

ON THE EFFECT OF AN ELECTRIC FIELD GRADIENT ON THE DIELECTRIC PERMITTIVITY OF A FLUID

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Summary. The effect of an electric field gradient on the dielectric permittivity of fluids is discussed. Numerical computations are carried out. These show the effect to be generally so small as to involve considerable experimental difficulties. On the other hand, if measurements were rendered possible, the value and sign of the quadrupole moment of the molecule could be determined immediately from the relatively simple formula derived for the effect under consideration.

1. INTRODUCTION

The present paper deals with the theory of the effect of a steep electric field gradient on the dielectric permittivity of fluids.

The solution of the problem with respect to the refractive index is due to Buckingham [1]. For the case of a field gradient produced by the four-wire condenser shown schematically in Fig. 1, Buckingham's theory yields a formula of the type

$$n_x - n_y = \left(A + \frac{B}{T} \right) E_{xx} \quad (1.1)$$

for the difference $n_x - n_y$ between the refractive indexes of a fluid for light beams travelling along the z -axis and with electric vectors in the x and y directions; herein, $E_{xx} = -E_{yy}$ is the electric field gradient, and T — the absolute temperature. The quantities A and B characterize properties of the medium; in the case of a gas, A is dependent on the molecular quadrupole moment induced in a molecule by the uniform field E , and B — on the permanent quadrupole moment and anisotropy of polarizability of the molecule. The effect described by eq. (1.1) provides a direct method of determining the numerical value and sign of the quadrupole moment.

With regard to the analogous effect consisting in a variation of the dielectric constant, the formula

$$\varepsilon_x - \varepsilon_y = \left\{ A + \frac{B}{T} + \frac{1}{T} \left(C + \frac{D}{T} \right) \right\} E_{xx}, \quad (1.2)$$

is derived in the present paper. In addition to the quantities A and B of eq. (1.1), it contains the two quantities C and D . C depends on the induced quadrupole and permanent dipole moments, and D is linear in the permanent quadrupole and quadratic in the permanent dipole moment of the molecule. In the case of a liquid, the quantities B , C and D depend, moreover, on the angular intermolecular correlations and on the fields existing between the molecules.

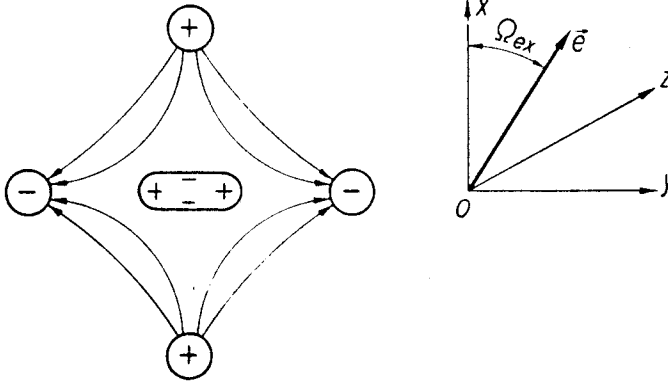


Fig. 1. Four-wire condenser yielding an electric field gradient $E_{xx} = -E_{yy}$ at its centre O . Conducting wires run $(\ominus \oplus \ominus)$ parallel to the z — axis (which is the axis of the condenser, perpendicular to the xy — plane); \mathbf{e} is the unit vector in the direction of the electric measuring field \mathbf{E} . If a fluid is introduced into this four-wire condenser, the quadrupole molecules $(\oplus \ominus)$ will be orientated, and electric anisotropy is induced in the medium. In the case of Buckingham's effect, \mathbf{E} is the electric vector at 45° to the x — axis of the linearly polarized light beam travelling in the z — direction.

Notwithstanding the fact that, in many substances the absolute value of the $|\varepsilon_x - \varepsilon_y|$ effect may well exceed the one represented by $|n_x - n_y|$, its measurement will surely involve considerable technical difficulties (e. g., how should one proceed in measuring with a weak uniform electric field so as to avoid perturbing the steep gradient of the field giving rise to the effect under consideration?). Apparently, the $\varepsilon_x - \varepsilon_y$ effect should be more easily accessible to measurement by microwave methods

employing a wave whose length is below the range of Debye dispersion of the substance investigated. However, the present paper is not intended to analyze the difficulties of measuring the "hypothetic" effect, but primarily to give an account of its theory and relationship with the readily measurable Kerr effect.

