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MAGNETOOPTICAL FARADAY EFFECT AND EFFECTIVE OPTICAL ANISOTROPY OF COMPLEXES IN LIQUID SOLUTIONS

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The magneto-optical Faraday effect is introduced as an experimental method for analysing molecular complexes and their optical anisotropy in highly dissolved binary solutions of liquids. The dependence of Verdet constant, V , on the mole concentration f_2 of *o*-bromoanisole in benzene and *o*-bromoanisole in *p*-xylene is used for calculation of the optical anisotropy δ^* and the anisotropy of optical polarizability ($b_{11}^* - b_{33}^*$) of complexes which are present in the solution as a result of molecular interactions in the investigated liquids. Numerical analysis of the values of V , δ^* and ($b_{11}^* - b_{33}^*$) are given. Measurements of the magneto-optical Faraday effect have been performed in a pulsed high magnetic field induction up to 20T and Laser beam $\lambda = 488$ nm.

KEY WORDS: Liquid solutions, Faraday effect, optical anisotropy.

1 INTRODUCTION

In the classical theoretical description the magneto-optical Faraday effect is determined by the square optical anisotropy, δ^* , of the medium molecules and by the atomic structure of individual molecules or molecular complexes. Existence of statistical complexes formed of dipolar and nondipolar molecules in highly dilute solutions of a dipolar liquid in nondipolar solvent has been reported in the papers on optical and magneto-optical analysis of intermolecular interactions in solutions of dielectric liquids^{1–7}. Coherent results of the studies performed by the methods of Rayleigh light scattering, molecular refraction, magneto-optical Cotton-Mouton and Faraday effect measurements provide the information on molecular characteristics of complexes made by the coupled dipolar and nondipolar molecules.

Reports of the results of Rayleigh light scattering, Cotton-Mouton and Faraday effect measurements for toluene solutions in benzene or *p*-xylene allowed drawing conclusions⁷ about the configuration of the dipolar (toluene) and nondipolar (benzene or *p*-xylene) molecules in their complex. This information was obtained from measurements of square optical anisotropy, δ^* , and the Cotton-Mouton, C , and Verdet, V , constants. The obtained results corroborate the conclusion that measurement of the Faraday effect in dilute solutions of liquids is a convenient method of determination of δ^* and its changes with mole concentration, f_2 , of a dipolar molecules in the solution.

That can be an alternative method to measurements of light scattering in a given solution. In many cases, e.g. in optically inhomogeneous systems, the proposed method of the Verdet constant measurement is a good solution to the problem of δ^* determination.

2 MAGNETOOPTICAL FARADAY EFFECT

Magneto-optical Faraday effect in a medium is a consequence of the magnetic field induced rotation, α , of the polarization plane of the light travelling in the medium at a distance \underline{L} , in parallel to the lines of the magnetic induction \underline{B} :

$$\alpha = VBL \quad (1)$$

where V is the Verdet constant of the medium. In the molecular-statistic approach the Faraday effect is expressed in terms of the dipolar moment \underline{m}_i , for the i -th molecule of the medium according to the following relation:

$$\underline{m}_i = \{b_{ij}^e + b_{ij}^{em} H_k^2(0) + \dots\} E_j^e(\omega). \quad (2)$$

Thus, the induced moment m_i , induced by the electric field $\underline{E}(\omega)$ of the light wave of frequency ω and by a static magnetic field $H(0)$ is composed of the electric component $m_i^e = b_{ij}^e E_j^e(\omega)$ and the magnetic one $m_i^m = b_{ij}^{em} H_k^2(0) E_j^e(\omega)$. The latter depends on the square of the intensity of the static magnetic field acting in the medium. In this approach the magnetic field dependent Verdet constant of the medium is expressed⁸ as:

$$V^{em}(\omega) = -i \frac{\pi\omega}{3c} \rho \frac{n^2(\omega) + 2}{3n(\omega)} \{b_{ijk}^{em} + (kT)^{-1} b_{ij}^e \mu_k^m\} \delta_{ijk}, \quad (3)$$

where c is the speed of light, $n(\omega)$ – light refraction index for the medium, ρ – density number of the medium, μ_k^m – the permanent magnetic moment of the medium molecules. For diamagnetic molecules we have $\mu_k^m = 0$. The parameter b_{ij}^e is the linear optical polarizability of molecules while b_{ijk}^{em} stands for their magneto-optical polarizability:

$$b_{ijk}^{em} \delta_{ijk} = b_{123}^{em} - b_{213}^{em} + b_{231}^{em} - b_{321}^{em} + b_{312}^{em} - b_{132}^{em}. \quad (4)$$

This magnetic polarizability, according to Eqs. (2) and (3), determines the contributions of the external magnetic field $H(0)$ and the optical field $E(\omega)$ to the dependence of the Faraday effect on the square of the magnetic field intensity. However, the nonlinear Faraday effect does not bring a contribution which could be experimentally verified in measurements of the Verdet constant. So far, in laboratory conditions the dependence of the Verdet constant on H^2 has been undetectable⁹. Because of that we can assume $b_{ijk}^{em} = 0$ for diamagnetic media $\mu_k^m = 0$ and $V^{em}(\omega) = 0$. Finally, $m_i = m_i^e$:

$$m_i^e = b_{ij}^e E_j(\omega), \quad (5)$$

and a classical description¹⁰ of the Verdet constant gives:

