Observation of Magnetoelectric Directional Anisotropy

G. L. J. A. Rikken,[1,2] C. Strohm,[1] and P. Wyder[1]

[1]Grenoble High Magnetic Field Laboratory, MPI-FKF/CNRS, 25 Avenue des Martyrs, F-38042 Grenoble, France

[2]Laboratoire National des Champs Magnétiques Pulsés, CNRS/INSA/UPS, 143 Avenue de Rangueil, F-31432 Toulouse, France

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We report the first observation of a new optical phenomenon, magnetoelectric directional anisotropy (MEA). MEA is a polarization-independent anisotropy which occurs in crossed electric field **E** and magnetic field **B** perpendicular to the wave vector **k** of the light. It is described by a contribution to the refractive index of the form $\delta n = \gamma \mathbf{k} \cdot \mathbf{E} \times \mathbf{B}$. Our experiment was performed on a $Er_{1.5}Y_{1.5}Al_5O_{12}$ crystal, but MEA should exist in all media. The relation of this new effect with recently discovered magnetoelectric birefringence is discussed.

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Recent experiments have shown that the combined action of static electric and magnetic fields perpendicular to the propagation direction of light can invoke new optical phenomena. For the case of parallel **E** and **B**, we have observed magnetoelectric Jones birefringence [1], a difference in refractive index for light linearly polarized under +45° and −45° with the external fields, which is bilinear in E and B. For the case of $\mathbf{E} \perp \mathbf{B}$, we have observed magnetoelectric linear birefringence [2], a bilinear difference in refractive index for light linearly polarized parallel to **E** and parallel to **B**. Symmetry arguments have shown that these two effects must have the same magnitude [3], and this was indeed observed in the experiments. The corresponding dichroisms, whose existence follows from that of the birefringences through the Kramers-Kronig relations, have not been observed so far. Calculations have also confirmed the existence of these two magnetoelectric birefringences and their interrelation for the quantum vacuum [4].

In addition to these birefringences other magnetoelectric optical effects have been predicted [5,6]. In particular, a polarization-independent difference in refractive index for light propagating parallel and antiparallel to $\mathbf{E} \times \mathbf{B}$ was predicted to exist for all media on the basis of symmetry arguments, i.e., the refractive index has a contribution of the form $\delta n = \gamma \hat{\mathbf{k}} \cdot \mathbf{E} \times \mathbf{B}$, where $\hat{\mathbf{k}}$ is the direction of propagation. The experimental observation of this phenomenon is the subject of this Letter.

The existence of this optical magnetoelectrical directional anisotropy can be understood in an elegant way by a relativistic argument: The electric and magnetic fields that are perpendicular to each other in the laboratory rest frame and perpendicular to the direction of light propagation transform to a single transverse magnetic field $\mathbf{B}'$ in the reference frame that moves along the light propagation direction with velocity $\mathbf{v} = c\mathbf{B} \times \mathbf{E}/B^2$, where $B' = B\sqrt{1 + \frac{v^2}{c^2}}$ [7] (Lorentz-Heaviside units, $E < B$). In this moving reference frame, the change of the refractive index under the action of B' is described by the Cotton-Mouton effect so $n'_{\|} \approx n'_0 + a_{\|}B'^2$ and $n'_{\perp} \approx n'_0 + a_{\perp}B'^2$. These refractive index values can be transformed back to the laboratory frame values by using the well-known result of the electrodynamics of moving media, as, e.g., used to describe Fizeau's experiment [7]

$$n_i^{-1} = n_i'^{-1} + \frac{\mathbf{v} \cdot \hat{\mathbf{k}}}{c}\left(1 - \frac{1}{n_i'^2} + \frac{\omega}{n_i'}\frac{\partial n_i'}{\partial \omega}\right), \quad (1)$$

with $i = \|, \perp$. When we neglect the dispersion term, Eq. (1) gives

$$n_i(\hat{\mathbf{k}}, \mathbf{B}, \mathbf{E}) \approx n_0 + a_i(B^2 + E^2 + 2\mathbf{B} \times \mathbf{E} \cdot \hat{\mathbf{k}}). \quad (2)$$

