Piezooptic effect in MnF_2

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A study was made of the change in the birefringence of a tetragonal MnF_2 crystal due to uniaxial compression at three temperatures: 300, 66, and 2°K. The magnitude of the piezooptic effect was determined for three experimental geometries at each of these temperatures. It was found that: a) the piezooptic effect was independent of temperature: b) for a given strain, the change in the birefringence had opposite signs for thermal and elastic compression.

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1. INTRODUCTION

Magnetic birefringence of light was discovered by Dillon[1] in an investigation of the domain structure of garnets and by Roth[2] in a study of antiferromagnetic NiO. Systematic investigations of this effect started in about 1970, but its origin has not yet been conclusively identified. Among the many possible mechanisms the most natural would seem to be the piezooptic effect due to the spontaneous deformation of the lattice (magnetostriction).[3-5]

In several investigations a comparison was made of the change in the birefringence occurring because of spontaneous deformation with the change produced by a similar deformation caused by an external uniaxial pressure. Experiments of this kind have been carried out so far only on cubic iron garnets.[6-8] It was found that spontaneous striction should give rise to an effect two orders (or in rare cases, one order) less than the observed magnetic birefringence.

The spontaneous deformation of the lattices of transition metal fluorides caused by antiferromagnetic ordering is approximately two orders of magnitude stronger than the magnetostriction of garnets. In the case of fluorides we can expect a much greater contribution of the striction to the birefringence. Jahn[4] found that at sufficiently high temperatures ($T > 3T_N$) the change in the birefringence is proportional to the temperature-induced change in the "degree of tetragonality":

$$d(n_e - n_o) = M\left(\frac{da}{a} - \frac{dc}{c}\right)$$

or, which is equivalent, to the difference between the thermal expansion coefficients along inequivalent axes[5]:

$$\frac{d}{dT}(n_e - n_o) = K(\beta_c - \beta_a).$$

However, the coefficient M (or K) changes on approach to the magnetic ordering temperature T_N. In the case of MnF_2, its sign is reversed below T_N and its value is approximately six times greater than at $T > T_N$. This means that if the striction mechanism is correct, the same strain should alter very differently (in respect of the magnitude and sign) the birefringence in the paramagnetic and antiferromagnetic regions. Therefore, it is stated in Ref. 9 that the striction mechanism of the magnetic birefringence can play no significant role in uniaxial MnF_2-type crystals. However, according to Jahn,[5] the birefringence is influenced strongly by the fluorine parameter and a very small change in this parameter may alter greatly the optical properties in the antiferromagnetic region because fluorine ions participate in the exchange interaction.

We investigated the piezooptic effect in MnF_2 with the aim of determining how a change in the "tetragonality parameter" caused by different types of deformation in a crystal affects the birefringence.

2. APPARATUS AND SAMPLES

Our measurements were carried out on single crystals of MnF_2. These crystals were transparent, rose-colored, and free of cracks.[2)] We selected regions free of blocks so as to avoid, particularly in the central part of a sample of $2\times2\times5$ mm dimensions, any inhomogeneities or bubbles. In two samples the long edges were directed along the [001] axis (samples Nos. 1 and 2) and the short edges along the [110] axis; in the case of the third sample (No. 3), the long edge was directed along [110] and the short edges along [110] and [001]. The samples were oriented by the x-ray method. The faces of the samples did not deviate by more than 1° from the crystallographic planes. The end faces of the samples were oriented particularly carefully and special measures were taken during grinding to ensure that these faces were exactly parallel. The grinding of all the faces was followed by polishing with a diamond paste on silk. The samples were not annealed after preparation because the layers deformed during the preparatory treatments were very thin compared with the thickness of the sample and they did not distort significantly the measured quantities.

The measurements were made at room and lower temperatures. The optical measurements in a helium bath were carried out most conveniently below the λ point when boiling of the liquid was avoided. To ensure approximately the same conditions also at other temperatures, all the measurements were carried out in an evacuated cryostat. This arrangement was also convenient because only one (external) pair of windows was under pressure. The difference between the pressures on the internal pair of windows was slight (a few torr).

