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Deflection switching of a laser beam by the Pockels effect of water

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Deflection of a laser beam in response to an electric field was detected with a Sagnac interferometer. A laser beam was aligned to travel between two electrodes immersed in aqueous electrolyte solution. When the alternating electric field was applied perpendicular to the beam axis, the direction of the beam deflection was switched synchronously with the field alternation as expected for the Pockels effect. Broken inversion symmetry is prerequisite to the linear electrooptic effect, but surprisingly the effect was observed even when the laser beam travels through the bulk water a few millimeters away from the electrode surface. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4705154>]

The linear electrooptic (EO) effect, Pockels effect, exists only in crystals that do not possess inversion symmetry.¹ Therefore, it is usually ruled out to use bulk liquid, where constituent molecules are randomly oriented to keep macroscopic centrosymmetry, as an EO device. However, the Pockels effect of water within the electric double layer (EDL) at the electrode-solution interface^{2,3} was reported, and the Pockels constants were measured to be one order of magnitude larger than that of typical electrooptic crystals such as LiNbO₃.^{1,4,5} The thickness of the EDL is given by the Debye-Hückel length² and is estimated to be a nanoscale-order. The response time of the effect is an order of 0.001 s (RC time constant) because it is limited by the formation time of the EDL.⁴ A possible mechanism of the Pockels effect of water is the initially induced broken centrosymmetry due to the presence of the electrode surface even before the electric field application. The electrode surface can induce orientational order of water⁶ or interaction of water molecules with the oxygen atoms of the electrode surface.^{3,7,8} Orientational response frequency of liquid water itself is as large as 10 GHz, which is comparable to that of an electrooptic modulator using LiNbO₃, so that the Pockels effect of water is potentially usable for nanoscale EO devices.⁹

The Pockels constant of water in the EDL was previously determined as follows. When a voltage is applied to an electrode in aqueous solution, two kinds of layers are formed at the water-electrode interface. The one is the EDL on the water side, and the other is the space-charge layer (SCL) on the electrode side. The Pockels effect was detected with normally incident beam on the electrode surface, so that the signal contains the contribution not only from the EDL but also from the SCL. The Pockels constant of the EDL was inde-

pendently determined by post-processing the combined signals based on the multiple layer analysis. Judging from the measurement and analyzing methods above, the magnitude of the Pockels constants is reliably determined. However, it was not experimentally proved whether the refractive index change occurs completely within a few nanometer-thick EDL because the probing light is normally incident on the electrode surface, and also it is not clear whether a nanoscale-order EDL is completely formed under the alternating electric field.

In this letter, we detect the Pockels effect of water as a function of the distance from the electrode. With the probe light incident parallel to the electrode surface, electric-field induced deflection of light beam is demonstrated, which is directly applicable to EO switching of light. The phenomenon we observed is schematically depicted in Fig. 1. The laser beam was deflected with its direction switched in synchronized response to the alternating electric field. Since the deflection angle was extremely small, a highly sensitive Sagnac interferometer was utilized for observation of the deflection. Surprisingly, the effect was observed even when the laser beam travels through the bulk water at the distance of a few millimeters from the electrode surface. This is an intriguing result from the textbook point of view of nonlinear optics.¹ The Pockels constant for “bulk water” estimated

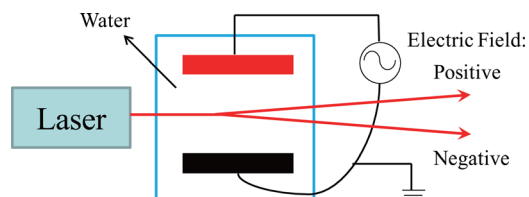


FIG. 1. Deflection switching of a laser beam under alternating electric field in water.

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from the deflection angle is shown to be one order of magnitude smaller than that previously evaluated for the EDL.

Fig. 2 shows the schematic of the experimental setup. The details of the experimental apparatus are described elsewhere.¹⁰ A Sagnac interferometer was used for the refractive index change measurement with a He-Ne Laser (Model 32734, Research Electro-Optics) at 633 nm with 3 mW output power. The laser beam was divided into two with a beam splitter which is the input port as well as the output port of the interferometer. The two beams were delivered into clockwise and counterclockwise propagating arms, which make a common path assuring the stability against fluctuation in the path length difference in the two arms and suited to measure a small imbalance between the two beams induced by a nonlinear effect.^{11,12} They were recombined at the beam splitter to be detected with a photodiode (S1336-BQ8, Hamamatsu Photonics) placed at the dark port. This is because if the two beams are completely superposed in space, they destructively interfere to yield zero intensity due to π phase difference brought in by fixed end reflection at the beam splitter for one of the beams. A measured sample, a glass cell filled with 0.1 M-NaCl aqueous solution, was placed along the common path. It was located at an asymmetrical position in the path such that the light traveling distance from the sample to the photodetector is significantly different between the two split beams. This difference introduced imbalance between the two beams in the common path interferometer to make sensitive signal detection possible as presented below.