So we find both linear birefringence $\Delta n_{\mathrm{LB}} \equiv n_{\|} - n_{\perp} = (B^2 + E^2 + 2EB)(a_{\|} - a_{\perp})$, with Cotton-Mouton, Kerr, and magnetoelectric contributions, respectively, and a polarization-independent magnetoelectric anisotropy $\Delta n_{\mathrm{MEA}} \equiv n_k - n_{-k} = 2(a_{\|} + a_{\perp})EB$. Two things can be learned from this argument: First, both the magnetoelectric directional anisotropy and the magnetoelectric linear birefringence can be regarded as relativistic corrections to the Cotton-Mouton or Kerr effects. Second, the order of magnitude of magnetoelectric linear birefringence and of magnetoelectric anisotropy are similar. Based on experimental results for the former, this implies that $\Delta n_{\mathrm{MEA}}/EB$ of the order of magnitude of 10^{-16} $\mathrm{mV^{-1}\,T^{-1}}$ can be expected in transparent molecular liquids [1]. Such small values call for the use of a ring laser measurement, as suggested by Ross *et al.* [6] and recently implemented by Vallet *et al.* for the observation of another weak polarization-independent optical anisotropy, namely, magnetochiral birefringence [8]. When further considering the analogy with the magnetochiral case, the observation of strong relative magnetochiral directional anisotropy in emission and absorption in rare-earth and transition metal ions [9,10] suggests that also the search for magnetoelectric anisotropy in rare-earth doped media could be successful. The strong electronic Cotton-Mouton effects that have been observed in such media [11,12] would, according to the argument above, correspond to $\Delta n_{\mathrm{MEA}}/EB \approx 10^{-15}$ $\mathrm{mV^{-1}\,T^{-1}}$. In particular those optical transitions that show strong magneto-optical Faraday effects and hypersensitivity to (crystal) electric fields would seem suitable candidates to study magneto-electrical anisotropy in absorption or emission, which corresponds to the imaginary part of Δn_{MEA}. It is along these lines that we have attempted to observe MEA.

The experimental setup used to observe MEA in optical absorption is shown in Fig. 1. As MEA depends only on the relative orientation of $\hat{\mathbf{k}}$, $\mathbf{B}$, and $\mathbf{E}$, the directional anisotropy can be observed by the change in optical absorption when changing the sign of one of these three parameters. For experimental convenience, we have chosen to study the absorption change upon changing the direction of $\mathbf{E}$. The transmission of light from a tunable Ti:sapphire laser through the sample is measured with a photodiode. The sample consists of an $Er_{1.5}Y_{1.5}Al_5O_{12}$ (ErYAG) crystal. This material was chosen because it is cubic, i.e., optically isotropic to first order and shows narrow and relatively strong transitions in the tuning range of the Ti:sapphire laser. The sample studied had dimensions of $1 \times 2 \times 5$ mm^3, with electrodes on the two opposite 2×5 mm^2 faces, the magnetic field aligned along the long axis, and the direction of light propagation perpendicular to the electric and magnetic field. The electric field is alternated at 1.4 kHz, and phase sensitive detection on the transmitted light is performed by means of a lock-in amplifier at the fundamental frequency. The polarization state of the incident light can be chosen to be either fully unpolarized by means of a Hanle depolarizer, or linearly polarized under an angle ϕ with the magnetic field by means of a Fresnel rhomb. By normalizing the lockin output signal by the dc output of the photodiode, we obtain $\Delta\alpha_{\mathrm{MEA}} l$, the magnetoelectric anisotropy in extinction, l being the optical path length. Figure 2 shows $\Delta\alpha_{\mathrm{MEA}} l$ as a function of wavelength, together with the ordinary absorption spectrum αl, for two different transitions of the crystal-field split $^4I_{9/2}$ multiplet of the Er^{3+} ion (for a description of these levels, see, e.g., Ref. [13], and references therein). Whereas around 815 nm [Fig. 2(b)], a derivative-like line shape for MEA is observed, the situation around 789 nm (Fig. 2(a)) is much more complicated and a rich spectral behavior is observed. Clearly, MEA may be an interesting spectroscopic tool if the corresponding theoretical modeling can be developed. The observation of MEA in absorption implies through the Kramers-Kronig relations the existence of MEA in refraction.