A mechanical force could easily be created in the

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evacuated cryostat by altering the gas pressure in a bellows. This was the principle adopted in the apparatus shown schematically in Fig. 1. This apparatus consisted of thin-walled stainless-steel tubes. A sample 1 was placed in a thick-walled copper tube 2 between two copper pistons, the lower of which was attached rigidly to the lower end of the copper tube and the other to a thin-walled stainless-steel tube which pressed against the lower end of the bellows. The latter tube transmitted the mechanical force to the sample. This tube was centered by a membrane 3 at the top end and by a tightly fitting Teflon guide ring 4 at the lower end. Two coils of thin copper wire were wound on the copper tube and were used to heat the tube and to monitor the constancy of its temperature.

The force acting on the sample was calculated from the measured pressure in the bellows and the area of the bottom of the bellows. However, since the whole apparatus consisted of thin tubes, it was necessary to allow for their deformation in the stress state and to find the correction for the force needed to stretch the bellows appropriately. The deformation of the tubes was calculated; the force needed to stretch the bellows was measured. The apparatus was calibrated by measuring the piezooptic effect in quartz glass (fused silica). A good agreement was obtained between our measurements and the values given in the AIP Handbook.[10]

The influence of a slight surface roughness of the pistons and of the end faces of the sample, which could create inhomogeneous stresses and fracture the sample, was minimized by placing thin indium sheets between the sample and the copper faces in contact with it. The most serious source of error was the irreproducibility of the mounting of the sample.

The error in the birefringence measurements did not exceed 10% of the maximum value. The scatter of the results obtained by repeated measurements was up to ±3%, but when temperature was altered it rose to ±5% and after remounting of the sample it rose to ±10%.

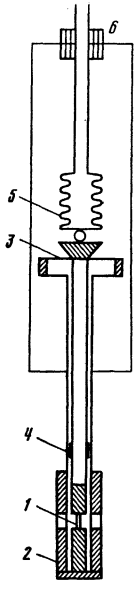

FIG. 1. Principal parts of the apparatus used to apply pressures: 1) sample between copper pistons; 2) thick-walled copper tube with windows; 3) membrane; 4) Teflon ring; 5) bellows; 6) guide for vertical motion of bellows.

The change in the birefringence was measured with a Berek compensator. This fairly simple instrument made it possible to carry out measurements with a very good precision if restriction was made to small angles of rotation of a calcite plate,[11] because its birefringence varied quadratically with the angle of rotation. Between 1 and 4° the compensator sensitivity varied from 0.1 to 0.4 nm per angular minute of rotation of the plate (this corresponded to a phase shift of the transmitted light by 3.5′–14′). For a sample 2 mm thick it was possible to measure the change in the birefringence with an error not exceeding 2×10^{-7}.

One of the sources of the experimental error was nonparallelism of the faces of the sample. In the dc measurements this had the effect of smearing the signal minimum. Therefore, modulation of light was used to ensure sufficiently accurate compensation of the signal. This was done using the apparatus shown in Fig. 2. Light from a helium–neon laser 1 passed through a polarizer 2, an electrooptic modulator 3 (of the ML-3 type), a sample 4, and a quartz wedge 5 which was used for rough compensation ensuring a shift of the working angle of a Berek compensator to the 0–4° range; light then passed through the Berek compensator 6 mounted on a stage, which could be used to measure angles to within 1′, an analyzer 7, and a photomultiplier 8. The direction of pressure was inclined at 45° to the crossed polarizer and analyzer. The electrooptic modulator was subjected to an alternating voltage from an oscillator 9 and this varied periodically the total birefringence so that the photomultiplier signal was modulated. This signal was amplified with a narrow-band measuring amplifier 10 tuned to the oscillator frequency.

