Isotropic and anisotropic magnetic refraction of light in the antiferromagnets $KNiF_3$ and $RbMnF_3$

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The thermo-optical and piezo-optical effects have been investigated in the cubic antiferromagnets $KNiF_3$ and $RbMnF_3$ and the isomorphic diamagnet $KMgF_3$. It is shown that, to high accuracy, the nonmagnetic contributions to the variation of the refractive index with temperature are proportional to the linear expansion of the lattice. The isotropic and anisotropic contributions to the change in the refraction of light have been separated out. At low temperatures the isotropic contribution in $KNiF_3$ may reach $\delta n_{is} = 3.7\times 10^{-3}$, which exceeds the anisotropic contribution by more than 2 orders of magnitude. It is asserted that because of the high symmetry of the ionic positions in the perovskite structure, the so-called "internal" ion displacements in the cell are not possible, and the magnetostrictive contribution can be calculated on the basis of data on bulk magnetostriction. It is shown that the magneto-optical mechanism makes the main contribution to the temperature dependence of the refraction. The critical exponents are found from the temperature dependences of the magnetic contribution to the birefringence and refraction. The principal difference between isotropic and anisotropic refraction of light is pointed out.

1. INTRODUCTION

Optical studies recently carried out on crystalline magnetic materials (see reviews[1–4]) demonstrated the existence in them of great changes in birefringence and in the absolute refractive index when short and long-range magnetic orderings are established. These phenomena can be divided into two groups, namely into anisotropic and isotropic magnetic refraction (or birefringence) of light. Anisotropic birefringence Δn_{aniso}, more often called magnetic linear birefringence (LMB), is determined by anisotropic spin correlation functions,[4] and its magnitude depends on the orientation of the magnetic moments relative to the crystallographic axes, for example

$$\Delta n^{zy}_{aniso} \sim \sum_{i,j} (\rho_{11}-\rho_{12})(\langle S_i^z S_j^z - S_i^y S_j^y\rangle), \quad (1)$$

where ρ_{11} and ρ_{12} are magnetic coefficients depending on the spin-orbit coupling and the anisotropic exchange interaction. In essence this effect is analogous to the well-known Cotton-Mouton effect in paramagnetics, but thanks to the large spontaneous magnetization in magnetic materials, it reaches larger values: $\Delta n \sim 10^{-4}$–10^{-5} (Refs. 1–4).

Isotropic magnetic refraction (IMR) δn_{iso} is radically different from LMB. Its magnitude is independent of the orientation of the magnetic moments relative to the crystallographic axes and is determined by the isotropic spin correlation function

$$\delta n_{iso} \sim \sum_{i,j} P_{ij}\langle S_i S_j\rangle, \quad (2)$$

where P_{ij} is the polarization component dependent on the exchange interaction between neighboring ions.[4] In the crystals studied so far[1–4] IMR reaches values $\delta n_{iso} \sim 10^{-2}$–$10^{-4}$ i.e. it is several orders of magnitude larger than the anisotropic birefringence.

A serious and interesting problem has arisen in the process of studying isotropic and anisotropic refraction, which has so far not been solved. This concerns the separating out of the different contributions which make up the changes in refraction under the action strictly of temperature (thermo-optic contribution), of deformation (elasto-optical contribution) and of magnetic ordering (magneto-optical and magnetostrictive contributions). In addition, a mechanism was proposed for a change in refraction due to internal shifts of atoms in the elementary cell, unconnected with a change in the cell itself.[5] For this investigation we chose crystals with the fairly simple perovskite cubic structure since, in our view, a separation of the different mechanisms for a change in refraction should be simpler and more reliable in them than in noncubic crystals with a complicated cell. For a more correct separation of the mechanisms we have, in the present work, studied together with the temperature dependences of the refraction for different wavelengths, the piezo-optical effect and the dispersion of refraction at room temperature, while in $KNiF_3$ the temperature dependences of the piezoelectric effect were also studied.