$\Delta\alpha_{\mathrm{MEA}} l$ at a fixed wavelength as a function of magnetic field and of electric field with unpolarized light is shown in Fig. 3. Clear linear dependences on B and E are

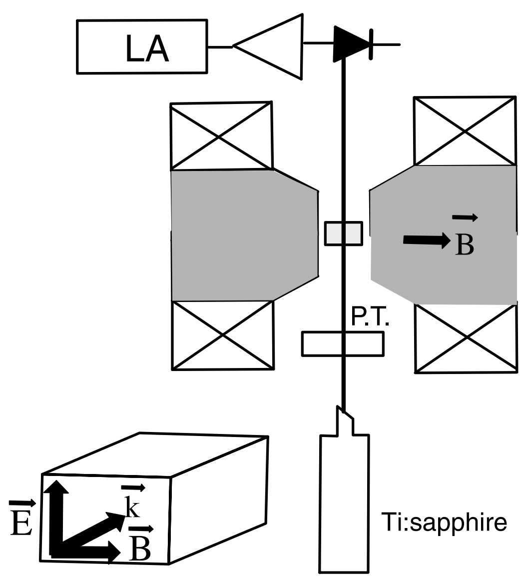

FIG. 1. Schematic setup of the experiment. The linear polarization of the laser output can be rotated (PT = Fresnel rhomb) or scrambled (PT = Hanle depolarizer). The electric field is alternated at 1.4 kHz, and phase sensitive detection of the photodiode signal by the lockin amplifier LA is used to determine MEA. The inset shows the relative orientation of the electric field $\mathbf{E}$, the magnetic field $\mathbf{B}$, and the wave vector of the light $\mathbf{k}$.