Since the birefringence of the sample and modulator was temperature-dependent and the ambient temperature in the laboratory was not specially stabilized, only those results were used which were obtained under constant temperature conditions. This could be determined quite easily by measuring the piezooptic effect by increasing the pressure and then reducing it. The two straight lines obtained in these cases should coincide. Moreover, use was made only of those measurements in which the dependence $\delta\Delta n(P)$ was linear. Deviation of $\delta\Delta n(P)$ from linearity could be due to a number of factors, usually temperature variations or inaccurate setting of the sample. Figure 3 shows examples of "good" measurements.

The straight lines obtained in this way were used to calculate the piezooptic effect for a given experimental

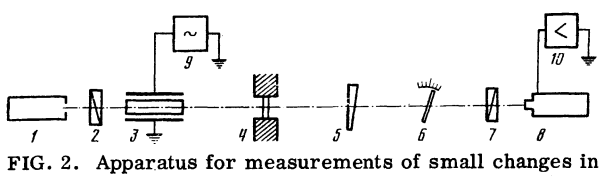

FIG. 2. Apparatus for measurements of small changes in the birefringence: 1) helium—neon laser; 2) polarizer; 3) light modulator; 4) sample; 5) quartz wedge; 6) Berek compensator; 7) analyzer; 8) photomultiplier; 9) oscillator; 10) measuring narrow-band amplifier.

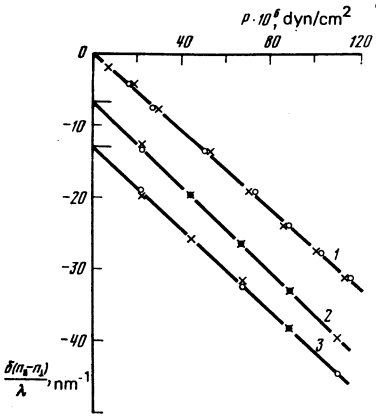

FIG. 3. Typical results of measurements of the piezooptic effect in an MnF_2 single crystal (sample No. 3, pressure along [110], light along [110]): 1) at helium temperatures (approximately 2 °K); 2) at nitrogen temperatures (approximately 55 °K); 3) at room temperature (approximately 295 °K). The circles were obtained by increasing the pressure and the crosses by reducing it.

geometry and a given temperature:

$$D=\delta\Delta n(P)/\delta P. \tag{1}$$

In all the experiments the direction of light k was perpendicular to the applied pressure P and measurements were made of the quantity $\Delta n = n_{\parallel} - n_{\perp}$, where $n_{\parallel}$ is the refractive index of light polarized parallel to P and $n_{\perp}$ is the refractive index of light polarized perpendicular to P.

3. RESULTS OF MEASUREMENTS

The piezooptic effect was investigated in three experimental geometries shown schematically in Fig. 4. Table I gives the calculated (with the aid of the elastic moduli taken from Ref. 12) strains of the samples for two directions of P: I) P‖[001]; II), III) P‖[110]. This table gives values calculated for the maximum pressure $P=-1.2\times10^8$ dyn/cm² used in the present study. The room-temperature values of the piezooptic effect D are given for all three geometries, as well as the maximum values of $\delta(n_{\parallel}-n_{\perp})$ obtained for $P=-1.20\times10^8$ dyn/cm². For comparison, the last row of Table I lists the values obtained on thermal compression of MnF_2.

Data on the temperature dependence of the piezooptic effect are given in Table II. It is clear from this table that the piezooptic effect is practically independent of temperature and also of the state (paramagnetic or antiferromagnetic) of MnF_2.

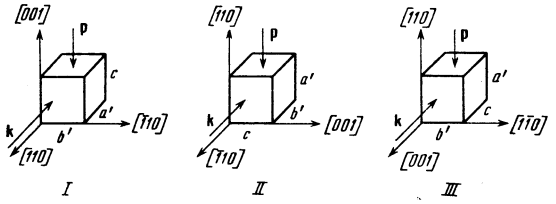

FIG. 4. Experimental geometries used to measure the piezooptic effect.

TABLE I. Strains created by pressure and thermal expansion (lower row) and corresponding changes in birefringence.

