Observation of Magnetoelectric Linear Birefringence

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We report the first experimental observation of optical linear birefringence induced in molecular liquids by crossed electric and magnetic fields perpendicular to the direction of light propagation. The optical axes coincide with the external fields, and the strength is bilinear in the electric and magnetic fields.

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In 1875, Kerr found a link between optics and electrostatic fields by his observation of electric field induced linear birefringence in glass [1], followed by a similar observation in liquids [2]. Later a phenomenologically identical effect was observed for magnetic fields by Majorana in colloids [3] and by Cotton and Mouton in liquids [4]. In all cases, the linear birefringence was quadratic in the applied external field strength, and had its optical axis parallel to this field. Intuitively, one could expect a type of cross effect between these two cases, where a linear birefringence is induced by combined magnetic and electric fields. Pockels was the first to address such a situation theoretically. He showed that within the electric dipole approximation such a cross effect is not allowed [5]. In more recent days, magneto-electro-optical effects were given a more solid theoretical base by the work of Kielich [6], Baranova *et al.* [7], Graham and Raab [8], and Ross *et al.* [9]. All authors agree on the existence of a linear birefringence in crossed electric and magnetic fields perpendicular to the direction of light propagation, with the optical axes parallel to the externally applied fields and with a strength bilinear in electric and magnetic fields. All authors also agree, with Pockels, that the necessary light-matter coupling for the occurrence of magnetoelectric linear birefringence goes beyond the electric dipole–electric dipole coupling responsible for both Kerr and Cotton-Mouton effects. There are, however, important discrepancies between the different predictions for the material systems in which such an effect could occur, and its absolute strength. Apparently, magneto-electro-optical effects constitute a severe test for our understanding of light-matter interaction. Here, we report the first experimental observation of the predicted magnetoelectric linear birefringence.

When considering the possible existence of a magnetoelectric linear birefringence, one immediately realizes that, because of the symmetry properties under time and parity reversal, such a birefringence cannot exist in arbitrary media as a simple bilinear function of the magnetic field **B** and the electric field **E**. This led Baranova *et al.* to the conclusion that such an effect can only exist in chiral, dissipative media [7], or, inversely, that the only optical effect strictly bilinear in **B** and **E** would be circular birefringence

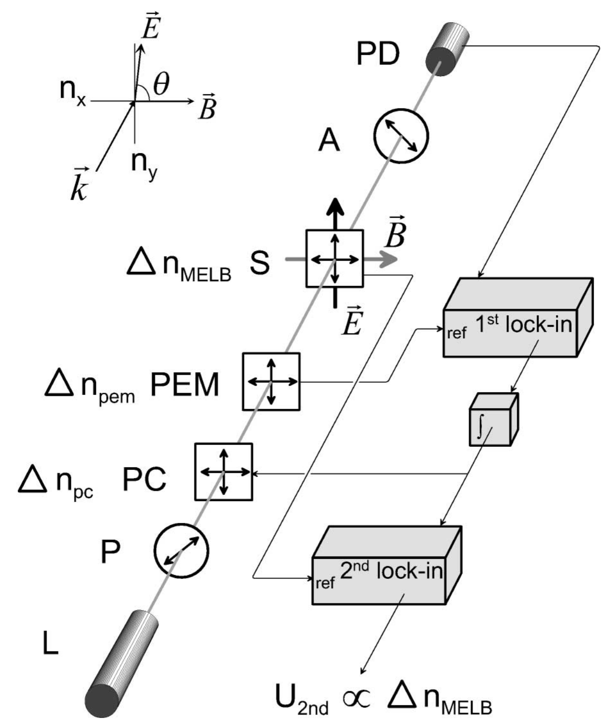

FIG. 1. Experimental geometry and setup. The inset shows a schematic view of MELB with definition of the axes. The main figure shows the experimental setup used to observe MELB, consisting of a HeNe laser at a wavelength of 632.8 nm (L), polarizer (P), photoelastic modulator (PEM), Pockels' cell (PC), sample cell (S), analyzer (A), and photodiode (PD). A phase-sensitive feedback loop drives the Pockels' cell to compensate the sample birefringence. Path lengths in the samples were 5–30 mm. In addition to the static magnetic field **B**, a low frequency alternating electric field $\mathbf{E}\cos\Omega t$ is applied and phase-sensitive detection of the resulting birefringence at the electric field frequency Ω ($\Omega \approx 60\ \mathrm{s}^{-1}$) is performed. The angle θ between **E** and **B** can be chosen by rotating the electrode assembly. By a proper choice of θ and the polarization state of the light, this setup can be used to measure electric linear (Kerr), magnetic linear (Cotton-Mouton), and magnetoelectric linear/Jones birefringence (MELB/MEJB). For the latter case, the resolution was found to be $\Delta n_{\mathrm{ME}} \approx 2 \times 10^{-12}$ with applied magnetic and electric fields up to 17 T and 2.5×10^5 V/m, respectively, and a path length of 25 mm.

