}{\lambda_{100} + 3\lambda_{111}}$ |

stress along $\langle 011 \rangle$. Since K must be positive for the considered fluoperovskites we expect a rapid variation of the LD under a $\langle 001 \rangle$ stress at S_{cr} because of the spin-flopping.

Our experimental results (Fig. 2) show a continuous dependence of the LD with stress and suggest that we have a polydomain structure in cubic antiferromagnets in the absence of a stress, as proposed earlier by Néel [20]. The origin of the spin-flopping in a cubic antiferromagnet differs markedly from that obtained in a uniaxial one because it is due to a reversible movement of the domain twin-walls. As in NiO [21] we conclude that we have an untwinned crystal for $S > S_{cr}$; the lower part of the curve ($S < S_{cr}$) (Fig. 2) is related to the displacement of the twin-walls. Other features allow the retention of such a mechanism instead of the rotation of the spins in a single domain :

— The K value deduced from our experiments and calculated from S_{cr} do not agree with the results deduced from Raman scattering experiments for KNiF_3 (Table I). For KCoF_3 and RbCoF_3 the small value of S_{cr} is not consistent with the rather large $\mathbf{k} = 0$ magnon frequencies (Table I).

— We found a dispersion of the S_{cr} values for different samples.

If we take into account the interaction with an applied stress S (direction cosines $\gamma_1, \gamma_2, \gamma_3$), the free energy per unit volume of the crystal has been defined as [19] :

$$F = F_S + F_A = -\frac{3}{2}\lambda_{100}S(\alpha_1^2\gamma_1^2 + \alpha_2^2\gamma_2^2 + \alpha_3^2\gamma_3^2) - \frac{3}{2}\lambda_{111}S(2\alpha_1\alpha_2\gamma_1\gamma_2 + 2\alpha_2\alpha_3\gamma_2\gamma_3 + 2\alpha_3\alpha_1\gamma_3\gamma_1) + K(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2)$$

where $\alpha_1, \alpha_2, \alpha_3$ stand for the direction cosines of the magnetization.

F_S comes from the stress effect and F_A contains both the magnetoelastic and elastic energy. The expression of the anisotropy coefficient K is given in [19] and the λ 's are the magnetostrictive coefficients.

Without stress the easy magnetization directions are related to the sign of K . In our case, K is positive and the spins are aligned along the x, y and z four-fold axes of the crystal giving rise three kinds of domains [15] labelled d_x, d_y and d_z .

Under a $\langle 001 \rangle$ stress $\alpha_3 = \cos \theta$ and F_S can be expressed by

$$F_S = -\frac{3}{2}\lambda_{100}S\cos^2\theta.$$

If we consider the case of two kinds of domains d_z and d_x (or d_y) the spin movement occurs in the zx (or zy) plane and :

$$F_A = \frac{K}{4}\sin^2 2\theta.$$

In order to interpret our results we can follow Hirakawa *et al.* [15] who introduce a restoring force proportional to the displacement x of the wall which separates a d_z from a d_x (or a d_y) domain. The variation ΔF of the total free energy per unit surface is expressed by :

$$\Delta F = \frac{1}{2}kx^2 - \frac{3}{2}\lambda_{100}S[(x)\cos^2\theta_1 - (-x)\cos^2\theta_2] + \frac{K}{4}[(x)\sin^2 2\theta_1 - (-x)\sin^2 2\theta_2]$$

where θ_1 and θ_2 represent the spin direction inside d_z and d_x (or d_y) domains.

The minimisation of ΔF with respect to θ_1, θ_2 and x leads to $\theta_1, \theta_2 = 0 + n\frac{\pi}{2}$ and $x = \frac{3}{2k}S\lambda_{100}$; the size

of the d_z domains therefore increases at the expense of the d_x or d_y domains and the wall moves linearly with the stress. There only remains d_z domains when x reaches the value l of their width at a stress $S_{cr} = 2kl/3\lambda_{100}$. These facts are in good agreement with our experimental results (Fig. 2) in particular with the linear variation of the LD versus S for $S < S_{cr}$. We have also demonstrated that for $S \parallel \langle 011 \rangle$, S_{cr} varies as kl/λ_{111} . Thus, the results are formally the same as those reported in table II (for a single domain) if we replace K by kl , but it is important to note that the mechanism involved differs markedly. As we have mentioned earlier the twin-wall movement gives a continuous effect under an applied stress, contrary to the classical *spin flop* inside a single domain.