2. THE GENERAL THEORY

Let us consider a homogeneous isotropic medium as represented by a large spherical specimen of macroscopic size. At the centre of this large sphere we shall be considering a small macroscopic sphere of volume V . We suppose that this specimen is in a weak uniform electric field E and in a strong field gradient E_{xx} . In this case, the dielectric constant of a dense medium is given by the general equation

$$\frac{\epsilon_s - 1}{\epsilon_s + 2} E = \frac{4\pi}{3V} \langle \mathbf{M} \cdot \mathbf{e} \rangle_{E, E_{xx}}, \quad (2.1)$$

where $\mathbf{M} = \mathbf{M}(\tau, E, E_{xx})$ is the dipole electric moment of the small sphere when its molecules are in the configuration τ and when the external field is E and the field gradient E_{xx} ; \mathbf{e} denotes the unit vector in the direction of the measuring electric field \mathbf{E} .

The brackets $\langle \rangle_{E, E_{xx}}$ in eq. (2.1) stand for the statistical average in the presence of the external fields E and E_{xx} . In the classical treatment, the configurational variables τ are continuous, and statistical mechanics leads to the formula

$$\langle \mathbf{M} \cdot \mathbf{e} \rangle_{E, E_{xx}} = \frac{\int \mathbf{M}(\tau, E, E_{xx}) \cdot \mathbf{e} \exp \left\{ -\frac{U(\tau, E, E_{xx})}{kT} \right\} d\tau}{\int \exp \left\{ -\frac{U(\tau, E, E_{xx})}{kT} \right\} d\tau}, \quad (2.2)$$

here, $U(\tau, E, E_{xx})$ is the total potential energy of the large sphere in the configuration τ and with the external fields E and E_{xx} , k - Boltzmann's constant, and T - the Kelvin temperature of the system.

$\langle \mathbf{M} \cdot \mathbf{e} \rangle_{E, E_{xx}}$ will now be expanded in a power series in E and E_{xx} :

$$\begin{aligned} \langle \mathbf{M} \cdot \mathbf{e} \rangle_{E, E_{xx}} = & \langle \mathbf{M} \cdot \mathbf{e} \rangle + \left\{ \frac{\partial}{\partial E} \langle \mathbf{M} \cdot \mathbf{e} \rangle_{E, E_{xx}} \right\}_0 E + \\ & + \left\{ \frac{\partial^2}{\partial E \partial E_{xx}} \langle \mathbf{M} \cdot \mathbf{e} \rangle_{E, E_{xx}} \right\}_0 E E_{xx} + \dots \end{aligned} \quad (2.3)$$

Since, at $E = E_{xx} = 0$,

$$\langle \mathbf{M} \cdot \mathbf{e} \rangle = \left\langle \frac{\partial U}{\partial \mathbf{E}_a} e_a \right\rangle = 0, \quad \left\langle \frac{\partial U}{\partial \mathbf{E}_{a\beta}} (x_\alpha x_\beta - y_\alpha y_\beta) \right\rangle = 0,$$

we obtain

$$\begin{aligned} \langle \mathbf{M} \cdot \mathbf{e} \rangle_{E, E_{xx}} &= \left\langle \left(\frac{\partial M_a}{\partial \mathbf{E}_\beta} - \frac{1}{kT} M_a \frac{\partial U}{\partial \mathbf{E}_\beta} \right) e_a e_\beta \right\rangle E + \\ &+ \left\langle \left(\frac{\partial^2 M_a}{\partial \mathbf{E}_\beta \partial \mathbf{E}_{\gamma\delta}} - \frac{1}{kT} \left(\frac{\partial M_a}{\partial \mathbf{E}_\beta} \frac{\partial U}{\partial \mathbf{E}_{\gamma\delta}} + \frac{\partial U}{\partial \mathbf{E}_a} \frac{\partial M_\beta}{\partial \mathbf{E}_{\gamma\delta}} + M_a \frac{\partial^2 U}{\partial \mathbf{E}_\beta \partial \mathbf{E}_{\gamma\delta}} \right) \right) \right. \\ &\left. + \frac{1}{k^2 T^2} M_a \frac{\partial U}{\partial \mathbf{E}_\beta} \frac{\partial U}{\partial \mathbf{E}_{\gamma\delta}} \right\rangle e_a e_\beta (x_\gamma x_\delta - y_\gamma y_\delta) \rangle E E_{xx} + \dots, \end{aligned} \quad (2.4)$$

where the brackets $\langle \quad \rangle$ without lower index denote the statistical mean value in the absence of external fields ($E = E_{xx} = 0$)

$$\langle \Phi \rangle = \frac{\int \Phi(\tau, 0) \exp \left\{ -\frac{U(\tau, 0, 0)}{kT} \right\} d\tau}{\int \exp \left\{ -\frac{U(\tau, 0, 0)}{kT} \right\} d\tau}. \quad (2.5)$$