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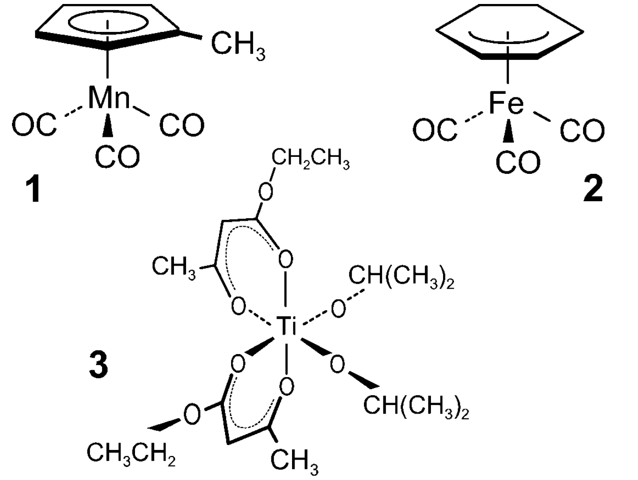

FIG. 2. Structures of the molecules studied. 1: methylcyclopentadienyl-Mn-tricarbonyl; 2: cyclohexadienyl-Fe-tricarbonyl; 3: Ti-bis(ethyl-acetoacetato) diisopropoxide.

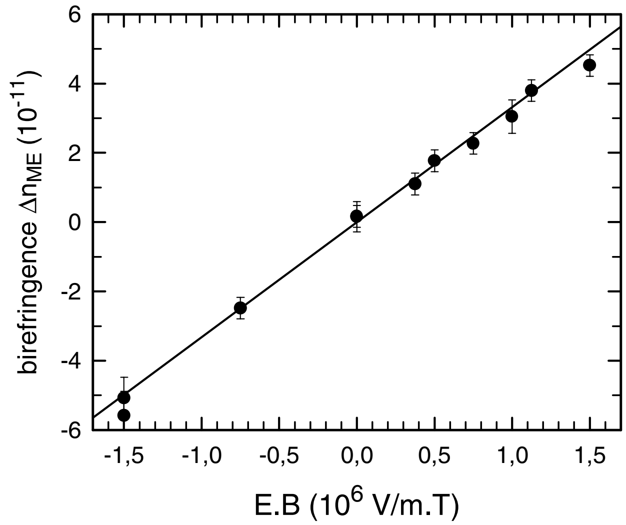

FIG. 3. Magnetoelectric linear birefringence. Δn_{ME} of "1" as function of $\mathbf{E} \cdot \mathbf{B}$, with a linear fit to the data.

in chiral media. The symmetry considerations do, however, allow for a linear birefringence that is trilinear in **k**, **B**, and **E**, where **k** is the wave vector of the light. In such a context, a symmetry allowed expansion of the refractive index of an arbitrary medium for transversely linearly polarized light with polarization vector $\hat{\mathbf{e}}$ may then be written as [9]:

$$n_{\mathrm{ME}}(\mathbf{E}, \mathbf{B}, \hat{\mathbf{k}}, \hat{\mathbf{e}}) = n_0 + \xi EB\{(\hat{e} \times \hat{E} \cdot \hat{k})(\hat{e}^* \cdot \hat{B}) + (\hat{e} \times \hat{B} \cdot \hat{k})(\hat{e}^* \cdot \hat{E}) + \text{c.c.}\}, \tag{1}$$

which in transverse, crossed external fields leads to fast and slow axes parallel to the external fields, and a birefringence

$$\Delta n_{\mathrm{ME}}(\lambda) \equiv n_B - n_E = k_{\mathrm{ME}}(\lambda)\lambda EB\,. \tag{2}$$

We shall refer to this effect as magnetoelectric linear birefringence (MELB). If the external fields are not crossed, but are still perpendicular to the propagation direction, the linear birefringence between the two polarization directions that are parallel and perpendicular to one of the fields becomes

$$\Delta n_{\mathrm{ME}}(\theta, \lambda) \equiv n_{\parallel} - n_{\perp} = k_{\mathrm{ME}}(\lambda)\lambda EB \sin\theta\,, \tag{3}$$

where θ is the angle between the two external fields. For parallel external fields, the MELB vanishes. However, for this case another magnetoelectric birefringence is maximal. This birefringence is called magnetoelectric Jones birefringence (MEJB) [10], and has its fast and slow optical axes tilted over $\pm 45°$ with respect to the external fields. MEJB was recently observed for the first time [11]. Note that MELB and MEJB are, from a macroscopic point of view, fundamentally different; within the Jones formalism, that classifies all possible optical effects in uniaxial media into four distinct classes, MELB and MEJB fall into different classes [12]. Still it will be intuitively clear from a microscopic point of view that a relation between MELB and MEJB should exist. Indeed, symmetry arguments show that the magnitudes of these two effects must be equal [8,9]. The explicit appearance of **k** in Eq. (1) shows that these two magnetoelectric effects are in essence spatial dispersive effects that result from a nonlocal optical response of the medium [13]. In this sense, both magnetoelectric birefringences are related to optical activity (natural circular birefringence), which also results from a nonlocal optical response. However, in the latter case, the nonlocality appears in the optical response because of the chirality of the medium. In contrast, in the theories of Ref. [8,9] it is clear that all molecules should show MELB. In fact, recent calculations have shown that even the vacuum, which is predicted to show both the Cotton-Mouton and the Kerr effect due to quantum-electrodynamical effects, should also show MELB and MEJB [14].