2. SPECIMENS AND EXPERIMENTAL METHOD

The cubic crystals $KNiF_3$, $RbMnF_3$ and $KMgF_3$, studied in the present work, have the perovskite type structure and are described by the space group O_h^7. The crystal $KMgF_3$ is diamagnetic, while $KNiF_3$ and $RbMnF_3$ go over to the antiferromagnetic state at temperatures of 246 and 83 K respectively.[6,7] Fluorides with the general formula ABF_3 are transparent in the visible part of the spectrum. The fundamental absorption edge lies at about 6 eV, and the center of gravity of the fundamental band lies in the region[8] 10–15 eV. There is an absorption line close to laser frequencies in compounds with nickel.[7] The $^3A_{2g}\rightarrow{}^3T_{1g}^b$ transition in $KNiF_3$ gives strong absorption near 24000 cm^{-1} and the crystal is opaque for laser radiation with wavelength 0.4416

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μm. There is a relatively weak $^3A_{2g} \rightarrow {}^1E_g^a$ transition near the laser radiation of wavelength 0.6328 μm, but transmission of laser light remains fairly strong.

The specimens were prepared from crystals oriented along [100] type axes. Prisms with angle $\approx 60°$ were cut out of single crystals for goniometric measurements. The faces of the prisms were polished with an error of ≈ 0.5 of an interference fringe.

Investigations of the temperature variations of refractive index were carried out with a homodyne interferometer, described before.[9] The sensitivity in measuring the change in refractive index for a 1 mm crystal thickness was $\delta n \lesssim 10^{-6}$, but the uncertainty increased to 10^{-5} for a long-duration measurement due to zero drift of the interferometer. For analyzing the results of the measurements according to Eq. (1) of Ref. 9, results[10–12] on the thermal expansion of the crystals were used, while for $KNiF_3$ and $KMgF_3$ the thermal expansion was also measured with a quartz dilatometer in the range 300–600 K.

The temperature investigations of refractive index were carried out at helium-neon laser wavelengths 0.6328, 1.1523 μm and a helium-cadmium laser wavelength 0.4416 μm. We made measurements at three wavelengths simultaneously, using the system illustrated in Fig. 1, in order to increase the accuracy of relative measurements at different frequencies. As a result, the zero drift of the interferometer itself does not contribute to the relative changes in the refraction of light, while zero drift associated with the lasers was roughly the same and small. The signal, comprising information on the changes $\delta n(T)$ was recorded simultaneously in three identical channels, from which the relatively weak dispersion of the temperature variations of refractive index could be determined reliably.

Helium and nitrogen cryostats were used for temperature measurements of the refraction of light, with which the temperature could be regulated from 20 to 350 K and from 80 to 700 K respectively. For the measurements, the specimen was housed on a cold finger in vacuo, the specimen temperature being controlled within an accuracy of 0.1 K by copper-constantan and copper/iron-copper thermocouples.

The piezo-optical effect was measured at room temperature using a homodyne interferometer, with the help of a beam press, by applying a uniaxial stress up to 300 kg·cm^{-2}

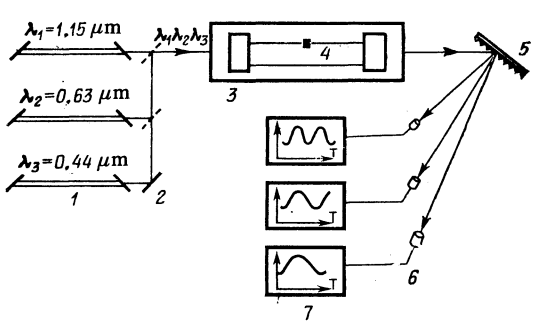

FIG. 1. Arrangement of the apparatus designed for simultaneous investigation at three wavelengths of the temperature variations of the absolute refractive index: 1) lasers, 2) mirrors, 3) interferometer, 4) specimen, 5) diffraction grating, 6) photoreceiver, 7) potentiometer.

along the crystallographic axes perpendicular to the light propagation in the crystal. We estimate the uncertainty in determining the piezo-optical coefficients as $\pm 20\%$.

