Magnetic, thermal, and elastic refraction of light in the antiferromagnet MnF_2

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The changes of the principal values of the refractive indices in the tetragonal MnF_2 crystal were investigated in the temperature interval from 15 to 400 K at three laser wavelengths, 0.44, 0.63, and 1.15 μm; the photoelastic constants at room temperature were also measured. An analysis of the temperature dependences of the refractive indices at high temperatures, when the magnetic contribution can be neglected, has shown that the changes are due primarily to the thermo-optical effect, and the contribution of the photoelastic effect plays a lesser role, and its sign is the opposite of that of the thermo-optical effect. The onset of short- and long-range magnetic order in the crystal influences the thermo-optical and photoelastic contributions and leads also to the appearance of magnetic contribution to the refraction. At low temperatures, the magneto-optical contribution for the ordinary refractive index reaches a value $\delta n_o^{mo} = 3.4 \times 10^{-3}$. The magnetic contribution due to the short-range order is observed up to temperatures (2.5–3) T_N. It is established that the temperature dependences of the derivative dn_o^{mo}/dT correlate well with the magnetic heat capacity of MnF_2, and the relative changes of the magneto-optical contribution with changing temperature agree with the relative changes of the first moment of the two-magnon scattering band. From the point of view of dispersion theory, the effect of magnetic refraction of light is due to the magnetic shift of the frequency and of the strength of the effective oscillator.

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1. INTRODUCTION

It is known that an important role is played in the description of optical phenomena by the extinction coefficient and by the refractive index of the medium with which the light interacts. Extinction by magnetically ordered crystals has been the subject of a rather large number of studies, and an extensive bibliography is contained in Ref. 1. Starting with about ten years ago, results have been reported of investigations of magnetic birefringence of light, i.e., the influence of magnetic ordering or of an external magnetic field on the difference between the two principal values of the refractive index. The results of these investigations were considered in part in a review article,[2] and a number of new papers were published in recent years.[3-6] It is well known, however, that a study of birefringence does not make it possible to determine all the constants that describe the influence of external actions, such as an electric or magnetic field, pressure, change of temperature, on the optical properties.[7] Complete information on this influence can be obtained only by studying the absolute refractive index and the changes of all its principal values under the influence of the perturbation. In a number of cases these changes turn out to be stronger than the changes of other parameters, such as the lattice constant. Recognizing that the refractive index and its changes can be measured with high accuracy, one can hope that a study of this characteristic will lead to a better understanding of the connection between the refraction of light and the energy structure of the crystal and its changes.

It should also be noted that in the transparency region the refractive index is an important characteristic that determines the reflection and propagation of light in crystals, and that many optical phenomena (such as light scattering, photoelasticity, and others) are proportional to high powers of this index. A study of the refractive index is also of practical use, since its variation under the influence of external action determines very important characteristics of optical instruments, such as resolution, sensitivity, endurance to high-power optical radiation, and others.

It was recently established that magnetic ordering influences substantially the refractive index of cubic[8,9] and noncubic[10] crystals. These investigations, as well as the already cited studies of magnetic birefringence, raised the serious problem of distinguishing between the different mechanisms that lead to changes of the refraction under the influence of temperature or pressure. To distinguish between different contributions to the change of the refractive index n in the paramagnetic and in the magnetically-ordered regions, we have undertaken the present investigation of the temperature dependences of $n(T)$ at various wavelengths and of the photoelastic effect in the tetragonal antiferromagnet MnF_2.

2. SAMPLES AND MEASUREMENT PROCEDURE

Manganese fluoride MnF_2 has a tetragonal structure of the rutile type and its symmetry is described by the space group D_{4h}^{14}. At a temperature $T_N \approx 67$K the crystal becomes antiferromagnetic with an antiferromagnetism vector L along the fourfold axis. In both the paramagnetic region and in the magnetically ordered region, in the absence of an external magnetic field, MnF_2 is in the visible band an optically uniaxial crystal, and the dispersion of its refractive index was investigated by Jahn.[11] In the visible region of the spectrum, the crystal has a weak dispersion due to interband electronic transitions with an approximate effective frequency 14

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eV, which agrees well with the position of the center of gravity of the spectrum of the imaginary part of the dielectric constant.[12] The width of the forbidden band is approximately 10 eV.

The samples were made of oriented MnF_2 single crystlas and their surfaces were subjected to optical grinding and polishing.

Investigations of the temperature-induced changes of the principal values of the refractive index were made with a two-beam homodyne interferometer illustrated in Fig. 1. The light sources l were helium–neon lasers operating at 0.6238 and 1.1523μ m and a helium–cadmium laser with wavelength 0.4416 m. The phase difference between the "object" beam E_1 and the reference beam E_2 was modulated at a frequency ~1 kHz by modulator 2, the modulation being effected between the orthogonal components E_1 and E_2 of the polarization of the entering light beam prior to their separation by the polarization beam splitter 3. Variation of the length of the optical path $\delta\psi$ of the light in the crystal placed in the object beam caused changes in the phase difference $\delta\varphi$ between the two interferometer beams. These changes are connected by the relation $\delta\psi = \delta\varphi\lambda/2\pi l$, where λ is the wavelength of the light and l is the crystal thickness. The signal amplitude of the first harmonic of the photocurrent, separated by selective amplifier 14 at the modulation frequency, was proportional to $\sin\delta\varphi$. This signal was transformed into a dc voltage, and an x–y potentiometer 10 registered its variation with temperature or with uniaxial compression.