In the solution, two electrode plates, tin-doped indium oxide (ITO, Geomatec) and Pt electrodes, were immersed in such a way that they are positioned parallel to each other and to the laser beam transmitted between them. An AC voltage of $2 V_{p-p}$ at frequency $f = 125$ Hz (Ref. 13) was applied with a function generator to the ITO electrode with the Pt electrode grounded. When a voltage is applied, the Pockels effect of water induces a change in the refractive index of water, which is proportional to the electric field. Note that the sign of the refractive index change is inverted if the polarity of the electric field changes. If the applied voltage is static, the EDL, where the applied voltage is concentrated on, is formed completely at the water-electrode interface. For 0.1 M electrolyte solution, the thickness of the layer (Debye-Hückel

length) is estimated to be 2 nm.^{2,4,5} However, as the alternating voltage is applied, the EDL is expected to be only transiently formed to make a less steep voltage gradient in the vicinity of the electrode than in the static case. The resulting electric field gradient induces the gradient in the refractive index change to deflect the transmitted beam. Both counter-propagating beams are deflected at the same deflection angle because they travel the common path between the electrodes, but the arm lengths from the sample to the photodetector are different to induce a lateral displacement in the beam positions on the detector surface. As a result, the effect of destructive interference is partially reduced resulting in the increase in the light intensity at the detector. To be precise, the two beams are slightly displaced initially because the light intensity change is larger for a displaced position than in the completely overlapped position. The light intensity change which is synchronous with the modulation frequency f of the electric field is detected with a lock-in amplifier (Model 7265, Signal Recovery). The beam diameter is about 1.4 mm at the detector, whose size is 5 mm square, and is about $170 \mu\text{m}$ at the electrodes in the cell, focused with a $f = 300$ mm focusing lens.

The experimental result is shown in Fig. 3, where the deflection angle is plotted against the distance of the beams from the ITO electrode surface. The distance was varied by moving a micrometer-controlled translational stage holding the cell. The light intensity change is transformed into the deflection angle by using a numerical simulation which defines the light intensity as a function of the displacement of two beams. The deflection angle is estimated to be about 4×10^{-9} rad. The deflection occurs not only when the beam passes through the close vicinity of the electrodes but also when it passes a few millimeters away from the ITO electrode. Furthermore, it should be noticed that the signal intensity is nearly constant almost over the whole distance between the electrodes, 4 mm. To check this signal is not spurious but specific to the volume between the two electrodes where the electric field is present, it is confirmed that there is no signal either without the electric field or without the laser beams (the beams are blocked). In addition, it is also confirmed that there is no signal outside the electrodes. With all these results taken into account, it is concluded that the observed deflection was induced in bulk water between

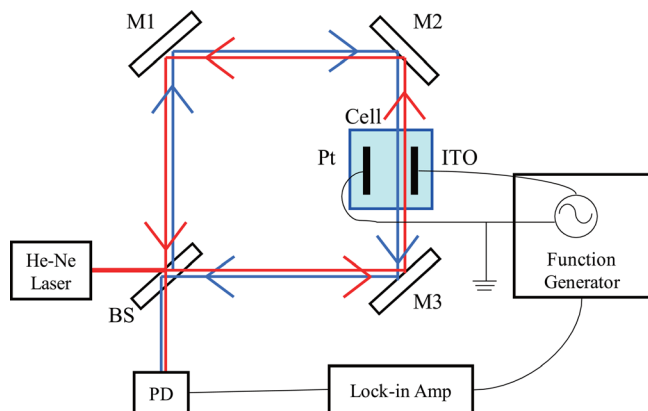


FIG. 2. Experimental setup with a Sagnac interferometer. AC voltage at frequency f is applied between electrodes in aqueous electrolyte solution.