Measurements were also made of the temperature dependences of the birefringence produced by uniaxial compression along the [010] axis in $KNiF_3$. The measurement was made by the method described earlier.[13] A beam press, housed in a nitrogen dewar was used to provide the uniaxial stress. The measuring sensitivity was $\Delta n \approx 10^{-8}$. The scatter in the measured values of $\Delta n(T)$ from specimen to specimen was $\approx 10\%$.

The piezo-optical coefficients were calculated from the results of the measurements according to the equations

$$\pi_{11} = -\frac{2}{n^3}\left(\frac{\delta\psi_1}{\delta\sigma_1} - (n-1)S_{12}\right), \tag{3}$$

$$\pi_{12} = \pi_{21} = -\frac{2}{n^3}\left(\frac{\delta\psi_2}{\delta\sigma_1} - (n-1)S_{12}\right), \tag{4}$$

where $\delta\psi_{1,2}$ are the measured changes in optical path for light polarizations along and perpendicular to the stress, σ_1 is the applied stress, S_{12} are the stiffness compliances calculated from the elastic stiffness.[14–16] The elasto-optical coefficients were found from values of the elastic stiffness coefficients and the piezo-optical coefficients.

The dispersion of the refractive index was studied at room temperature using a G-1.5 goniometer, by the minimum deviation method, with an uncertainty $\pm 5 \times 10^{-5}$.

3. EXPERIMENTAL RESULTS

A. Dispersion and temperature variations of refractive index

Results on the dispersion of the refractive indices in the three crystals are given in Table I. The values for $KNiF_3$ are somewhat higher ($\sim 5 \times 10^{-3}$) than are given by Ganot,[18] who used the Pulfrich method. The dispersion of the refractive index for all the crystals can be described by the single-oscillator equation

$$n^2 - 1 = f/(E_0^2 - E^2), \tag{5}$$

where f is the effective oscillator strength, E_0 is its energy, E is the photon energy. The parameters of this equation for the three crystals are also shown in Table I.

The temperature variations of refractive index are shown in Fig. 2. On approaching the temperature of the magnetic phase transition, an appreciably larger increase in refractive index is observed in the magnetically ordered dielectrics than in the diamagnetic $KMgF_3$. The largest changes take place at the Néel point at which a break is clearly observed in the $\delta n(T)$ plot for $KNiF_3$ and $RbMnF_3$, while the temperature derivative of refractive index experiences a discontinuity (Fig. 3). It can be seen clearly from Figs. 2 and 3 that short range magnetic ordering is reflected in the variations of refractive index well before the Néel temperature. The refractive index increases under the influence of magnetic ordering.

We evaluated the temperature dependence of the dispersion parameters f and E_0 in $RbMnF_3$ (Fig. 4) on the single oscillator approximation, using the temperature dependences of the refractive index at these wavelengths. This de-

TABLE I. Dispersion of the refractive index of cubic crystals.

λ, μm	n		
	$KMgF_3$ [17]	$KNiF_3$	$RbMnF_3$
1,1523	–	1,4852	–
0,6328	–	1,4915	1,4825
0,6433	1,4021	–	–
0,629	1,4023	–	–
0,605	1,4027	–	–
0,5790	1,4032	1,4932	1,4840
0,5770	1,4033	1,4933	1,4840
0,5461	1,4039	1,4946	1,4852
0,5016	–	–	1,4872
0,4922	–	–	1,4877
0,495	1,4056	–	–
0,4415	–	–	1,4911
0,4358	1,4076	–	1,4915
0,4047	–	–	1,4943
E_0, eV	16,6	13,2	13,8
f, eV^2	267,2	210,7	221,9

pendence shows that the magnetic changes in refractive index are associated with a change both in frequency and in effective oscillator strength. The frequency shift is approximately $\delta E_0 = 0.05$ eV or 0.4% of the frequency 14 eV, while the change in oscillator strength $\delta f = 1.4$ eV^2 or 0.6%.