The installation permitted also null-method measurements with the aid of an optical compensator 5. In measurements of the temperature dependence of the refractive index, the variation of the latter was calculated from the equation

$$\delta n = \delta\psi - (n-1)\delta l/l. \qquad (1)$$

At a crystal thickness $l \approx 1$ mm the sensitivity to changes of the refractive index was $\delta n \leq 10^{-6}$, but in prolonged measurements, owing to the slow drift of the interferometer null point, the measurement accuracy decreased to $\delta n \approx 10^{-5}$. The measurement results were reduced

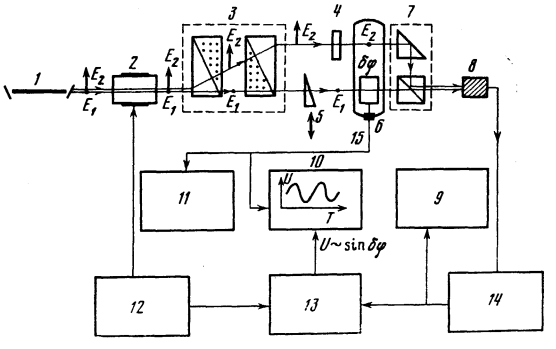

FIG. 1. Diagram of homodyne interferometer: 1—laser, 2—ML-3 electro-optical modulator, 3—polarizing light-beam splitter, 4—quartz 90° plate, 5—phase compensator, 6—cryostat with sample (or press with sample), 7—beam-convergence unit, 8—FTT-5 phototransistor, 9—monitoring oscilloscope, 10—LKD-4 x-y recorder, 11—digital microvolt meter F-30, 12—sound-frequency generator, 13—ac voltage converter V9-2, 14—U2-6 tuned amplifier, 16—thermocouple.

with the aid of the Gibbons data[12] on the thermal expansion of MnF_2, whose accuracy $\approx 10^{-5}$ corresponded to the accuracy of our measurements.

In the temperature measurements we used a helium cryostat that made it possible to regulate the temperature from 15 to 400 K. In the measurements the samples were placed on a cold finger in vacuum, and the sample temperature was monitored with accuracy ~0.1 deg by two thermocouples, copper–constantan and copper–iron–copper.

The photoelastic effect at room temperature was measured with a lever press[13] with uniaxial pressure up to ≈ 100 kgf/cm^2 applied along the crystallographic axes perpendicular to the propagation of the light in the crystal. The piezo-optical constants π_{ij} were calculated from the results of measurements using equations in the form (the equation is given for π_{11})

$$\pi_{11} = -\frac{2}{n_o^3}\left(\frac{\delta\psi_1}{\sigma_1} - (n_o-1)S_{12}\right), \quad \mathbf{k}\|\mathbf{c}, \qquad (2)$$

where $\mathbf{k}$ is the wave vector of the light wave, $\delta\psi_i$ are the measured values of the optical path, σ_i is the applied pressure, and S_{ij} are the elastic compliances calculated from the values of the elastic rigidities.[14] The photoelectric coefficients were calculated next from the values of the piezoelectric constants and elastic rigidities.

The accuracy with which the changes of the refractive indices were measured under pressure was $\approx 10^{-6}$, but the results depended of the reproducibility of the sample mounting, of its correct orientation, and of the uniformity of the pressure. According to our estimates, these factors decreased the accuracy of the photoelectric constants to 15–20%.

3. EXPERIMENTAL RESULTS

The temperature changes $\delta n_o(T)$ and $\delta n_e(T)$ of the principal values of the refractive index of MnF_2 at various wavelengths are shown in Fig. 2. The subscripts o and e pertain respectively to the ordinary and extraordinary refractive indices. These results make it possible to calculate the temperature dependence of the birefringence $\delta(n_e - n_o)$, which agreed within $\approx 10^{-5}$ in the para-region and within $\approx 5\times 10^{-5}$ at 15 K with the data of Jahn,[11] Borovik-Romanov,[15] and others. At high temperature the principal indices of MnF_2 increase monotonically, and their difference $n_e - n_0$ decreases with increasing temperature. However, as the magnetic-ordering point is approached from the paramagnetic region, the decrease of the indices stops gradually and they begin to increase rapidly. Obviously, the latter is due to establishment of magnetic order in the crystal. At a temperature $T_N = 67$ K the changes of the refractive index are maximal, and the plots of $\delta n_{o,e}(T)$ show clearly a kink at this temperature (see the inset of Fig. 2a), while the derivative with respect to temperature $dn_{o,e}/dT$ changes jumpwise. Figure 2 shows clearly that the influence of the short-range magnetic order on the refractive index sets in at temperatures greatly exceeding the magnetic-ordering temperature.

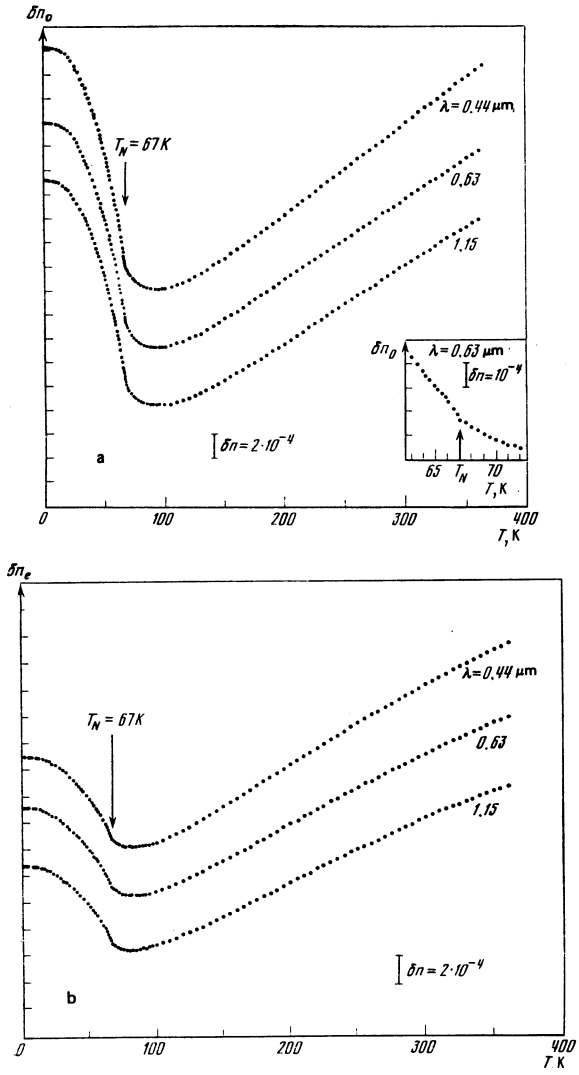

FIG. 2. Temperature changes of ordinary (a) and extraordinary (b) refractive indices in MnF_2 at different wavelengths.