The experimental setup to observe MELB is shown in Fig. 1. It consists of a modification of the setup used to

TABLE I. Electric and magnetic dipole moments and results for MELB, MEJB, Kerr, and Cotton-Mouton effects. Kerr and Cotton-Mouton constants are defined as $\Delta n_K = k_K(\lambda)\lambda \mathbf{E}^2$ and $\Delta n_{\mathrm{CM}} = k_{\mathrm{CM}}(\lambda)\lambda \mathbf{B}^2$.

Molecule	μ(D)	$m(\mu_B)$	$k_{\mathrm{ME}}(\frac{10^{-12}}{\mathrm{VT}})$	$k_{JB}(\frac{10^{-12}}{\mathrm{VT}})$	$k_K(\frac{10^{-15}m}{\mathrm{V}^2})$	$k_{\mathrm{CM}}(\frac{10^{-5}}{m\mathrm{T}^2})$
1	≈4	1.7	51	47	340	−51
2	≈4	≈3	22	21	100	−130
3	≈1	≈0.5	5.3	5.1	32	7

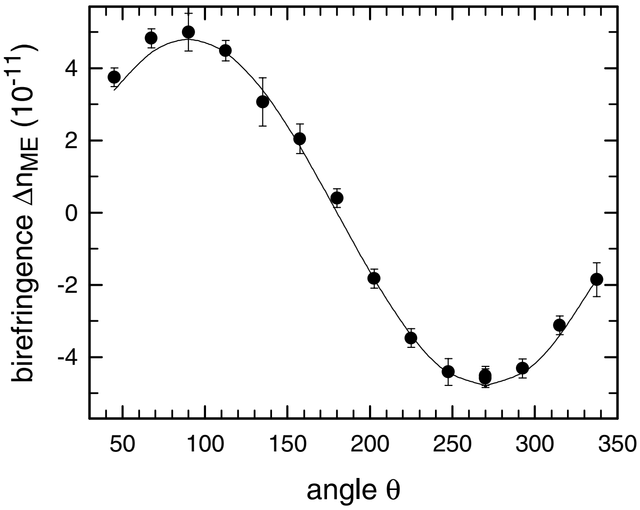

FIG. 4. Angular dependence of the magnetoelectric linear birefringence. Δn_{ME} of "1" as a function of θ. A fit to a $\sin\theta$ dependence is shown. Data were obtained at $B = 14$ T and $E = 1.1 \times 10^5$ V/m.

measure magnetoelectric Jones birefringence [11]. All the samples that have shown MEJB [11] were investigated for the occurrence of MELB. They were pure molecular liquids, shown in Fig. 2 and listed in Table I, and were nonabsorbing at the wavelength used ($\lambda = 632.8$ nm). Our results for electric (Kerr), magnetic (Cotton-Mouton), and magnetoelectric linear birefringence are summarized in Table I. All samples were found to show a significant MELB. The claim by Baranova *et al.* [7] that MELB can only occur in chiral, dissipative liquids is clearly refuted. Typical results for 1 are shown in Fig. 3, where the linear dependence of Δn_{ME} on $\mathbf{E} \cdot \mathbf{B}$ is demonstrated. It was checked that the observed Δn_{ME} was independent of sample length. By rotating the electrode assembly around the light propagation direction at fixed magnetic field and polarization state of the light, we can vary the angle θ. Our results as a function of $\sin\theta$ (Fig. 4) confirm the predictions by Ross *et al.* [9] for the geometric dependence. Figures 3 and 4 together thereby prove the existence of MELB and confirm Eq. (3).

A further test of the validity of our experimental results consisted of comparing the observed MELB of 1, 2, and 3 to the observed MEJB for these compounds [11]. This birefringence, which has its optical axis under $\pm 45°$ with the parallel external fields, was indeed found to have the same magnitude as the MELB within the experimental inaccuracy ($\pm 10\%$), as can be seen in Table I. The discussion in Ref. [11] on the magnitude of the MEJB therefore also applies to the case of MELB, and any quantitative theory that addresses one of the two effects will also automatically cover the other one.

In summary, we report the first experimental observation of magnetoelectric linear birefringence in molecular liquids. We find qualitative agreement with predictions by [8] and [9], but quantitative understanding is still lacking. We confirm that the magnitude of MELB is identical to that of MEJB, as required by symmetry arguments.

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