B. Piezo-optical effect

The temperature dependences of double refraction for light of wavelength 0.6328 μm propagating along the [100] axis of a $KNiF_3$ crystal are shown in Fig. 5 for different values of the uniaxial stress σ, applied along the [001] axis. There is practically no birefringence in the cubic $KNiF_3$ for $\sigma = 0$ and room temperature. The small birefringence associated with crystal imperfection had a value less than 10^{-6}. On reducing the temperature below $T_N = 246$ K, a magnetic contribution to the birefringence appears, associated with the transition of the crystal into the antiferromagnetic state. The reproducibility of the results was poor for small stresses, which is most likely connected with the multidomain magnetic structure. We observed antiferromagnetic domains visually. They are banded structures with walls, in agreement with Safa *et al.*[19] parallel to axes of the [110] type. The dependence of Δn on stress at temperatures of 294 and 121 K is shown in the inset to Fig. 6. At a temperature above T_N the birefringence is a linear function of stress, while below T_N the linearity appears after the specimen becomes single domain at $\sigma > 6\times10^7$ dyne cm^{-2}. The temperature dependence of the difference between the piezo-optical coefficients $\pi_{11}-\pi_{12}$, obtained from measurements of birefringence, is shown in Fig. 6. An important conclusion follows from this result, namely that within the limits of experimental accuracy, the piezo-optical coefficients do not change on varying the temperature, including in this magnetic ordering. The departure of the points from a linear dependence in the region of 200 K is most probably associated with the nearby $^1E_g^a$ absorption line which shifts with change in temperature.[20] A similar conclusion about the temperature independence of the piezo-optical coefficients was drawn earlier[21] for the uniaxial antiferromagnetic MnF_3. A further increase in the accuracy of the experiment is evidently required to elucidate the magnetic contribution to the piezo-optical coefficients. In principle, the piezo-optical coefficients can also change under the influence of short-range magnetic ordering, which can be detected on the $\Delta n(T)$ curves at different stresses. As the stress is increased, the difference between the directions along the perpendicular to the stress should grow, and this should lead to the appearance of an isotropic contri-

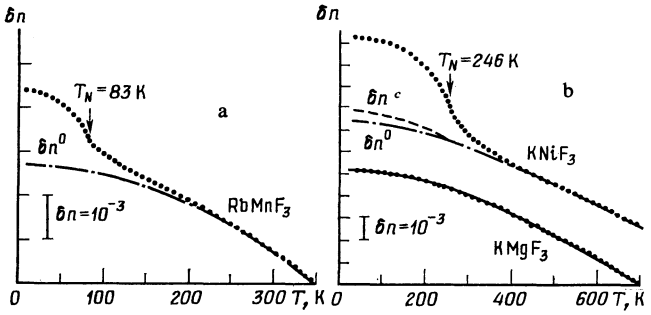

FIG. 2. The temperature variations of refractive index of $RbMnF_3$ (a); of $KNiF_3$ and $KMgF_3$ (b) at a wavelength 0.6328 μm (points): the dash-dot curves of δn^0 are calculated according to Eq. (7); the dashed curve of δn^0 is calculated by taking account of magnetostriction.

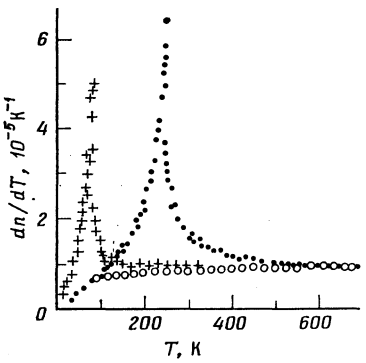

FIG. 3. Temperature dependence of dn/dT in the three crystals studied at a wavelength $\lambda = 0.6328$ μm: +) $RbMnF_3$, ●) $KNiF_3$, ○) $KMgF_3$.