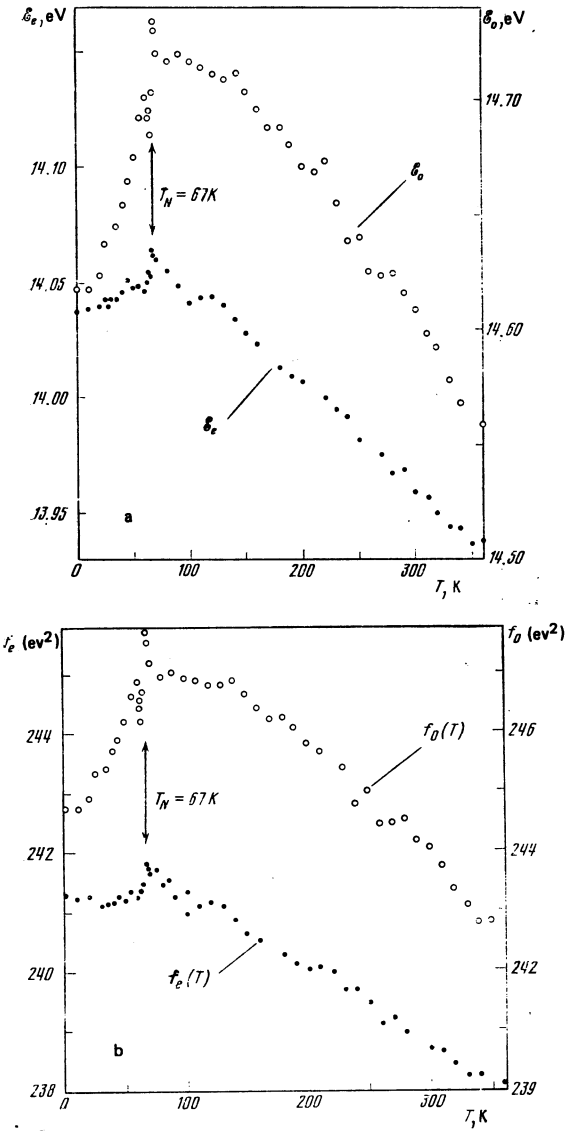

FIG. 3. Temperature dependences of the frequency (a) and of the strength (b) of the effective oscillator for the ordinary and extraordinary refractive indices in MnF_2.

If it is assumed that the dispersion of the refractive index in MnF_2 in the visible and in the near-infrared regions is determined by the high-lying transitions with energies exceeding 10 eV, then we can describe this dispersion by using the single-oscillator formula

$$n_{o,e}^2-1=f_{o,e}/(\mathscr{E}_{o,e}^2-\mathscr{E}^2), \qquad (3)$$

where $f_{o,e}$ is the strength of the effective oscillator, $\varepsilon_{o,e}$ is its energy, and ε is the photon energy. On the basis of data on the dispersion of the refractive index and on the basis of the results shown in Fig. 2 for several wavelengths we can then calculate the temperature dependences of $f_{o,e}(T)$ and $\varepsilon_{o,e}(T)$, which are shown in Figs. 3a and 3b. It is seen there that the magnetic ordering leads to a change of both the frequency and the strength of the effective oscillator. These changes are respectively

$$\delta\mathscr{E}_o^m\approx 0.16 \text{ eV } (1.0\%), \quad \delta\mathscr{E}_e^m\approx 0.08 \text{ eV } (0.6\%),$$
$$\delta f_o^m\approx 4.1 \text{ (eV)}^2 \ (1.6\%), \quad \delta f_e^m\approx 2.2 \text{ (eV)}^2 \ (0.9\%).$$

The results of the investigation of the photoelastic effect at the wavelength $0.63\mu m$ are given in Table I. The variance of the photoelastic constants lies in the range $\approx 20\%$, which is at the borderline of the error with which the constants themselves are determined.

The values of the constant π_{11} measured in two different experimental geometries agreed within $\approx 15\%$. In the calculation of the photoelastic constants we chose the value $\pi_{11}=1.31\times10^{-13}$ cm^2/dyn. Our results agree within the limits of experimental error with the data of Borovik-Romanov, Kreines, and Paces,[16] who investi-

TABLE I. Relative changes of the optical thickness of the crystal per unit pressure under uniaxial compression, and the piezo- and photoelastic constants in MnF_2.

Index ij of tensor component	$(d\psi/d\sigma)_{ij}\cdot10^{13}$, cm^2/dyn	$\pi_{ij}\cdot10^{13}$, dm^2/dyn	p_{ij}	Direction of light
11	+0.30	−1.22	+0.10	k∥c
11	−6.10	−1.41		k⊥c
12	−5.40	+2.36	+0.19	k∥c
31	−10.10	+0.73	+0.12	k⊥c
13	−2.80	+0.72	+0.19	k⊥c
33	−1.36	−0.23	+0.06	k⊥c

gated the piezo-optical effect with the aid of the birefringence of the light. Our measurements yield

$$(d\psi/d\sigma)_{33}-(d\psi/d\sigma)_{13}=1.44\cdot 10^{-13}\,\text{cm}^2/\text{dyn},$$

while the birefringence data yield for the analogous quantity also the value 1.44×10^{-13} cm^2/dyn. According to our data

$$\pi_{11}+\pi_{12}-2.11\pi_{31}=-0.48\cdot 10^{-13}\,\text{cm}^2/\text{dyn},$$

as against $+0.48\times10^{-13}$ cm^2/dyn in accordance with the data of Ref. 16. These results are in agreement within a 15–20% variation of all the constants π_{11}, π_{12}, and π_{31}. We call attention to the fact that all the photoelastic constants are positive, i.e., the photoelastic effect should cause the refractive index of the crystal to increase under compression and to decrease under expansion.

