Experimental observation of a new nonreciprocal magneto-optical effect

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Results are presented of an experimental investigation of nonreciprocal linear birefringence (difference between the phase velocities of opposing waves of equal linear polarization) in a lithium-iodate ($LiIO_3$) placed in a magnetic field. The effect is due to the spatial dispersion and is possible in a medium without a symmetry center.

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It is known that allowance for the spatial dispersion in crystals leads to a number of qualitatively new optical phenomena.[1] These include nonreciprocal linear birefringence (the difference between the phase velocities of opposing waves of equal linear polarization). It was noted in[2–4] that a similar effect can be obtained in magnetically ordered crystals, but this effect, in view of its smallness, was not observed in experiment heretofore.

One of us[5] has shown that nonreciprocal linear birefringence is produced also in noncentrosymmetric crystals when an external magnetic field is applied. According to estimates, the value of the nonreciprocal phase shift, even at relatively low intensities of the applied magnetic field, is fully sufficient for measurement if the investigated crystal is placed in the resonator of a ring laser; in this case the nonreciprocal phase shift influences the frequency difference between the opposing waves of the laser.

We present here the results of an experimental investigation of the effect of nonreciprocal linear birefringence in a lithium iodate ($LiIO_3$) crystal placed in a magnetic field. The sample was cut from the crystal in such a way that the laser beam propagated perpendicular to the optical axis with the polarization of the ordinary ray (the entrance faces were cut at the Brewster angle).

We used in the experiment an He—Ne ring laser with perimeter $L = 100$ cm, capable of operating (by changing mirrors) at wavelengths $\lambda = 1.15$ and $0.63\ \mu$. To eliminate the effect of frequency locking of the transverse waves, the laser was mounted on a rotating platform; this produced an initial frequency on the order of 100 kHz. The measurements were performed by a modulation procedure similar to that described in[6]. An alternating magnetic field (of frequency 80 Hz and amplitude on the order of several dozen oersteds) was applied to the crystal, perpendicular both to the laser beam direction and to the optical axis. We measured the opposing-wave frequency-difference deviation due to the nonreciprocal magneto-optical effect. The value of this deviation is $\Delta f = \Delta n d c/\lambda L$, where Δn is the nonreciprocal birefringence, d is the sample length, and c is the speed of light.

The values obtained for the effect in an $LiIO_3$ crystal ($d = 1.8$ cm) where $\Delta n/H = (1.3 \pm 0.2) \times 10^{-12}$ at $\lambda = 1.15\ \mu$ and $\Delta n/H = (1.9 \pm 0.3) \times 10^{-12}$ Oe^{-1} at $\lambda = 1.63\ \mu$. Since $LiIO_3$ is a polar crystal, the sign of the observed effect changes when the direction of the optical axis is reversed, as was indeed confirmed by experiment. It was also verified that this effect does not appear if the magnetic field is directed parallel to the optical axis.

To describe the magneto-optical effects we used the expansion of the dielectric tensor in the following form[1]:

$$\epsilon_{ij}(\omega, \mathbf{k}, \mathbf{H}) = \epsilon_{ij}(\omega) + \gamma_{ijk} H_k + \gamma_{ijkl} H_k k_l(\omega), \tag{1}$$

where **k** is the wave vector and **H** is the vector of the constant magnetic field. The first term of the expansion (1) is responsible for the optical effects in the crystal without allowance for spatial dispersion and external magnetic fields (only nonmagnetic crystals are considered). For the nonreciprocal effects to appear, it is necessary to satisfy the condition

$$\epsilon_{ij}(\omega, \mathbf{k}, \mathbf{H}) \neq \epsilon_{ji}(\omega, -\mathbf{k}, \mathbf{H}).$$

This condition is satisfied by the dielectric-constant increments connected with the tensors γ_{ijk} and γ_{ijkl}, since the Onsager relation[1] imposes on these increments constraints of the type

$$\gamma_{ijk} = -\gamma_{jik}; \qquad \gamma_{ijkl} = \gamma_{jikl}.$$

In addition, in the frequency region where there is no absorption, the γ_{ijk} are imaginary and γ_{ijkl} are real. The tensor γ_{ijk} is responsible for the traditional nonreciprocal magneto-optical effects (the Faraday effect and others), and will henceforth not be considered. As seen from the expansion (1), the tensor γ_{ijkl} is of the axial type. This means that the discussed effect can be observed only in media without a symmetry center. The form of the fourth-rank axial tensor is given for different crystal classes in[7]. The tensor γ_{ijkl} is symmetrical in the first two indices and appears, depending essentially on the type of the anisotropy, along a preferred direction of light propagation. It can be shown that the effect connected with the tensor γ_{ijkl} becomes very small if this anisotropy is circular in character. This circumstance hinders the observation of the effect, against the background of the Faraday effect, in optically active cubic crystals as well as in isotropic media. The most favorable situation occurs when large natural birefringence takes place along the propagation direction of the light, when analysis shows that the Faraday effect makes a small contribution to the nonreciprocal phase shift. The Faraday effect can be separated to an even greater degree by choosing the transverse variant of the magneto-optical effect and directing the light perpendicular to the optical axis of a uniaxial crystal. In this case the induced nonreciprocal linear birefringence is superimposed on the natural linear birefringence. As a result, different phase shifts will occur for the opposing directions for both the ordinary and the extraordinary waves. This was precisely the configuration chosen to observe the nonreciprocal magneto-optical birefringence in the lithium iodate crystal, whose symmetry corresponds to C_6. A more detailed theoretical analysis of the new nonreciprocal magneto-optical effect will be presented in forthcoming publications.

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