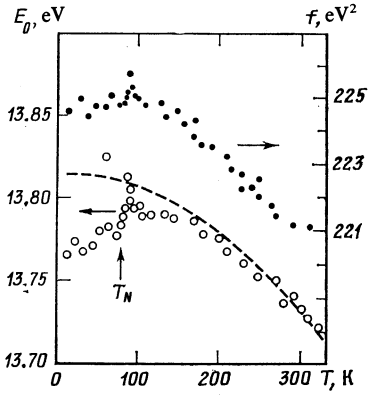

FIG. 4. Temperature dependences of the dispersion parameters f (●) and E_0 (○) from Eq. (5) in $RbMnF_3$. Dashed curve is the change in the parameter E_0 unrelated to magnetic ordering.

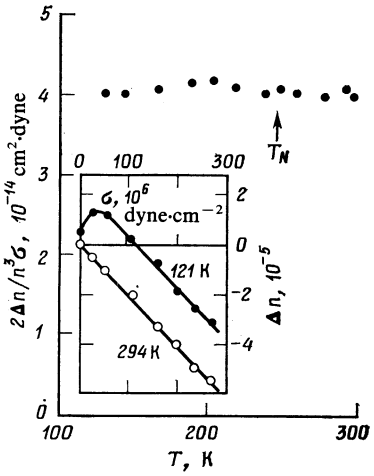

FIG. 6. Temperature dependence of the piezo-optical effect in $KNiF_3$ for a wavelength $\lambda = 0.6328\,\mu$m. The inset shows the birefringence induced at two temperatures.

bution to the birefringence in cubic crystals. However, calculations based on a comparison of the intrinsic optical anisotropy and isotropic magnetic birefringence and refraction with that induced by uniaxial stress, showed that a stress 3–4 orders of magnitude greater than used in the present work is required to observe such a magnetopiezo-optical effect.

Results on the piezo-optical effect at room temperature, obtained by measurement of the principal values of the piezo-optical tensor, are shown in Table II. Values of the difference between the coefficients $\pi_{11}-\pi_{12}$, obtained by us in measurements of birefringence and refractive index, agree with recently published[7] results of measuring the linear birefringence in $KNiF_3$. We draw attention to the fact that all the elasto-optical coefficients are positive, i.e. compression of the crystal leads to an increase in refractive index.

4. THE MECHANISMS FOR TEMPERATURE VARIATIONS OF THE REFRACTIVE INDEX IN CUBIC CRYSTALS

In the general case the change in the inverse dielectric susceptibility tensor of a magnetic crystal for a small temperature change is described by Eq. (4) of Markovin and Pisarev.[9] An equation can be obtained from this expression for the temperature variations of the refractive index of cubic crystals studied in the present work:

$$\Delta[\delta n(T)] = -{}^1/_2 (n^0)^3 [\mu^0 + (p_{11}{}^0 + 2p_{12}{}^0)\alpha^0]\Delta T + \Delta(\delta n^m), \quad (6)$$

where μ^0 is the thermo-optical effect tensor; p_{11}^0 and p_{12}^0 are elasto-optical coefficients, α^0 is the principal value of the thermal expansion tensor. The term with μ^0 describes the isochoric temperature variation of the refraction of light, the term $(p_{11}^0 + 2p_{12}^0)\alpha^0$ determines the change in refractive index due to the photoelastic effect on thermal expansion of the crystal, δn^m is the magnetic addition due to short and long-range magnetic order.

By using results of measurements of the temperature variations of refractive index, elasto-optical coefficients, the literature[10–12] and our own results on thermal expansion, we have calculated the magnitude of the photoelastic and thermo-optical contributions to the temperature derivative in $KNiF_3$, $RbMnF_3$ and $KMgF_3$ at room temperature, where the influence of short and long-rang magnetic ordering on these effects can be neglected (Table III). Analysis of Table III shows that both effects, the thermo-optical and photoelastic, provide an appreciable contribution to dn/dT in the diamagnetic $KMnF_3$, while in $RbMnF_3$ the variation dn/dT at room temperature is mainly determined by the photoelastic effect.