4. MECHANISMS OF THERMAL CHANGE OF THE REFRACTIVE INDEX

In the general case the change of the inverse dielectric tensor ε_{ij}^{-1} of a magnetic crystal following a small change of temperature ΔT is given by

$$\Delta(\varepsilon_{ij}^{-1})=(\mu_{ij}^{0}+p_{ijkl}^{0}\alpha_{kl}^{0})\Delta T+\Delta\delta(\varepsilon_{ij}^{-1})^{M}, \tag{4}$$

where μ_{ij} is the tensor of the thermo-optical effect and describes the isochoric temperature change of the refraction of the light, p_{ijkl} is the tensor of the photoelastic effect, and α_{kl} is the thermal-expansion tensor.[17] The term $p_{ijkl}\alpha_{kl}$ determines the change of the refractive index on account of the photoelastic effect in thermal expansion of the crystal; the zero superscript labels the part of the tensor which is not connected with the magnetic ordering; $\delta(\varepsilon_{ij}^{-1})^{M}$ is the magnetic increment due to the onset of short- and long-range magnetic order in the crystal; this increment is discussed in detail in the next section.

We have established in Sec. 3 that in the investigated part of the optical band the refractive index of MnF_2 is determined by electronic interband transitions. The change of the temperature influences primarily the crystal-lattice vibrations, and these lead in turn to a change of the electron-phonon interaction. From the microscopic point of view, the temperature dependence of the refractive index (both the thermo-optical and the photoelastic contributions) is therefore due to changes of the electronic spectrum under the influence of the electron-phonon interaction.

For those temperature changes of the principal values of the refractive index of MnF_2 which are not connected with the magnetic ordering we can write

$$\begin{aligned}\Delta n_o^{0}&=-{}^1/_2(n_o^{0})^3[\mu_1^{0}+(p_{11}+p_{12})\alpha_1^{0}+p_{13}\alpha_3^{0}]\Delta T,\\ \Delta n_e^{0}&=-{}^1/_2(n_e^{0})^3[\mu_3^{0}+2p_{31}\alpha_1^{0}+p_{33}\alpha_3^{0}]\Delta T.\end{aligned} \tag{5}$$

Measurements of the photoelastic effect[16] based on the birefringence of light in MnF_2 have shown that different combinations of the photoelastic constants depend very little on the temperature and are hardly altered by a magnetic phase transition. It was therefore concluded in Ref. 16 that the photoelastic constants of MnF_2 are themselves independent of the temperature and of the magnetic ordering. We can thus put $p_{ijk}^{0}{}^{l}\approx p_{ijkl}$, in (5) and conclude that the photoelastic contribution to the temperature dependence of $n_{o,e}(T)$ changes only as a result of thermal expansion, i.e., it is proportional to a linear combination of α_1 and α_3.

To calculate the temperature dependence of the thermo-optical effect, we differentiate Eq. (3) with respect to temperature:

$$\begin{aligned}\Delta n_{o,e}=&\frac{n_{o,e}^2-1}{2n_{o,e}}\Big[\frac{1}{f_{o,e}}\Big(\frac{\partial f_{o,e}}{\partial u_{1,1}}+\frac{\partial f_{o,e}}{\partial u_{2,1}}\Big)\alpha_1+\frac{1}{f_{o,e}}\frac{\partial f_{o,e}}{\partial u_{3,3}}\alpha_3\\ &-\frac{2\mathscr{E}_{o,e}}{\mathscr{E}_{o,e}^2-\mathscr{E}^2}\Big(\frac{\partial\mathscr{E}_{o,e}}{\partial u_{1,1}}+\frac{\partial\mathscr{E}_{o,e}}{\partial u_{2,1}}\Big)\alpha_1-\frac{2\mathscr{E}_{o,e}}{\mathscr{E}_{o,e}^2-\mathscr{E}^2}\frac{\partial\mathscr{E}_{o,e}}{\partial u_{33}}\alpha_3\\ &-\frac{2\mathscr{E}_{o,e}}{\mathscr{E}_{o,e}^2-\mathscr{E}^2}\Big(\frac{\partial\mathscr{E}_{o,e}}{\partial T}\Big)_u+\frac{1}{f_{o,e}}\Big(\frac{\partial f_{o,e}}{\partial T}\Big)_u\Big]\Delta T,\end{aligned} \tag{6}$$

where $u_1=u_2$ and u_3 are the strains along the crystallographic axes. The last two terms of (6) describe the thermo-optical effect and the remainder the thermoelastic effect.

We consider first the thermo-optical part of the temperature dependence of the effective-oscillator frequency. The electron–phonon interaction produces an isochoric shift of the electron energy levels in the crystal.[18,19] We assume, as in Refs. 18 and 19, that in MnF_2 this temperature shift is proportional to the internal (phonon) crystal energy $\varepsilon'(T)$. We can then write for thermo-optical frequency shift of the effective oscillator

$$\delta\mathscr{E}_{o,e}^{\text{to}}=d_{1,3}\mathscr{E}'(T). \tag{7}$$

On the other hand, from the thermodynamic theory of crystals we have an equation that connects the internal energy with the thermal expansion[20]:

$$\delta\mathscr{E}'(T)=\gamma_1 V(c_{11}^{t}+c_{12}^{t})\Big(\frac{\delta l}{l}\Big)_1+\gamma_1 V c_{13}^{t}\Big(\frac{\delta l}{l}\Big)_3, \tag{8}$$

where γ_1 is the Grüneisen parameter, V is the molar volume, and c_{ij}^{t} are the isothermal elastic constants. It follows from (7) and (8) that the isochoric temperature dependence of the oscillator frequency can be approximated by the equation

$$\delta\mathscr{E}_{o,e}^{\text{to}}=a_{1,3}\Big(\frac{\delta l(T)}{l}\Big)_1+b_{1,3}\Big(\frac{\delta l(T)}{l}\Big)_3, \tag{9}$$

where $a_{1,3}b_{1,3}$ are constants that do not depend on the temperature (accurate to the temperature dependence of the Gruneisen parameters and of the elastic constants). Taking into account the statements made above concerning the photoelastic contribution, we arrive at the conclusion that on the whole the temperature dependence of the oscillator frequency can be approximated by an equation similar to (9).