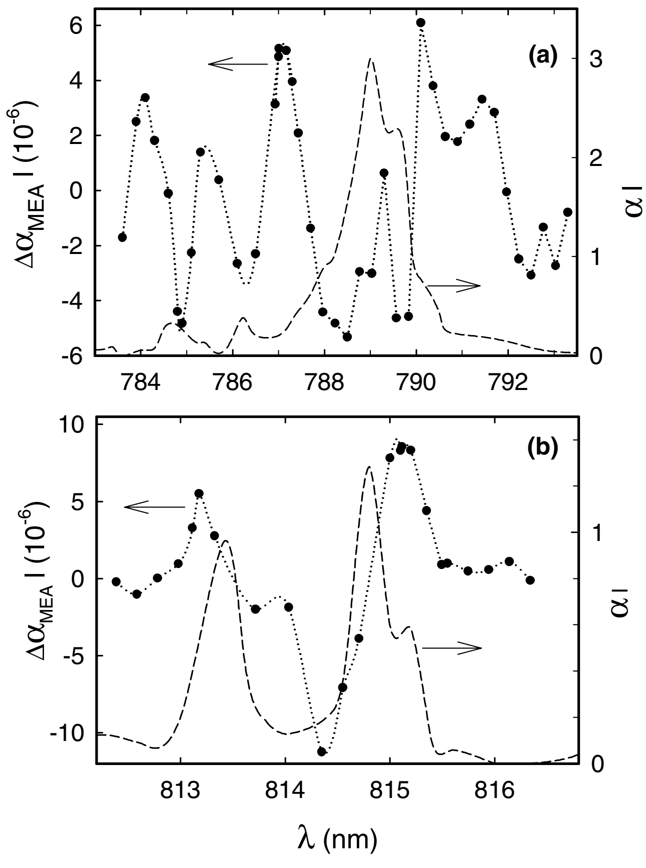

FIG. 2. MEA (full symbols) and absorption spectra (dashed lines) with unpolarized light of an $Er_{1.5}Y_{1.5}Al_5O_{12}$ crystal with $B = -1.85$ T and $E = 236$ V/mm for two different transitions of the crystal-field split $^4I_{9/2}$ multiplet of the Er^{3+} ion. The dotted lines are only meant to guide the eye.

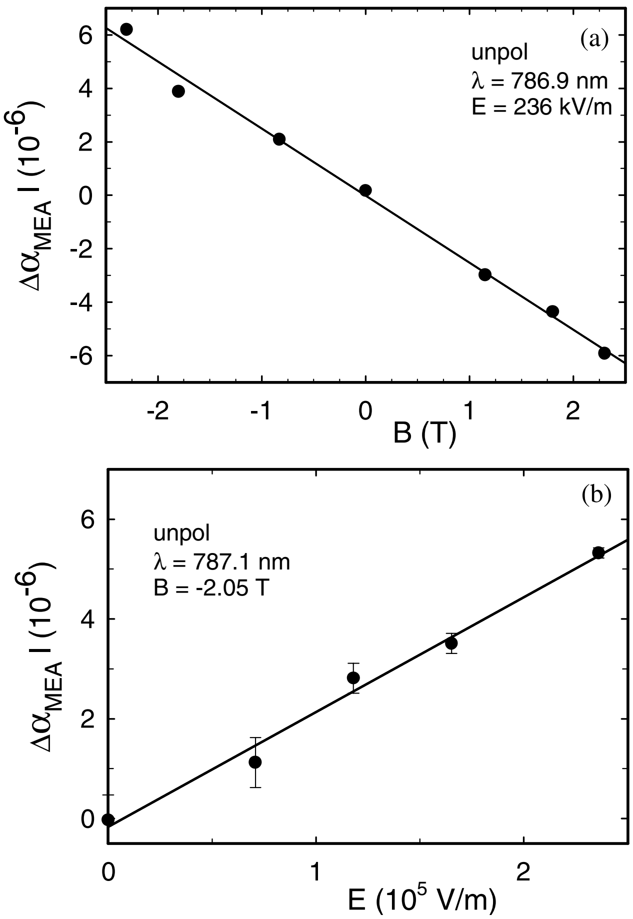

FIG. 3. Dependence of MEA measured with unpolarized light on (a) magnetic field B, with $E = 236$ V/mm and $\lambda =$ 786.9 mm (b) electric field amplitude E with $B = -2.05$ T and $\lambda = 787.1$ nm. Sample is a $Er_{1.5}Y_{1.5}Al_5O_{12}$ crystal, the lines are linear fits to the experimental data.

observed, proving that $\Delta\alpha_{\rm MEA} = \gamma EB$. Figure 4 shows the dependence of $\Delta\alpha_{\rm MEA} l$, measured with linearly polarized light, on the angle ϕ between polarization and magnetic field. Also shown is a fit to an $a + b\cos 2\phi$ dependence, with $a = 6.8 \times 10^{-6}$ and $b = 4.9 \times 10^{-6}$. The a term we identify as the true polarization-independent magnetoelectric anisotropy and it corresponds to the result found with unpolarized light in Fig. 2, whereas the $b\cos 2\phi$ term is attributed to magnetoelectric linear dichroism, the absorptive counterpart of magnetoelectric linear birefringence, which should have such a polarization dependence [2]. As the weak oscillator strength of the optical transition under study implies that $\partial n_i/\partial\omega$ is small, the relativistic argument outlined above should apply. The similar values found for a and b support the validity of this argument. The maximum observed value of $\Delta\alpha_{\rm MEA}$ translates through $\alpha = 4\pi {\rm Im} n/\lambda$ into ${\rm Im}\Delta n_{\rm MEA}/EB \approx 10^{-15}$ mV^{-1} T^{-1}. The agreement of this value with the estimates above, based on experimental results for magnetoelectric linear birefringence