The temperature variation of the photoelastic and thermo-optical effects must be correctly extrapolated in order to separate out the contributions to the thermal changes in the refractive index. We conluded above, on the basis of results of studying the piezo-optical effect in $KNiF_3$, that the elasto-optical coefficients in this crystal practically do not change on changing the temperature and magnetic order, in comparison with the thermal expansion tensor. In what follows we shall assume that this situation is true for $RbMnF_3$ and $KMgF_3$. The temperature dependence of the photoelastic contribution will then be determined by the formula

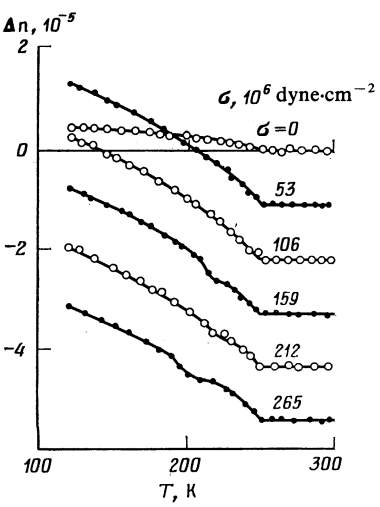

FIG. 5. Temperature dependences of the birefringence in $KNiF_3$ at a wavelength $\lambda = 0.6328\,\mu$m at different values of the uniaxial stress along the [001] axis.

TABLE II. Piezo-optical π_{ij} (in units of 10^{-13} cm^2 dyne^{-1}) and elasto-optical p_{ij} coefficients of cubic fluorides at a wavelength of 0.6328 μm.

Crystal	π_{11}	π_{12}	p_{11}	p_{12}
$KNiF_3$	+0.22	+0.67	+0.10	+0.15
$RbMnF_3$	+0.49	+0.87	+0.13	+0.15
$KMgF_3$	+0.20	+1.36	+0.13	+0.24

$$\delta n^{ph}(T)=A^{ph}\delta l(T)/l, \quad A^{ph}=1/2(n^0)^3(p_{11}+2p_{12}), \qquad (7)$$

where $\delta l(T)/l$ is the thermal expansion of the crystal. We assumed that this formula can also be extended to those crystals for which the thermo-optical effect provides an appreciable contribution to the temperature variation of the refractive index. The results of such an approximation for $KMgF_3$ are shown in Fig. 2. The coefficient A^{ϕ} is determined in the high temperature region. Approximate curves for this crystal give an error of only 5×10^{-5} at low temperatures. We can understand the physical meaning of such an approximation for the thermo-optical effect in cubic crystals on the basis of the same considerations as we introduced[9] for the tetragonal MnF_2.

5. ISOTROPIC AND ANISOTROPIC MAGNETIC CONTRIBUTIONS TO THE REFRACTIVE INDEX OF ANTIFERROMAGNETIC FLUORIDES

When using Eq. (7) for magnetic dielectrics, the magnetostriction must be removed from the thermal expansion. We carried out such a calculation for $KNiF_3$ and $RbMnF_3$, using the coefficients from Table III. To exclude the magnetostriction, the thermal expansion was extrapolated from the high temperature region on the Debye approximation. In Fig. 2 are shown the experimental temperature dependences of refractive index and the calculated curves for the thermal behavior of the refraction, unrelated to magnetic ordering. The difference between the $\delta n(T)$ and $\delta n^0(T)$ curves is determined by the total, i.e. the isotropic and anisotropic, magnetic contribution to the refractive index. The total contribution is $\delta n^m = 3.7\times10^{-3}$ in $KNiF_3$ and 1.7×10^{-3} in $RbMnF_3$.