We discuss now the thermal changes of the effective-oscillator strength. It is seen from Fig. 3 that the temperature dependences of the strength and of the frequency of the oscillator are similar for the corresponding polarizations of the light. This makes it possible to extend the approximating equation of the type (9) to cover also the change of the oscillator strength [including the thermo-optical contribution to $\delta f_{o,e}(T)$, i.e., the last term of (6)].

From the foregoing analysis we can conclude that although the thermo-optical contribution is not a consequence of the thermal expansion of the crystal but is

determined by the isochoric electron–phonon interaction, its temperature dependence is similar to that of a linear combination of the lattice parameters. This connection between the thermo-optical effect and the thermal expansion is similar to the connection between the heat capacity and the thermal expansion. We can now approximate those temperature changes of the refractive index of MnF_2 which are connected with magnetic ordering by the expression

$$\delta n_{o,e}^{0}=A_{1,3}\left(\frac{\delta l(T)}{l}\right)_{1}^{0}+B_{1,3}\left(\frac{\delta l(T)}{l}\right)_{3}^{0}, \qquad (10)$$

where $A_{1,3}$ and $B_{1,3}$ are coefficients independent of temperature. We emphasize once more that Eq. (10) should be regarded just as a convenient approximation of the temperature dependence of $n_{o,e}(T)$, since only part of the thermal change of the refractive index, namely the photoelastic contribution, is directly caused by the change of the lattice parameters.

The coefficients $A_{1,3}$, $B_{1,3}$ were calculated by a least-squares matching of Eq. (10), in which the thermal-expansion parameters were taken to be the temperature changes of the MnF_2 lattice constants from the paper of Gibbons,[12] to the experimental plots of $\delta n_{o,e}(T)$ (Fig. 2) in the temperature interval 250–360 K. In this temperature interval we can neglect the influence of the magnetic ordering. The comparison is clearly reflected in Fig. 4. The values of the expected temperature changes of the refractive index $\delta n_{o,e}$, calculated from Eq. (10) lie on straight lines when plotted in the coordinates indicated in the figure. The light and dark circles are the experimentally determined values of the ratio $(dn_{o,e}^{exp}/dT)/\alpha_1$ for the ordinary and extraordinary rays, respectively. The calculated relations agree well with the experimental ones down to ≈200 K. The discrepancy with decreasing temperature is due to the appearance of a magnetic contribution to the refractive index.

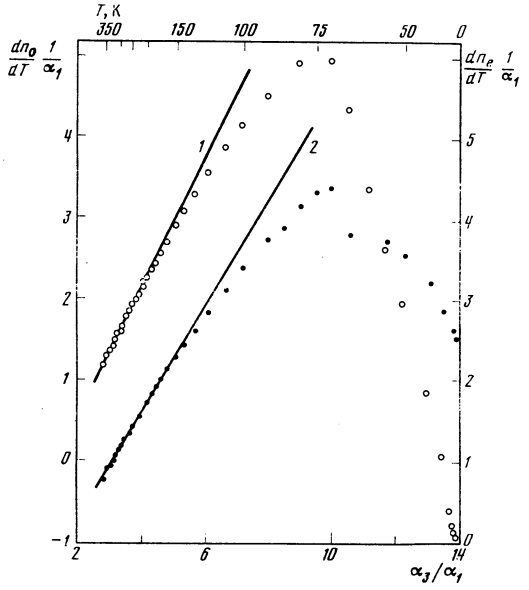

FIG. 4. Approximation of the experimental temperature dependences of the ordinary (○) and extraordinary (●) refractive indices of MnF_2 at a wavelength 0.63 μm by direct lines calculated in accordance with Eq. (10): line 1—$(dn_o^0/dT)/\alpha_1 = -1.03 + 0.79\alpha_3/\alpha_1$, 2—$(dn_e^0/dT)/\alpha_1 = -1.06 + 0.66\alpha_3/\alpha_1$.

TABLE II. Coefficients of the approximating formula (10) and values of the magneto-optical contribution to the refraction of light in MnF_2 at various wavelengths.

λ, μm	A_1	B_1	A_3	B_3	δn_o^{mo}, 10^{-3}	δn_e^{mo}, 10^{-3}
0.44	−1.11	+0.90	−1.32	+0.78	3.38	1.84
0.63	−1.03	+0.79	−1.06	+0.66	3.06	1.59
1.15	−1.03	+0.77	−1.15	+0.65	3.07	1.62

The coefficients $A_{1,3}^{pe}$ and $B_{1,3}^{pe}$ are given in Table II for different wavelengths. We present below the photoelastic-contribution coefficients $A_{1,3}^{pe}$ and $B_{1,3}^{pe}$ and the temperature dependences of the ordinary and extraordinary refractive indices at the wavelength 0.63 μm:

$$A_1^{pe}=-\tfrac{1}{2}n_o^3(p_{11}+p_{12})=-0.46, \quad B_1^{pe}=-\tfrac{1}{2}n_o^3p_{13}=-0.31,$$
$$A_3^{pe}=-\tfrac{1}{2}n_e^3\cdot 2p_{31}=-0.40, \quad B_3^{pe}=-\tfrac{1}{2}n_e^3p_{33}=-0.11, \qquad (11)$$

as well as the thermo-optical constants $A_{1,3}^{to}$ and $B_{1,3}^{to}$, defined as the differences of $A_{1,3}$ and $B_{1,3}$ from the corresponding photoelastic coefficients:

$$A_1^{to}=-0.57, \quad B_1^{to}=+1.10, \quad A_3^{to}=-0.66, \quad B_3^{to}=+0.77 \qquad (12)$$

From a comparison of the values and signs of these coefficients we see that the principal role in the temperature dependence of the refractive index of MnF_2 is played by the thermo-optical mechanism, which in fact determines the positive slope of the $n_{o,e}(T)$ plot at high temperatures. If the photoelastic contribution were to predominate, the slope would be negative.