Experimental geometry (Fig. 4)	Sample No.	Strain (in units of 10^{-5}) at $P=-1.2\times10^8$ dyn/cm²						$\delta(n_{\parallel}-n_{\perp})\cdot10^5$	$D\cdot10^{13}$ cm²/dyn
		$\frac{\delta c}{c}=u_{zz}$	$u_{xx}=u_{yy}$	u_{xy}	$\frac{\delta a'}{a'}$	$\frac{\delta b'}{b'}$	$\frac{\delta a'}{a'}-\frac{\delta c'}{c'}$		
I	1	−10.8	4.2	0	4.2	4.2	15.0	−1.73 **	1.44
	2							−1.74 **	1.45
II	3	4.2	−4.9	−8.3	−9.1	−0.8	−13.3	−1.48 ***	1.23
III	3	4.2	−4.9	−8.3	−9.1	−0.8	−13.3	−1.44	1.20
$\Delta T=-10$°K at $T=300$ K		−12.1	−2.9	0	−2.9	−2.9	9.2	+1.66 **	

*The x, y, and z axes directed along the unit-cell edges; the a' and b' axes along [110] and [1$\bar{1}$0].
**In this experiment, $\delta(n_{\parallel}-n_{\perp})=\delta(n_e-n_0)$.
***In this experiment, $\delta(n_{\parallel}-n_{\perp})=-\delta(n_e-n_0)$.

4. DISCUSSION OF RESULTS

1. It is known (see, for example, Ref. 13) that the piezooptic effect in tetragonal crystals is described by seven piezooptic coefficients π_{ik} which form the following tensor:

$$\begin{pmatrix} \pi_{11} & \pi_{12} & \pi_{13} & 0 & 0 & 0 \\ \pi_{12} & \pi_{11} & \pi_{13} & 0 & 0 & 0 \\ \pi_{31} & \pi_{31} & \pi_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & \pi_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & \pi_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & \pi_{66} \end{pmatrix}. \tag{2}$$

The piezooptic coefficients relate in the following way the change in the reciprocal permittivity tensor δB_i to the stresses:

$$\delta B_i=\pi_{ij}\sigma_j, \tag{3}$$

where σ_j is the elastic stress tensor ($i=1-6, j=1-6$).

Since

$$\delta n_i=-{}^1/_2 n_i^3\Delta B_i \quad (i=1, 2, 3),$$

we can show that the three measured quantities $D_{\rm I}$, $D_{\rm II}$, and $D_{\rm III}$ are related to the piezooptic coefficients:

$$D_{\rm I}={}^1/_2[n_o^3\pi_{13}-n_e^3\pi_{33}], \tag{4}$$

$$D_{\rm II}={}^1/_4[n_o^3(\pi_{11}+\pi_{12}+{}^1/_2\pi_{66})-2\pi_{13}n_e^3], \tag{5}$$

$$D_{\rm III}={}^1/_4 n_o^3\pi_{66}, \tag{6}$$

where $n_o=1.472$ and $n_e=1.499$ are the refractive indices for the ordinary and extraordinary rays, respectively.

Thus, our experiments give unambiguously only the value of π_{66}:

$$\pi_{66}=1.51\cdot10^{-13}\ \text{cm}^2/\text{dyn}. \tag{7}$$

In the case of the other three piezooptic coefficients, all that we can do is to give the following relationships:

$$\pi_{13}-1.06\pi_{33}=0.90\cdot10^{-13}\ \text{cm}^2/\text{dyn}, \tag{8}$$

TABLE II. Temperature dependence of piezooptic effect.