If the total magnetic contribution to the refractive index is found from the temperature variation of the absolute refractive index, then the anisotropic magnetic contribution can be found from measurements of the magnetic linear birefringence $\Delta n = n_{\parallel}-n_{\perp}$, to which the isotropic part does not make a contribution. The temperature dependence of LMB in $KNiF_3$ is shown in Fig. 7, obtained by approximating to zero stress the temperature dependences of birefringence at different stresses. The temperature dependence of the total magnetic contribution is shown in the same figure. There is no anisotropic magnetic birefringence above T_N, while below T_N it increases on decreasing the temperature. In $KNiF_3$ it reaches a value $\Delta n_{aniso} = 2.2\times10^{-5}$ at 120 K, which is about two orders of magnitude less than the total magnetic contribution. The anisotropic contribution in $RbMnF_3$, $\Delta n_{aniso} = (1.3\text{–}2.2)\times10^{-6}$ (Ref. 7), which is three orders of magnitude less than the total magnetic contribution we measured. In these two crystals the magnetic contribution to the refractive index is thus isotropic to high accuracy. Applying the terminology used in crystal optics, we can say that the main effect numerically of the influence of magnetic ordering on the refraction of light in cubic crystals is not the change in the symmetry of the optical indicatrix due to anisotropic additions, as often suggested earlier, but is the isotropic change in its dimensions, i.e. the isotropic magnetic refraction effect. It is interesting to note that in all the crystals studied by us so far, namely in the fluorides $KNiF_3$, $RbMnF_3$, in the uniaxial antiferromagnet MnF_2 (Ref. 9), in the biaxial antiferromagnet $BaMnF_4$ (Ref. 22), in oxides with the garnet structure[23] and in chalcogenides,[24] the isotropic magnetic contribution is positive and leads to an increase in refractive index on reducing the temperature. In contrast to this, the anisotropic linear magnetic birefringence can be

TABLE III. Photoelastic and thermo-optical contributions to the temperature dependences of the refractive index of cubic fluorides at a temperature $T = 295$ K (for $KNiF_3$ at $T = 650$ K) and wavelength 0.6328 μm.

Crystal	$(dn/dT)^{ph}$, 10^{-5} K^{-1}	$(dn/dT)^{TO}$, 10^{-5} K^{-1}	$A=(dn/dT)/\alpha$
$KMgF_3$	−1.49	+0.74	−0.5
$KNiF_3$	−1.15	+0.05	−0.632
$RbMnF_3$	−1.38	+0.12	−0.631

Note: $(dn/dT)^{ph} = -1/2n^3(p_{11}+2p_{12})\alpha$; $(dn/dT)^{TO} = dn/dT - (dn/dT)^{ph}$.

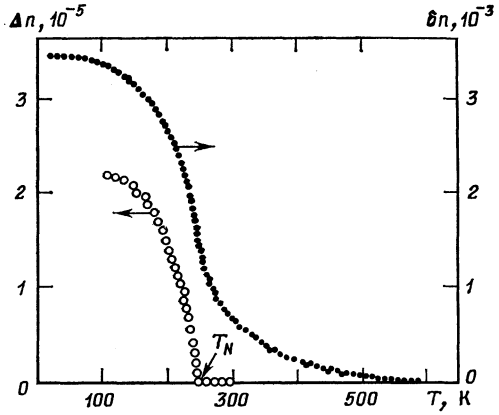

FIG. 7. The temperature dependences of the total (●) and anisotropic (○) magnetic contributions to the refraction of light in $KNiF_3$ at a wavelength $\lambda = 0.6328$ μm. The scale for the two contributions differs by two orders of magnitude.

both positive and negative. These results require a microscopic theoretical explanation.

As has been noted before,[7,9] magnetic ordering has an effect on the energy bands (the magneto-optical mechanism), on the expansion of the crystal (magnetostriction) and on the electron-phonon interaction (magnetothermo-optical mechanism). In addition, a mechanism for the change in birefringence has been proposed,[5] associated with a shift of the ions in the elementary cell, without a change in its symmetry, in particular of the fluorine ions in the tetragonal rutile structure. Such a mechanism is possible, but calculations showed that the present experimental methods do not yet permit the demonstration of such shifts. This mechanism has been criticized by Borovik-Romanov *et al.*[21] who established that on deforming MnF_2 crystals along the [110] axis the changes in birefringence are small, although the shifts of the fluorine ions are then large.