The validity of the approximating formula (10) was confirmed by us by measurements made on magnetically disordered cubic garnets and on tetragonal zinc di-

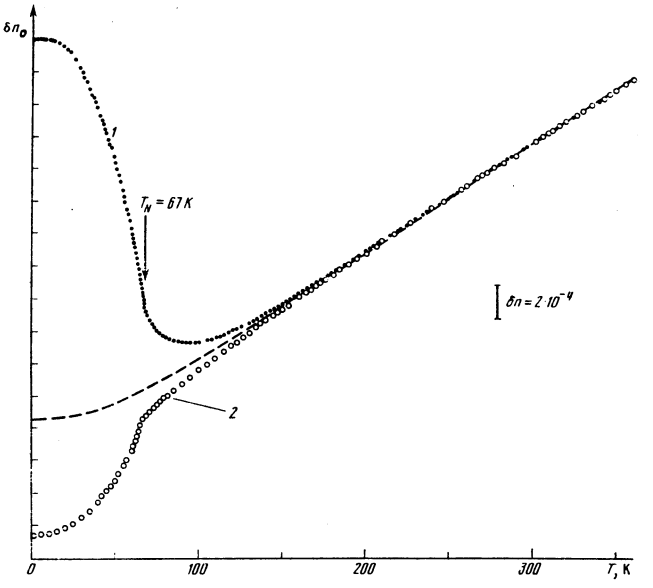

FIG. 5. Temperature dependences of ordinary refractive index of MnF_2 0.63 μm: 1—experiment, 2—obtained from Eq. (10), where the Gibbons data[12], with magnetostriction taken into account, were used for the thermal expansion: $\delta n_o^p = -1.03(\delta l/l)_1 + 0.79(\delta l/l)_3$; the dashed line corresponds to the Debye approximation of $\delta n_o^0(T)$.

phosphide, in which, just as in MnF_2, the decisive role in the thermal behavior of the refractive index is played by the thermo-optical effect.

To calculate by means of Eq. (10) the temperature dependence of that part of the refractive index which is not connected with the magnetic ordering, it is necessary to exclude the magnetostriction from the thermal expansion of MnF_2. The corresponding calculation was made in the Debye approximation, and the theoretical curve was fitted to the experimental one at high temperatures by using as parameters the scale coefficient and the Debye temperature T_D. This curve is shown dashed in Fig. 5. Better agreement in the high-temperature region between the calculated and experimental relations is observed at $T_D \approx 150$ K, which is lower than the Debye temperature ≈260 K, determined in Ref. 14 for acoustics phonons. This indicates that the actual picture of the phonon spectrum in MnF_2 is poorly represented by the simplest Debye approximation. Nonetheless, the scatter of the Debye curves at $T=0$ K for $T_D=150-450$ K amounts to only 2.5×10^{-4}, so that the dashed curve of Fig. 5 can be assumed to be the approximate plot of $\delta n_o^0(T)$ at the temperature and used to estimate the total magnetic contribution to the refractive index.

5. ISOTROPIC MAGNETIC CONTRIBUTION TO THE REFRACTIVE INDEX

We have shown in the preceding section that those changes of the refractive index which are not connected with the magnetic ordering can be approximately described within the framework of the Debye approximation. Then the difference between this calculated curve and the experimental plot of δn_o constitutes the total magnetic contribution to the refractive index, which is microscopically connected with the isotropic exchange interaction.[2] The exchange influences the electron– phonon interaction (which should lead to a change in the thermo-optical contribution) and the thermal expansion of the crystal (the photoelastic contribution due to exchange magnetostriction), as well as directly on the structure of the electron bands (the magneto-optical contribution). Thus, the total magnetic contribution to the refraction can be described by the following expression:

$$\delta n_o^{m}=-\frac{n_o^3}{2}\left[\int_{300}^{T}\mu_1^{m}dT+(p_{11}+p_{12})\left(\frac{\delta l}{l}\right)_1^{ms}+p_{13}\left(\frac{\delta l}{l}\right)_3^{ms}\right]+\delta n_o^{mo}, \quad (13)$$

where μ_1^m is the magnetic increment to the thermo-optical effect tensor (the magneto– thermo-optical contribution), $(\delta l/l)_1^{ms}$ and $(\delta l/l)_3^{ms}$ are the magnetostriction changes of the crystal dimensions, and δn_o^{mo} is the magneto-optical contribution to the refraction.

The magnetic contributions to the thermo-optical and photoelastic effects (i.e., the terms in the square brackets of [13]) can be found as the difference between the Debye curve and the δn_o^{ex}, curve calculated from the approximating formula (10) using data[12] on the thermal expansion of MnF_2 (see Fig. 5). We see that these contributions differ in sign from the total magnetic contribution to the refraction, and consequently differ in sign from the magneto-optical contribution $\delta n_0^{mo}=\delta n_o^{exp}-\delta n_0^{ex}$.

We have already noted above that according to the data of Ref. 16 the photoelastic constants hardly change with temperature and that this circumstance makes it also possible to separate the magnetostriction contribution to the refraction. Estimating approximately the magnetostriction from the data of Ref. 12 at $T=0$ K as $\delta l/l)_3^{ms}\approx-7\times10^{-4}$ and $(\delta l/l)_1^{ms}\approx+4\times10^{-4}$, and using the photoelastic coefficients (11), we find that

$$\delta n_o^{ms}\approx+0.3\cdot10^{-4}, \quad \delta n_e^{ms}\approx-0.8\cdot10^{-4}$$

at the wavelength 0.63 m. For the magnetic contribution to the thermo-optical effect we now get

$$\delta n_o^{mto}\approx\delta n_e^{mto}\approx-10\cdot10^{-4}.$$

At the same temperature and wavelength, the magneto-optical contribution amounts to

$$\delta n_o^{mo}\approx3\cdot10^{-3}, \quad \delta n_e^{mo}\approx1.6\cdot10^{-3}.$$

The contributions at other wavelengths are given in Table II.