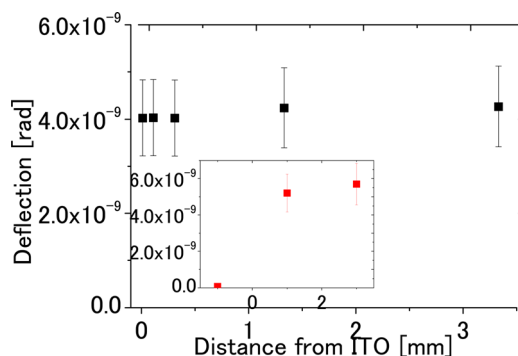


FIG. 3. Dependence of the deflection angle on the spatial-position of the laser beam, measured with lock-in detection at f . The inset shows that there is no signal outside the electrodes.

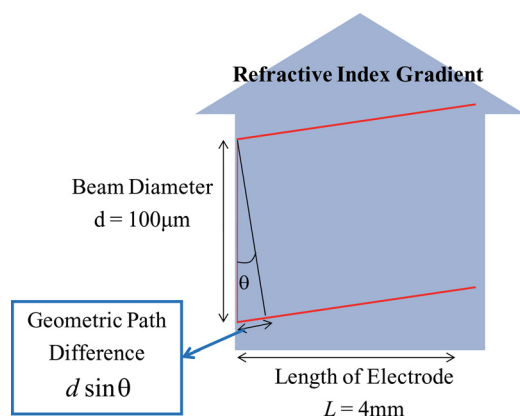


FIG. 4. Laser beam deflection under a gradient of the refractive index close to the electrode.

the two electrodes when the voltage is applied. Because there was no appreciable signal detected at $2f$, the signal is confirmed to be caused by the Pockels effect, not by the Kerr effect. The constant signal intensity over the whole volume between the electrodes, therefore, means the constant gradient of the electric field. In other words, the voltage is quadratically dependent on the distance from the electrode surface.

Since bulk water has macroscopic centrosymmetry, the second-order optical nonlinearity or the first order optoelectric effect (Pockels effect) is inactive.¹ Therefore, the result is intriguing in view of the basics of nonlinear optics. One possible reason presently conceivable is that nonequivalent electrodes, the ITO and Pt electrodes, are used to inevitably introduce some asymmetry into the bulk water volume. Another possible mechanism is that one electrode is grounded and the voltage relative to the ground is applied to the other. That is, the absolute potential is asymmetric relative to the vacuum. For example, the redox potential of water is defined as the absolute energy relative to the vacuum level or to the standard electrode in electrochemistry. Although there are asymmetric elements in the inter-electrode space as described above, how they are responsible for the Pockels effect of water is not trivial. As a preliminary experiment, we obtained the result that the magnitude of the deflection signal was much lower for Pt-Pt electrodes than for ITO-Pt or ITO-ITO electrodes. This shows that the signal size depends on the combination of the electrodes, but the relevance to the symmetry breaking effect is presently not clear.

From the experimental deflection angle, the size of the Pockels coefficient, r_{33} , relevant to the present experimental condition (the external electric field is parallel to the optical electric field^{4,5}) can be estimated in the following way as shown in Fig. 4. When the beam travels parallel to the elec-

trode under the refractive index gradient, the optical path difference (OPD) arises within the beam cross section to distort the wave front. The maximum OPD within the beam diameter d caused by propagation through the electric field applied volume over the distance L , which is the length of the electrodes, is given by $(\frac{\Delta n}{\Delta x} d)L$. Here it is assumed that $\frac{\Delta n}{\Delta x}$ is constant as inferred from the experimental observation. Since the light beam is propagated in such a way that the propagation direction is orthogonal to the wavefront, this OPD should be equivalent to the OPD caused by beam deflection, $nd \sin \theta$, with n and θ being, respectively, the refractive index of water and the deflection angle. With $n = 1.33$, $\theta = 4 \times 10^{-9}$, and $L = 4$ mm, we obtain $\Delta n = 1.0 \times 10^{-10}$. From the impedance analysis of the equivalent circuit of the system, the voltage falls in bulk water is estimated to be 0.05 V. With the electrode separation of 4 mm, the average electric field between the electrodes is estimated to be 12 V/m. The order of magnitude of r_{33} for “bulk water” is therefore estimated to be 10 pm/V, which is one order of magnitude smaller than that for the EDL but well comparable to that for typical electrooptic crystals.¹

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¹³We measured the frequency dependence of the signal from $f = 125$ to 1525 Hz to obtain that the signal magnitude was kept until $f = 225$ Hz and decreased with frequency from $f = 225$ Hz to 1525 Hz. The frequency dependence is needed to be more thoroughly studied in terms of both the impedance of the experimental system and the microscopic response of water.