The cubic ABF_3 crystals with perovskite structure studied by us have a unit cell of high symmetry, O_h^7, and in addition *all* the ions in the cell are in special positions with high symmetry having a center of inversion. Ions A and B are in positions with O_h symmetry, while the fluorine ions are in positions with D_{4h} symmetry. In this case internal shifts of the ions within the limits of the cell turn out to be forbidden, since they must change the dimensions and number of atoms in the cell. Due to this the magnetostrictive contribution to the change in refraction and birefringence of perovskites is completely determined by bulk deformations. As a result, the calculated magnetostrictive contribution in $RbMnF_3$ is negligibly small (because of the small magnetostriction), while it represents 10–12% of the total contribution in $KNiF_3$ (Fig. 2). The isotropic magneto-optical mechanism is thus the main source of the changes in refractive index with temperature.

We shall discuss briefly the temperature dependence of the magnetic contributions to the refractive index. According to theoretical ideas,[25] the anisotropic magnetic contribution in $KNiF_3$ (Fig. 7) near the magnetic ordering temperature should vary as the square of the order parameter, which in the present case is the antiferromagnetism vector. Analysis of the results gave a value $\beta = 0.41 \pm 0.02$ for the critical exponent, which agrees with the results of Ferré *et al.*[6] We also calculated values of the critical exponent for $KNiF_3$ and $RbMnF_3$ from results on the temperature dependence of the isotropic magnetorefraction. Since in this case the changes in refraction are proportional to the correlation function,[2] they are determined by both long-range and short-range order. We assumed that near T_N the changes in refractive index due to fluctuations are the same at the same departure from T_N on the high and low temperature sides. Then

$$[\delta n^m(T_N-\Delta T)-\delta n^m(T_N+\Delta T)]\sim\tau^{2\beta}, \qquad (8)$$

where $\tau = (T_N - T)/T$ is the reduced temperature. It turned out that in both antiferromagnetics $\beta = 0.39 \pm 0.02$. This value for $RbMnF_3$ is very different from the value $\beta = 0.32 \pm 0.02$ found from studies of neutron scattering.[26] The reason for such a great disagreement between values of the critical exponent in optical and neutron investigations is not clear to us.

6. CONCLUSIONS

The main experimental results of the present work consist of studies of the changes in refractive index and the piezo-optical effect in three cubic crystals with perovskite structure. The different mechanisms for the temperature variations of refractive index could be justifiably separated out thanks to the comprehensive nature of the investigation. It was established that the nonmagnetic contributions can, with good accuracy, be considered proportional to the linear expansion of the lattice. The isotropic and anisotropic magnetic contributions to the changes in refraction were separated out and it was shown that the former is dominant. At low temperatures these contributions are $\delta n_{iso} = 3.7\times10^{-3}$, $\Delta n_{aniso} = 2.2\times10^{-5}$ in $KNiF_3$ and $\delta n_{iso} = 1.7\times10^{-3}$ and $\Delta n_{aniso} = (1\text{–}2)\times10^{-6}$ (Ref. 7) in $RbMnF_3$. It was shown that because of the high symmetry of the local atomic positions in the cell, internal shifts of atoms in the cell should not provide contributions to the change in refraction, and the magnetostrictive contribution can be considered as the product of a certain combination of the piezo-optical coefficients and the linear expansion resulting from magnetostriction. The magnetostrictive contribution in $RbMnF_3$ turned out to be negligibly small, while in $KNiF_3$ it represented 10–12% of the total magnetic contribution. Values of the critical index could be found from analysis of the temperature variations of the magnetic contribution to the refraction.

We will make another remark as follows. The results of the present and earlier studies[1–4] show that the isotropic and anisotropic refraction (LMB) are essentially different effects. Isotropic refraction is independent of the orientation of the magnetization in the crystal, and also in cubic crystals, of the polarization of the light, while LMB depends both on the orientation of the magnetization and on the polarization of the light. These two effects differ appreciably in magnitude, by 2–3 orders of magnitude, and in their temperature dependences, in that LMB disappears at the magnetic transition temperature, while isotropic refraction is observed at temperatures 2–3 times greater than the ordering temperature. The microscopic mechanisms for these phenomena must also be different. All these facts give one the right to consider the isotropic refraction the sort of effect which should not be called the Cotton-Mouton effect.

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