We now discuss the temperature changes of the magnetic contribution to the refractive index. We confine ourselves to the magneto-optical contribution, since it is the largest and its separation does not involve the approximations of the Debye theory. Figure 6 shows the temperature dependence of the magneto-optical contribution δn_o^{mo} for the ordinary refractive index at the wavelength 0.63 μm. The magnitude of this contribution does not depend on the polarization of the light in the basal plane, and Borovik-Romanov *et al.*[15] have also shown that it does not change when the antiferromagnetism vector deviates from the tetragonal axis in an external magnetic field. The isotropic magneto-optical refraction can in turn be subdivided into terms due to the long- and short-range magnetic orders:

$$\delta n_o^{mo}=\lambda_1 L^2+\delta n_o^{fl},$$

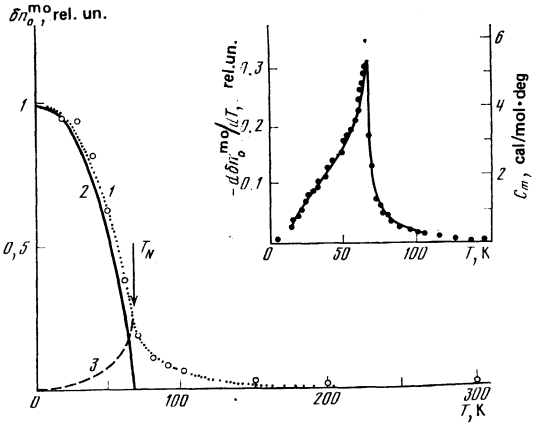

FIG. 6. Temperature dependences: 1—of the magneto-optical contribution to the ordinary refractive index at 0.63 μm, and 2—of the square of the static magnetization of the sublattices; curve 3 corresponds to the contribution of the magnetic fluctuations to the refractive index below T_N; ○—theoretical dependence of the first moment of the two-magnon light-scattering band in MnF_2 on the temperature. Inset: ●—temperature dependence of the derivative $d(\delta n_o^{mo})/dT$ of the magneto-optical contribution with respect to temperature; solid curve—temperature dependence of the magnetic heat capacity.

where λ_1 is a phenomenological parameter. The part connected with the long-range magnetic order is proportional to the square of the antiferromagnetism vector L^2 and vanishes at the Neel point (curve 2 in Fig. 6). The fluctuating part, which is connected with the short-range magnetic order, has its maximum at the point T_N and decreases with increasing distance from T_N in both the low-temperature and high-temperature directions (curve 3 of Fig. 6). The presence of a large fluctuating part in a wide temperature interval is a very important feature of isotropic magnetic refraction. Contribution of the long- and short-range magnetic orders manifests itself also in the magnetostriction and thermo-optical contributions.

The presence of short-range order in the crystals is reflected also in other physical phenomena, for example in the heat capacity and in light and neutron scattering. The inset of Fig. 6 shows the relative changes of the magnetic heat capacity C_m (Refs. 21, 22) and the derivative $d(\delta n_o^{mo})/dT$ of the isotropic magneto-optical refraction with respect to temperature. These two relations correlate very well in the entire temperature interval. A reduction of the data on the magneto-optical refraction near T_N shows that, just as in the case of the magnetic heat capacity,[22] the changes of the refraction can be characterized by a power-law with an exponent ≈ 0.1. However, the insufficient accuracy of the measurements of the temperature near T_N did not permit us to make an unequivocal choice between the power-law and logarithmic dependences of the magneto-optical refraction on the temperature near T_N.

We have also reduced the experimental data using an equation proposed in a theoretical paper by Peĭsakhovich[23] for the fluctuating part of the magnetic birefringence. When modifying the form of the final equation (10) of Ref. 23 and replacing the birefringence in it by the fluctuating contribution to the magneto-optical refraction δn_o^{fl}, we can write down the relation

$$\lg\left|\frac{\delta n_o^{fl}(T)}{\delta n_o^{fl}(T_N)-\delta n_o^{fl}(T)}\right| = \lg C - \mu \lg \tau, \qquad (14)$$

where C is a parameter that depends on the band structure of the crystal and μ is the critical exponent of the correlation radius. It turned out that in a temperature interval of approximately ±15 deg near T_N the experimental points satisfy this relation, but the values obtained for critical exponent above and below T_N were too high, $\mu \approx 1.0$.

The short-range magnetic order manifests itself well also in two-magnon light scattering.[1] An examination of the published data on two-magnon scattering in MnF_2 has shown that neither the relative changes of the intensity nor the relative shift of the maximum of the asymmetrical band of the two-magnon scattering[24] agree with the relative change of the magneto-optical refraction. It turned out, however, that the changes of this refraction correlate splendidly with the relative changes of the first moment of the two-magnon scattering band,[24] which reflects the position of the center of gravity of this band (circles in Fig. 6). Brya, Richards, and Bartkowski[24] also estimated theoretically the relative value of the first moment at $T = T_N$ and obtained a value 0.23 as against the experimental 0.15. In our experiments we obtained for the relative isotropic magneto-optic refraction a value 0.23 which agrees with the indicated theoretical estimate. It appears that measurements of the changes of the refractive index can result in the present case in a better accuracy (approximately 1%) than the light-scattering method, in which one encounters the serious problem of determining the first moment of the two-magnon band, particularly at higher temperatures, where this band is substantially broadened and the background changes.