Since S_{cr} values for KNiF_3 are comparable for $S \parallel \langle 001 \rangle$ or $S \parallel \langle 011 \rangle$ we deduce that λ_{100} and λ_{111} have the same order of magnitude. On the contrary, the absence of the LD in KCoF_3 and RbCoF_3 for $S \parallel \langle 011 \rangle$ is consistent with the quadratic distortion of the crystal below T ($\lambda_{100} \gg \lambda_{111} \simeq 0$).

In contrast to NiO [21] we had difficulties maintaining untwinned KCoF_3 samples after removing the stress. Since the magnetostrictive deformation value is

similar and the dimension of the domains is large (about 1 mm) [22] for the two crystals, we conclude that the reversibility of the spin-flopping in KCoF_3 might arise from the high purity of the samples.

3.3 COMPARISON BETWEEN TWIN-WALLS MOVEMENT UNDER A STRESS AND UNDER A MAGNETIC FIELD. — We have recently demonstrated [7] by the use of magnetic circular dichroism that the magnetic field necessary to flop the spin from the field direction to the plane perpendicular to it is less than 1 tesla. Note that the critical field H_{cr} is very small compared to $\sqrt{2 H_E H_A}$ where H_A is the anisotropy field and H_E the exchange field. Assuming the same origin, i.e., the movement of domain twin-walls, for the spin-flopping under a stress or a magnetic field, we obtain the following relation between H_{cr} and S_{cr} :

$$\Delta F_{\text{mag}} = -\frac{1}{8k}(\chi_{\perp} - \chi_{\parallel})^2 H_{\text{cr}}^4 = \\ = \Delta F_{\text{stress}} = -\frac{9}{8k} \lambda_{100}^2 S_{\text{cr}}^2$$

if ΔF_{mag} (or ΔF_{stress}) represents the variation of the free energy in the presence of a magnetic field (or of a stress). Thus, $H_{\text{cr}} = \sqrt{3 \lambda_{100} S_{\text{cr}} / (\chi_{\perp} - \chi_{\parallel})}$ and since $\lambda_{100} \leq 10^{-5}$ in KNiF_3 we deduce $H_{\text{cr}} \leq 1$ Tesla.

We therefore conclude that the two sets of experiments are consistent, especially if we take into account the fact that they were not performed on the same crystal and that the critical field (and stress) depends upon the sample.

3.4 TENTATIVE EXPLANATION FOR THE ABSENCE OF SATURATION OF LD FOR $S > S_{\text{cr}}$. — The slope of the LD curve versus S ($S > S_{\text{cr}}$) may be related to different mechanisms: the photoelastic effect, the modification of the exchange integral J under stress or the incomplete alignment of the spins along the stress direction at S_{cr} . The large difference between the values of the slope above and below T_N indicates that the LD depends not only on the photoelastic effect but on other effects also. In KNiF_3 the deduced magnetic

part of the LD for $S > S_{\text{cr}}$ shows a smaller slope for $S \parallel \langle 011 \rangle$ than for $\langle 001 \rangle$.

This could be due to a larger modification of J for $S \parallel \langle 001 \rangle$ which is the direction of the superexchange than for $S \parallel \langle 011 \rangle$. The KNiF_3 [23] or RbCoF_3 and KCoF_3 [9] values of $\partial J / \partial a$ lead to a negligible effect at low temperature on single ion transitions compared to those considered here. The last explanation is that at $S = S_{\text{cr}}$ we have not observed the entire saturation of the LD due to the spin rotation. This would confirm the existence of S-type domains [21] in these perovskites as observed by Lang's method in KCoF_3 [22], a fact consistent with the large attenuation of ultrasound below T_N for a stressed crystal, for $S > S_{\text{cr}}$.

4. Conclusion. — We have demonstrated that the stress induced linear dichroism in KNiF_3 , KCoF_3 and RbCoF_3 has a magnetic origin. The stress S is only necessary to remove the domain structure so as to give an untwinned crystal. The corresponding value of S_{cr} is small compared to the stress which is necessary to rotate the spins in a single domain in an uniaxial antiferromagnet. This is a general effect in cubic antiferromagnets. In particular, we have observed the same behaviour for RbMnF_3 where

$$S_{\text{cr}} (S \parallel \langle 111 \rangle) \simeq 20 \text{ kgp.cm}^{-2}$$

is small compared to the value of 400 kgp.cm^{-2} previously attributed to the spin-flop phenomenon [24]. Thus LD is an interesting and sensitive method to follow twin-wall movements.

The dispersion of the LD versus the light frequency contains also spectroscopic information which has not been discussed here. This will be considered in the second part of our paper [8] in the light of a comparison of the LD under stress on KNiF_3 and KCoF_3 to the magnetic linear dichroism of $\text{Ni}^{2+}/\text{KMgF}_3$ and $\text{Co}^{2+}/\text{KMgF}_3$ diluted compounds.

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