The general relationships:

$$M_a^T = -\frac{\partial}{\partial \mathbf{E}_a} \left\{ U(\tau, E, E_{xx}) \right\}, \quad \Theta_{a\beta}^T = -3 \frac{\partial}{\partial \mathbf{E}_{a\beta}} \left\{ U(\tau, E, E_{xx}) \right\} \quad (2.6)$$

define the total dipole and quadrupole moments of the large sphere.

With the definitions (2.6), and with the isotropic mean values of the functions of the unit vector components e_a , x_a and y_a :

$$\begin{aligned} \overline{e_a e_\beta} &= \overline{x_\alpha x_\beta} = \overline{y_\alpha y_\beta} = \frac{1}{3} \delta_{\alpha\beta}, \\ \overline{e_a e_\beta (x_\gamma x_\delta - y_\gamma y_\delta)} &= \frac{1}{30} (2 \cos^2 \Omega_{ex} - 1) \{ -2 \delta_{a\beta} \delta_{\gamma\delta} + 3 (\delta_{a\gamma} \delta_{\beta\delta} + \delta_{a\delta} \delta_{\beta\gamma}) \}, \end{aligned} \quad (2.7)$$

the expansion (2.4) leads to the result

$$\begin{aligned} \langle \mathbf{M} \cdot \mathbf{e} \rangle_{E, E_{xx}} &= \frac{1}{3} \left\langle \frac{\partial M_a}{\partial \mathbf{E}_a} + \frac{1}{kT} M_a M_a^T \right\rangle E + \frac{1}{15} (2 \cos^2 \Omega_{ex} - 1) \left\langle 3 \frac{\partial^2 M_a}{\partial \mathbf{E}_\beta \partial \mathbf{E}_{a\beta}} + \right. \\ &+ \left. \frac{1}{kT} \left(\frac{\partial M_a}{\partial \mathbf{E}_\beta} \Theta_{a\beta}^T + 3 M_a \frac{\partial M_\beta^T}{\partial \mathbf{E}_{a\beta}} + 3 M_a^T \frac{\partial M_\beta}{\partial \mathbf{E}_{a\beta}} \right) + \frac{1}{k^2 T^2} M_a M_\beta^T \Theta_{a\beta}^T \right\rangle E E_{xx} + \dots, \end{aligned} \quad (2.8)$$

where Ω_{ex} is the angle subtended by the unit vector \mathbf{e} and the direction of the x -axis (see Fig. 1), and $\delta_{\alpha\beta} = \begin{cases} 1 & \text{for } \alpha = \beta \\ 0 & \text{for } \alpha \neq \beta. \end{cases}$

Substituting (2.8) in eq. (2.1), the equation for ε_s may be written in the following general form:

$$\frac{\varepsilon_s - 1}{\varepsilon_s + 2} = \frac{\varepsilon - 1}{\varepsilon + 2} + \frac{4\pi}{45V} (2 \cos^2 \Omega_{ex} - 1) \left\langle 3 \frac{\partial^2 M_\alpha}{\partial E_\beta \partial E_{\alpha\beta}} + \frac{1}{kT} \left(\frac{\partial M_\alpha}{\partial E_\beta} \Theta_{\alpha\beta}^T + 3 M_\alpha \frac{\partial M_\beta^T}{\partial E_{\alpha\beta}} + 3 M_\alpha^T \frac{\partial M_\beta}{\partial E_{\alpha\beta}} \right) + \frac{1}{k^2 T^2} M_\alpha M_\beta^T \Theta_{\alpha\beta}^T \right\rangle E_{xx}, \quad (2.9)$$

wherein [2]

$$\frac{\varepsilon - 1}{\varepsilon + 2} = \frac{4\pi}{9V} \left\langle \frac{\partial M_\alpha}{\partial E_\alpha} + \frac{1}{kT} M_\alpha M_\alpha^T \right\rangle \quad (2.10)$$

is the equation for the dielectric constant in the absence of the field gradient ($E_{xx} = 0$).