Experimental geometry (Fig. 4)	Sample	$D\cdot10^{13}$, cm²/dyn		
		2 K	55 K	300 K
I	1	1.44	1.48	1.44
I	2	1.42	1.43	1.45
II	3	1.23	1.29	1.23
III	3	1.13	1.15	1.20

$$\pi_{11}+\pi_{12}-2.11\pi_{13}=0.48\cdot10^{-13}\,\text{cm}^2/\text{dyn}. \quad (9)$$

2. A comparison of the change in the birefringence with the change in the degree of tetragonality in two experiments (in geometries I and II) and with the change as a result of thermal expansion shows (Table I) that one quantity (degree of tetragonality) is insufficient for the description of the change in the birefringence due to different types of deformation. This conclusion follows from an analysis of the relationship between the change in the reciprocal permittivity tensor and the strain u_k. This relationship can be expressed in terms of the tensor of the elastooptic coefficients p_{ik} (Ref. 13):

$$\Delta B_i=p_{ik}u_k, \quad p_{ik}=\pi_{ir}c_{rk}. \quad (10)$$

The form of the tensor p_{ik} is identical with the form of the tensor π_{ik} given by Eq. (2).

Using Eq. (10), we easily obtain an expression for the relationship between the change in the birefringence $\delta(n_e-n_o)$ and the strain. In the simplest case of uniaxial compression along a fourfold axis or thermal expansion, we have

$$u_{xy}=u_{xz}=u_{yz}=0, \quad u_{xx}=u_{yy}=u_1, \quad u_{zz}=u_3$$

and the corresponding relationship is

$$\delta(n_e-n_o)=\tfrac{1}{2}n^3[(p_{11}+p_{12}-2p_{31})u_1+(p_{13}-p_{33})u_3]. \quad (11)$$

It is assumed in the above relationship that $n_e\approx n_o=n$.

Denoting the coefficients in Eq. (11) by A and B,

$$A=\tfrac{1}{2}n^3(p_{11}+p_{12}-2p_{31}), \quad B=\tfrac{1}{2}n^3(p_{13}-p_{33}), \quad (12)$$

we obtain the following relationship for the experiments in geometry II:

$$\delta(n_{[110]}-n_{[001]})-\frac{n_o^3}{2}\frac{p_{66}}{2}u_6=-Au_1-Bu_3. \quad (13)$$

Substituting the experimental values of δn, u_1, u_3, and u_6 into Eqs. (11) and (13) (and bearing in mind that $p_{66}=\pi_{66}c_{66}=0.11$), we obtain a system of two equations for the determination of the coefficients A and B. In this way we obtain the change in the birefringence in the presence of the simplest type of deformation:

$$\left.\begin{aligned}&u_{xx}=u_{yy}\neq u_{zz}, \quad u_{xy}=u_{xz}=u_{yz}=0;\\&\delta(n_e-n_o)=-0.01u_{xx}+0.16u_{zz}.\end{aligned}\right\} \quad (14)$$

We can thus see that the change in the birefringence is in no way proportional to the change in the degree of tetragonality $(u_{xx}-u_{zz})$. According to Eq. (14), the decisive factor is the strain along the c axis of a tetragonal crystal. An increase in the strain (when the unit cell approaches the cubic form) increases the algebraic value of the birefringence n_e-n_o.

3. The fact that the piezooptic effect is practically independent of temperature (including the range below T_N) in every configuration used allows us to draw the conclusion that all the piezooptic constants vary slowly with temperature and do not change significantly even on transition to a new antiferromagnetic state. Therefore, we can assume that Eq. (14) is valid in the paramagnetic and antiferromagnetic states.

4. We shall now turn to the main problem formulated at the beginning of this paper, which is the relationship between the magnetic birefringence, which appears in MnF_2 on transition to the antiferromagnetic state, and the spontaneous striction. According to the results of Gibbons,[14] the spontaneous striction along the c axis is very large and its magnitude can be estimated to be $u_{zz}=-7\times10^{-4}$. Thermal expansion along the u_{xx} axis is very irregular. The temperature dependence of this expansion changes its sign and it is largely governed by the establishment of the short-range antiferromagnetic order.