Whereas the temperature changes of the isotropic magnetic refraction correlate well with the changes of the heat capacity and of the first moment of the two-magnon band, substantial discrepancies are observed between these changes and the data on the critical scattering on neutrons.[25] It is obvious, that different aspects of magnetic interactions in crystals manifest themselves in experiments on neutron scattering and in experiments on magnetic refraction, since light has a much smaller wave vector than neutrons and a much higher energy. In addition, the refractive index is a macroscopic quantity and its measurement is accompanied by averaging of fluctuations of the magnetic order over the frequency, over the volume of the crystal, etc.

6. CONCLUSION

Thus, our investigations have shown that the temperature changes of the principal values of the refractive index in MnF_2 are determined by different mechanisms. An analysis of these changes at high temperatures has made it possible to conclude that they are connected with the thermo-optical and photo-elastic effects. We succeeded also in showing that to approximate the thermo-optical and photoelastic changes at low temperatures one can use Eq. (10), although the thermo-optical effect is in fact not connected directly with the linear expansion coefficient. To analyze the expected changes of the linear-expansion coefficients in the absence of magnetic ordering, we used the Debye approximation, which enabled us to find the total magnetic contribution to the refraction and then resolve this contribution into magneto-optical, magneto-striction, and magneto–thermo-optical components. It turned out that the first component is the largest and is more than ten times larger than the magnetostriction contribution.

On the basis of the measurements of the refractive index at three wavelengths, assuming the single-oscillator approximation to be valid, we were able to show that the measured magnetic refraction is due to changes in both the frequency and in the effective-oscillator strength.

A characteristic of the magnetic contribution to refraction is its connection with both the long-range and the short-range magnetic order in the crystal. The short-range magnetic order influences the refraction below and above the magnetic-ordering temperature T_N and is preserved up to temperatures $(2.5-3)T_N$. At the present time we still have no consistent theory capable of calculating the values of the different magnetic con-

tributions to the refraction, particularly the magneto-optical contribution, and of describing the temperature changes of these contributions, including the immediate vicinity of T_N. We have therefore confined ourselves here to a comparison of the magneto-optical contribution to the refraction with the magnetic heat capacity and with two-magnon scattering (with the first moment of the two-magnon band), and established very good correlation between the corresponding quantities. Taking into account the high accuracy of the measurements of the refractive index and of its changes, one can hope that they can provide in the future a very accurate method of measuring the magnetic correlation functions that determine the magnetic contribution to the refraction.

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[1]V. V. Eremenko, Vvedenie v opticheskuyu spektroskopiyu magnetikov (Introduction to Optical Spectroscopy of Magnets), Naukova dumka, Kiev, 1975.
[2]G. A. Smolenskii, R. V. Pisarev, and I. G. Sinii, Usp. Fiz. Nauk 116, 231 (1975) [Sov. Phys. Usp. 18, 410 (1975)].
[3]G. Heygster and W. Kleeman, Physica (Utrecht) 89B+C, 165 (1977).
[4]I. R. Jahn, J. B. Merkel, G. A. Gehring, and P. J. Becker, Physica (Utrecht) 89B+C, 177 (1977).
[5]K. Iio, M. Sakatani, and K. Nagata, J. Phys. Soc. Jpn. 45, 1567 (1978).
[6]H. Von Kanel, Preprint (to be published in Physica B).
[7]J. F. Nye, Physical Properties of Crystals, Oxford, 1957. Russ. transl. Mir, 1967, p. 300.
[8]P. A. Markovin, R. V. Pisarev, G. A. Smolensky, and P. P. Syrnikov, Solid State Commun. 19, 185 (1976).
[9]G. A. Smolensky, R. V. Pisarev, P. A. Markovin, and B. B. Krichevzov, Physica (Utrecht) 86–88B, 1205 (1977).
[10]B. B. Kruchevtsov, P. A. Markovin, and R. V. Pisarev, Proc. 20th All-Union Conf. on Low-Temperature Physics, part II. M., 1979, p. 165.
[11]I. R. Jahn, Phys. Status Solidi B 57, 681 (1973).
[12]D. F. Gibbons, Phys. Rev. 115, 1194 (1959).
[13]A. A. Kaplyanskiĭ and L. G. Suslina, Fiz. Tverd. Tela (Leningrad) 7, 2327 (1965) [Sov. Phys. Solid State 7, 1881 (1966)].
[14]R. L. Melcher, Phys. Rev. B 2, 733 (1970).
[15]A. S. Borovik-Romanov, N. M. Kreĭnes, A. A. Pankov, and M. A. Talalev, Zh. Eksp. Teor. Fiz. 64, 1762 (1973) [Sov. Phys. JETP 37, 890 (1973)].
[16]A. S. Borovik-Romanov, N. M. Kreĭnes, and J. Paces, Zh. Eksp. Teor. Fiz. 77, 2477 (1979) [Sov. Phys. JETP 50, 1198 (1979)].
[17]A. S. Sonin and A. S. Vasilevskaya, Elektroopticheskie kristally (Electro-optical Crystals), Atomizdat, Moscow, 1971, p. 156.
[18]R. W. Keyes, Solid State Phys. 11, 179 (1960).
[19]B. Di Bartolo, Optical Interactions in Solids, John Wiley and Sons, Inc., N.Y., 1968.
[20]S. I. Novikova, Teplovoe rasshirenie tverdykh tel (Thermal Expansion of Solids), Nauka, Moscow, 1974.
[21]J. W. Stout and E. Catalono, J. Chem. Phys. 23, 2013 (1955).
[22]D. T. Teaney, Phys. Rev. Lett. 14, 898 (1965).
[23]Yu. G. Peĭsakhovich, Pis'ma Zh. Eksp. Teor. Fiz. 22, 506 (1975) [JETP Lett. 22, 247 (1975)].
[24]W. J. Brya, P. H. Richards, and R. R. Bartkowski, Phys. Rev. Lett. 28, 826 (1972).
[25]M. P. Schulhof, P. Heller, R. Nathans, and A. Linz, Phys. Rev. B 1, 2304 (1970).

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