The electric field gradient E^s inside the sphere is related to the field gradient E_{xx} in *vacuo* by the formula [1]:

$$E_{xx}^s = \frac{5}{2\varepsilon + 3} E_{xx}, \quad (2.11)$$

and, from eq. (2.9) we have¹

$$\varepsilon_x - \varepsilon_y = \frac{8\pi(\varepsilon + 2)^2(2\varepsilon + 3)}{675V} \left\langle 3 \frac{\partial^2 M_\alpha}{\partial E_\beta \partial E_{\alpha\beta}} + \frac{1}{kT} \left(\frac{\partial M_\alpha}{\partial E_\beta} \Theta_{\alpha\beta}^T + 3 M_\alpha \frac{\partial M_\beta^T}{\partial E_{\alpha\beta}} + 3 M_\alpha^T \frac{\partial M_\beta}{\partial E_{\alpha\beta}} \right) + \frac{1}{k^2 T^2} M_\alpha M_\beta^T \Theta_{\alpha\beta}^T \right\rangle E_{xx}^s, \quad (2.12)$$

wherein ε_x and ε_y are the values of ε_s for $\Omega_{ex} = 0^\circ$ and $\Omega_{ex} = 90^\circ$, i.e. when the unit vector \mathbf{e} is parallel and perpendicular to the direction of the x -axis, respectively.

3. APPLICATIONS OF THE THEORY TO SOME SPECIAL CASES

If the sphere contains N similar molecules, we may expand M_α^T and $\Theta_{\alpha\beta}^T$ as the sums of the moments of the individual molecules:

$$M_\alpha^T = M_\alpha = \sum_{p=1}^N m_\alpha^{T(p)}, \quad \Theta_{\alpha\beta}^T = \sum_{p=1}^N \Theta_{\alpha\beta}^{T(p)} \quad (3.1)$$

¹ From eq. (2.9) we can also obtain the quantity $\Delta\varepsilon_s = \varepsilon_s - \varepsilon$, determining the effect of electric saturation produced in the medium by a strong electric field gradient. Between $\Delta\varepsilon_s$ and the gradient-induced anisotropy $\varepsilon_x - \varepsilon_y$, given by eq. (2.12), exists the following relation:

$$\Delta\varepsilon_s = \frac{1}{2} (2 \cos^2 \Omega_{ex} - 1) (\varepsilon_x - \varepsilon_y).$$

where $m_a^{T(p)}$ and $\Theta_{\alpha\beta}^{T(p)}$ are the total dipole and quadrupole moments of the p -th molecule within the sphere. Using the expansions as given by Buckingham [1] for the isolated molecule, we have analogously for the p -th molecule immersed in the medium (the hyperpolarizability tensors are neglected):

$$m_a^{T(p)} = \mu_a^{(p)} + a_{\alpha\beta}^{(p)} (E_\beta + F_\beta^{(p)}) + \frac{1}{3} A_{\alpha;\beta\gamma}^{(p)} (E_{\beta\gamma} + F_{\beta\gamma}^{(p)}) + \frac{1}{3} B_{\alpha\beta;\gamma\delta}^{(p)} (E_\beta + F_\beta^{(p)}) (E_{\gamma\delta} + F_{\gamma\delta}^{(p)}) + \dots, \quad (3.2)$$

$$\Theta_{\alpha\beta}^{T(p)} = \Theta_{\alpha\beta}^{(p)} + A_{\gamma;\alpha\beta}^{(p)} (E_\gamma + F_\gamma^{(p)}) + \frac{1}{2} B_{\alpha\beta;\gamma\delta}^{(p)} (E_\gamma + F_\gamma^{(p)}) (E_\delta + F_\delta^{(p)}) + \dots, \quad (3.3)$$

wherein $\mu_a^{(p)}$ and $\Theta_{\alpha\beta}^{(p)}$ are the electric permanent dipole and quadrupole moments of the p -th isolated molecule and $a_{\alpha\beta}^{(p)}$ — its electric polarizability tensor. The tensors $A_{\alpha;\beta\gamma}^{(p)}$ and $B_{\alpha\beta;\gamma\delta}^{(p)}$ describe the quadrupole moments induced in the p -th molecule by a uniform electric field. $F_\alpha^{(p)}$ and $F_{\alpha\beta}^{(p)}$ are the α — component of the intermolecular field and $\alpha\beta$ — component of the field gradient at the centre of molecule p due to the charge distribution of its neighbours.