The striction along the a axis can be estimated only very approximately and the result is $u_{xx}=+4\times10^{-4}$. Hence, it follows that the change in the birefringence caused by the magnetostriction is

$$\delta(n_e-n_o)_{\text{strict}}\leqslant-1\cdot10^{-4}.$$

This change has the reverse sign and a value approximately 20 times smaller than that found experimentally

$$\delta(n_e-n_o)_{\text{magn}}=1.8\cdot10^{-3}.$$

It follows that as in the case of ferrites, the magnetic birefringence observed in tetragonal antiferromagnets with high values of the spontaneous magnetostriction is considerably greater than that which can be expected from the magnetostriction alone.

5. Experiments carried out in configurations II and III allow us to draw two additional conclusions:

a) Application of a pressure along the [110] axis results in a shear deformation of a crystal and this gives rise to a considerable difference between the parameters describing the position of the fluorine ions on the diagonal a' of the unit-cell base (this diagonal is parallel to the pressure) and on the perpendicular diagonal b'. Experiments indicate that such deformation is not accompanied by any anomalous change in the birefringence. Therefore, there is no support for Jahn's hypothesis[5] that the fluorine parameter has a significant influence on the birefringence.

b) The characteristic features of the tensor of the elastic constants have the effect (Table I) that compression of a crystal along the [110] axis has practically no influence on its dimension along the $[1\bar{1}0]$ axis, i.e., along this direction the Poisson ratio is zero. We may assume that the change in the corresponding refractive index $n_{[1\bar{1}0]}$ is also smaller than the other changes. We shall check this hypothesis by writing down the results of the experiments in configurations II and III. According to Table I, we have

$$\delta(n_{[110]}-n_{[1\bar{1}0]})=-1.44\cdot10^{-5},$$
$$\delta(n_{[110]}-n_{[001]})=-1.48\cdot10^{-5}.$$

Subtracting one expression from the other, we find that

$$\delta(n_{[001]}-n_{[1\bar{1}0]})=0.04\cdot10^{-5},$$

i.e., this value is an order of magnitude less than all the other measured quantities.

6. We can easily see that Eq. (14) is unsuitable for the calculation of the temperature dependence of the birefringence. It is found experimentally that $n_e - n_o$ decreases on increase in temperature. If the only reason for this were the thermal expansion, then—in accordance with Eq. (14)—the birefringence would have increased on increase in the lattice constants, because the coefficient of expansion along the z axis is an order of magnitude greater than the coefficient of expansion along the x axis. Thus, the thermooptic effect not associated with the change in the lattice parameters should predominate.

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Scattering of light by parametric magnons and phonons in $CoCO_3$

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Mandel'shtam-Brillouin light scattering is used in antiferromagnetic $CoCO_3$ at $T \lesssim 2$ K to detect and investigate magnons parametrically excited by microwave pumping, and propagating along the C_3 axis and in the basal plane of the crystal. The excitation thresholds for parametric magnons are determined, and the maximum number of parametric particles observed in the experiment is estimated. Disintegration of a single uniform-precession ($k = 0$) magnon into two phonons, with $k \neq 0$ and with a frequency equal to half the antiferromagnetic-resonance frequency, is also observed.

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I. INTRODUCTION

The study of parametrically excited spin waves PSW (or parametric magnons PM) in magnetically ordered materials has been the object of much research (see, for example, Refs. 1 and 2). Most of these investigations have been made by radio-frequency methods, involving absorption by the material of microwave power incident on it.

A comparatively small part of the work has been devoted to optical methods of investigating PM. In these papers, the magnetooptical Faraday effects[3-5] and magnetic birefringence[6] have been used to record the change of the component of the magnetization of the crystal parallel to the external magnetic field when PM were excited.

All these experiments give information about the relaxation times of PM, their total number, etc. But they do not permit a direct answer to the question: in what direction do the parametric excitations propagate, and to what wave vector **k** do they correspond? It is also difficult to determine, by the methods indicated above, which kind of quasiparticles, phonons or magnons, are excited parametrically in the experiment. Some of these questions, it seems, can be answered by investigation of inelastic scattering of light by elementary excitations. Since parametric excitation produces quasiparticles of the low-frequency branch of the spectrum of

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