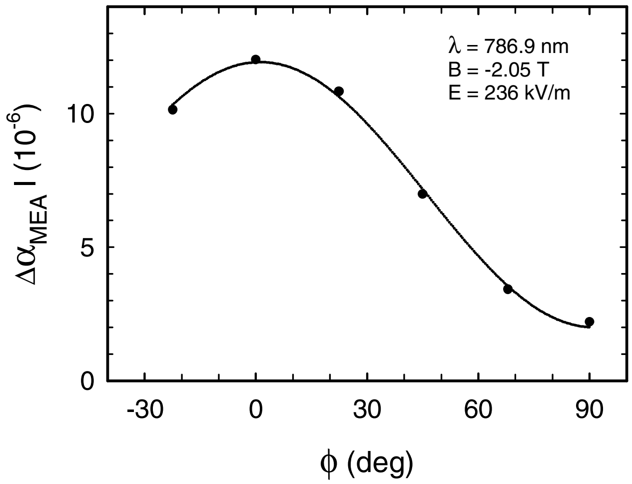

FIG. 4. MEA with linearly polarized light, as a function of the angle ϕ between the polarization and the magnetic field, of an $Er_{1.5}Y_{1.5}Al_5O_{12}$ crystal with $B = -2.05$ T, $E = 236$ V/mm, and $\lambda = 786.9$ nm. The solid line is a fit to an $a + b\cos 2\phi$ dependence, with $a = 6.8 \times 10^{-6}$ and $b = 4.9 \times 10^{-6}$.

in transparent molecular liquids (10^{-16} mV^{-1} T^{-1}) or Cotton-Mouton constants in rare-earth crystals (10^{-15} mV^{-1} T^{-1}), supports our relativistic model qualitatively.

In conclusion, we have reported the first observation of magnetoelectric directional anisotropy, an optical effect predicted to exist for all media. We have also observed magnetoelectric linear dichroism, and the observed magnitudes support our suggestion that these effects are interrelated, and related to the Cotton-Mouton effect through special relativity.

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[1] T. Roth and G. L. J. A. Rikken, Phys. Rev. Lett. **85**, 4478 (2000).
[2] T. Roth and G. L. J. A. Rikken, Phys. Rev. Lett. **88**, 063001 (2002).
[3] E. B. Graham and R. E. Raab, Mol. Phys. **52**, 1241 (1984).
[4] G. L. J. A. Rikken and C. Rizzo, Phys. Rev. A **63**, 012107 (2000).
[5] N. B. Baranova, Yu.V. Bogdemov, and B.Ya. Zel'dovich, Sov. Phys. Usp. **20**, 870 (1977).
[6] H. J. Ross, B. S. Sherborne, and G. E. Stedman, J. Phys. B **22**, 459 (1989).
[7] J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1975).
[8] M. Vallet et al., Phys. Rev. Lett. **87**, 183 003 (2001).

[9] G. L. J. A. Rikken and E. Raupach, Nature (London) **390**, 493 (1997).
[10] G. L. J. A. Rikken and E. Raupach, Phys. Rev. E **58**, 5081 (1998).
[11] J. Ferré and G. A. Gehring, Rep. Prog. Phys. **47**, 513 (1984).
[12] K. Cho, S. P. Bush, D. L. Mazzoni, and C. C. Davis, Phys. Rev. B **43**, 965 (1991).
[13] C. A. Morrison and R. P. Leavitt, in *Handbook on the Physics and Chemistry of Rare Earths*, edited by K. A. Gschneider, Jr. and L. Eyring (North-Holland, Amsterdam, 1982), Vol. 5, p. 461.