With respect to (3.1), we may write eq. (2.12) in the form involving molecular parameters:

$$\begin{aligned} \varepsilon_x - \varepsilon_y = & \frac{8\pi(\varepsilon + 2)^2(2\varepsilon + 3)}{675V} \left\langle 3 \sum_{p=1}^N \frac{\partial^2 m_a^{T(p)}}{\partial E_\beta \partial E_{\alpha\beta}} + \right. \\ & + \frac{1}{kT} \sum_{p=1}^N \sum_{q=1}^N \left(\frac{\partial m_a^{T(p)}}{\partial E_\beta} \Theta_{\alpha\beta}^{T(q)} + 6 m_a^{T(p)} \frac{\partial m_\beta^{T(q)}}{\partial E_{\alpha\beta}} \right) + \\ & \left. + \frac{1}{k^2 T^2} \sum_{p=1}^N \sum_{q=1}^N \sum_{r=1}^N m_a^{T(p)} m_\beta^{T(q)} \Theta_{\alpha\beta}^{T(r)} \right\rangle E_{xx}^s. \end{aligned} \quad (3.4)$$

Neglecting in the foregoing equation the small terms $\partial F_\alpha^{(p)} / \partial E_\beta$, $\partial F_{\alpha\beta}^{(p)} / \partial E_{\alpha\beta}$, ... we obtain a form better adapted to further discussion, namely:

$$\begin{aligned} \varepsilon_x - \varepsilon_y = & \frac{8\pi(\varepsilon + 2)^2(2\varepsilon + 3)}{675} \rho \left\{ B_{\alpha\beta;\alpha\beta} + \frac{a_{\alpha\beta} \Theta_{\gamma\delta}}{kT} \left\langle \sum_{q=1}^N \omega_{\alpha\gamma}^{(pq)} \omega_{\beta\delta}^{(pq)} \right\rangle + \right. \\ & \left. + \frac{2}{kT} m_\alpha A_{\beta;\gamma\delta} \left\langle \sum_{q=1}^N \omega_{\alpha\gamma}^{(pq)} \delta_{\beta\delta} \right\rangle + \frac{1}{k^2 T^2} m_\alpha m_\beta \Theta_{\gamma\delta} \left\langle \sum_{q=1}^N \sum_{r=1}^N \omega_{\alpha\gamma}^{(pq)} \omega_{\beta\delta}^{(pr)} \right\rangle \right\} E_{xx}^s, \end{aligned} \quad (3.5)$$

where $\omega_{\alpha\gamma}^{(pq)}$ denotes the cosine of the angle subtended by the a and γ axes of the molecular systems of reference rigidly attached to the p -th and q -th molecules, and $\varrho = N/V$.

(i) **Perfect Gas.** For a perfect gas, eq. (3.5) assumes the form:

$$\varepsilon_x - \varepsilon_y = \frac{8\pi}{15} \varrho \left\{ B_{a\beta:a\beta} + \frac{1}{kT} a_{a\beta} \Theta_{a\beta} + \frac{2}{kT} \mu_a A_{\beta:a\beta} + \frac{1}{k^2 T^2} \mu_a \mu_\beta \Theta_{a\beta} \right\} E_{xx}. \quad (3.6)$$

If the molecules are non-dipolar, $\mu_a = A_{\beta:a\beta} = 0$, and eq. (3.6) reduces to

$$\varepsilon_x - \varepsilon_y = \frac{8\pi}{15} \varrho \left\{ B_{a\beta:a\beta} + \frac{1}{kT} a_{a\beta} \Theta_{a\beta} \right\} E_{xx} \quad (3.7)$$

which is analogous with the one derived by Buckingham [1] for $n_x - n_y$ (see, eq. 1.1).

For axially symmetric polar molecules, eq. (3.6) yields

$$\varepsilon_x - \varepsilon_y = \frac{8\pi}{15} \varrho \left\{ B_{a\beta:a\beta} + \frac{1}{kT} (a_{||} - a_{\perp}) \Theta + \frac{2}{kT} \mu A + \frac{1}{k^2 T^2} \mu^2 \Theta \right\} E_{xx}, \quad (3.8)$$

where A and Θ are the induced and permanent quadrupole moments of the molecule possessing axial symmetry, and $a_{||}$, a_{\perp} — its electric polarizabilities parallel and perpendicular to the axis of symmetry.

(ii) **Liquids.** For polar liquids whose molecules have the axial symmetry we obtain from eq. (3.5);

$$\varepsilon_x - \varepsilon_y = \frac{8\pi(\varepsilon + 2)^2(2\varepsilon + 3)\varrho}{675} \left\{ B_{a\beta:a\beta} + \frac{2}{kT} mAR_P + \frac{1}{kT} (a_{||} - a_{\perp}) \Theta R_{CM} + \frac{1}{k^2 T^2} m^2 \Theta R_K \right\} E_{xx}^s \quad (3.9)$$

The quantities R_P , R_{CM} and R_K appearing herein are termed the angular intermolecular correlation factors for molecules possessing the axial symmetry, and are of the form

$$R_P = \left\langle \sum_{q=1}^N \cos \theta^{(pq)} \right\rangle, \quad (3.10)$$

$$R_{CM} = \frac{1}{2} \left\langle \sum_{q=1}^N (3 \cos^2 \theta^{(pq)} - 1) \right\rangle, \quad (3.11)$$

$$R_K = \frac{1}{2} \left\langle \sum_{q=1}^N \sum_{r=1}^N (3 \cos \theta^{(pq)} \cos \theta^{(pr)} - \cos \theta^{(qr)}) \right\rangle, \quad (3.12)$$

where $\theta^{(pq)}$ denotes the angle between the axes of symmetry of the p -th and q -th molecules. The factors R_P , R_{CM} and R_K of the same form also appear in the theory of molecular orientational effects given by Piekara and Kielich [3, 4].

For strongly polar molecules, the first two terms in eq. (3.9), accoun-

ting for the induced quadrupole moments A and B , can be omitted, yielding the following relations

$$\varepsilon_x - \varepsilon_y = \frac{36n^2(2\varepsilon + 3)}{5(n^2 + 2)^2} \frac{\Theta}{(a_{\parallel} - a_{\perp})} KE_{xx}^s, \quad (3.13)$$

where [4]

$$K = \frac{2\pi(n^2 + 2)^2(\varepsilon + 2)^2 \rho}{1215n^2 kT} (a_{\parallel} - a_{\perp}) \left\{ (a_{\parallel} - a_{\perp}) R_{CM} + \frac{m^2}{kT} R_K \right\} \quad (3.14)$$

is the Kerr constant. Here, a_{\parallel} and a_{\perp} are the optical polarizabilities of the molecule parallel and perpendicular to its axis of symmetry.

4. DISCUSSION

If, for a given substance, the gradient-induced anisotropy $\varepsilon_x - \varepsilon_y$ were known experimentally, and considering that Kerr's constant is easily measurable, eq. (3.13) would immediately yield the quadrupole moment of its molecule.

For carbon disulphide, we have the following experimental data at $t = 20^{\circ}\text{C}$: $n = 1.63$, $\varepsilon = 2.63$, $a_{\parallel} - a_{\perp} = 9.6 \cdot 10^{-24} \text{ cm}^3$ and $K = 12.1 \cdot 10^{-12} \text{ e.s.u.}$; by eq. (3.13), this yields

$$\varepsilon_x - \varepsilon_y = 9.2 \cdot 10^{12} \Theta E_{xx}.$$

For a field gradient of $E_{xx}^s = -1300 \text{ e.s.u.}$ as calculated by Buckingham [1] for $V = 20 \text{ kV}$ in the special condenser (fig. 1), and with a quadrupole moment of $\Theta = 5 \cdot 10^{-26} \text{ e.s.u.}$, we have for CS_2 :

$$|\varepsilon_x - \varepsilon_y| = 6 \cdot 10^{-10}.$$

Within existing techniques, an effect of this order of magnitude should hardly be accessible to detection. The chances for this should be better in liquids presenting a very high Kerr constant. Thus, for nitrobenzene: $n = 1.55$, $\varepsilon = 33.4$, $a_{\parallel} - a_{\perp} = 7.3 \cdot 10^{-24} \text{ cm}^3$ and $K = 13 \cdot 10^{-10} \text{ e.s.u.}$; hence, eq. (3.13) yields

$$\varepsilon_x - \varepsilon_y = 1.1 \cdot 10^{16} \Theta E_{xx}^s$$

and, with $E_{xx}^s = -1300 \text{ e.s.u.}$ and $\Theta = 15 \cdot 10^{-26} \text{ e.s.u.}$, a value of

$$|\varepsilon_x - \varepsilon_y| = 2.2 \cdot 10^{-6}.$$

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