$$V = -\frac{\pi(n^2 - 1)^2}{2n\lambda^2 N_L e p} (1 - \delta^*/2). \quad (6)$$

where N_L is the Lösschmidt number, λ – the light wave-length corresponding to the frequency ω , e – elementary charge, p – the number of dispersive electrons in a molecule. The quantity δ^* is the square optical anisotropy of the medium effective molecule and is expressed as:

$$\delta^* = \frac{(b_{11}^* - b_{22}^*)^2 + (b_{22}^* - b_{33}^*)^2 + (b_{33}^* - b_{11}^*)^2}{(b_{11}^* + b_{22}^* + b_{33}^*)^2}, \quad (7)$$

in which b_{11}^* , b_{22}^* , b_{33}^* are the components of the optical polarizability tensor in the directions of the three main symmetry axes of the optical polarizability of a molecule. The quantity δ^* is identical to the square of the optical anisotropy, δ^2 , used in description of the Rayleigh light scattering and Cotton-Mouton effect¹⁰⁻¹². The formulation of Eq. (6) suggests a possibility of using the Faraday effect measurements for determination of δ^* of the medium molecules. The proposed method involves measurement of the Verdet constant and light refraction index of the medium. Knowledge of a value of the p factor describing the effective number of dispersive electrons in a molecule is not required for determination of δ^* for molecular complexes.

3 OPTICAL ANISOTROPY OF A BINARY MOLECULAR COMPLEXES

Optical anisotropy of an individual molecule is determined by the differences of its optical polarizability along the three main directions of its symmetry axes. Eq. (7) determines quantitatively the optical anisotropy of an isolated molecule or an effective molecule (complex) where $(b_{11}^* + b_{22}^* + b_{33}^*)/3$ is defined as the mean optical polarizability.

A real liquid or a real undiluted gas are a medium of interacting molecules or atoms. Intermolecular interaction in a liquid leads to formation of statistical binary, ternary and higher molecular complexes. However, in liquids the most probable is formation of binary complexes. A binary complex is characterized by optical anisotropy δ^* different from the anisotropy δ of an isolated individual molecules in the medium. One of the methods used to gain information on δ^* is its determination from the Faraday effect measurements in a binary liquid solution⁷.

From Eq. (6) we have:

$$V \frac{n}{(n^2 - 1)} = - (A/p) (1 - \delta^*/2), \quad (8)$$

where $A = \pi/2N_L e = \text{const}$. Thus, Eq. (8) can be rewritten as:

$$\left(1 - \frac{\delta^*}{2}\right) = -V \frac{np}{(n^2 - 1)^2 A}. \quad (9)$$

For individual components “1” and “2”, of the binary solutions, we get, respectively:

$$\left(1 - \frac{\delta_1^*}{2}\right) = -V_1 \frac{n_1 p_1}{(n_1^2 - 1)^2 A}; \quad \left(1 - \frac{\delta_2^*}{2}\right) = -V_2 \frac{n_2 p_2}{(n_2^2 - 1)^2 A}. \quad (9a)$$

For solution containing component “1” at a mole concentration f_1 and component “2” at a mole concentration f_2 , where $f_2 + f_1 = 1$, we have:

$$\left(1 - \frac{\delta_{21}^*}{2}\right) = -V_{21} \frac{n_{21} p_{21}}{(n_{21}^2 - 1)^2 A}. \quad (10)$$

Applying the additivity rule to the number p of dispersive electrons¹⁰ in the molecular system of the solution we get: $p_{21} = p_1 f_1 + p_2 f_2$, and combining Eqs. 9a and 10 we arrive at:

$$\frac{\delta_{21}^*}{2} = 1 + \left\{ \left(\frac{\delta_1^*}{2} - 1 \right) \frac{(n_1^2 - 1)^2}{n_1 V_1} (1 - f_2) + \left(\frac{\delta_2^*}{2} - 1 \right) \frac{(n_2^2 - 1)^2}{n_2 V_2} f_2 \right\} \frac{n_{21} V_{21}}{(n_{21}^2 - 1)^2}. \quad (11)$$

As follows from Eq. (11), the optical anisotropy δ_{21}^* of a complex in a binary solution with component “2” at a concentration f_2 is determined by the values of δ_1^* and δ_2^* , optical anisotropies of pure components “1” and “2”, the Verdet constants V_1 and V_2 of the solution components and light refraction indices n_1 , n_2 and n_{21} of the components and the solution. Thus, when the additivity rule for the effective number of dispersive electrons for two coupled molecules is fulfilled we obtain a value of δ_{21}^* which is the square optical anisotropy of a complex. It should be noted that δ_{21}^* is determined by the dependence of the Verdet constant V_{21} on mole concentration f_2 of the solution, Eq. (11). Changes in V_{21} with f_2 provide information on the configuration of coupled molecules forming statistical complexes. Thus, measurements of the Faraday effect in liquid solutions are a method of analysis of intermolecular interactions. An illustration of the above described method of δ^* measurements are the values of δ^* (Fig. 2) and the anisotropy ($b_{11}^* - b_{33}^*$) of optical polarizability (Fig. 4) determined for binary *o*-bromoanisole–benzene and *o*-bromoanisole–*p*-xylene complexes.

4 RESULTS AND DISCUSSION

Magneto-optical Faraday effect of the studied solutions was measured in a strong pulse magnetic field of induction, B , up to 20T using laser light of a wavelength $\lambda = 488$ nm. The angle of rotation of the light polarization plane in the Faraday effect was recorded by the photo-electric method¹³.

Figures 1a and 1b present the measured Verdet constant of *o*-bromoanisole solution in benzene and *p*-xylene, respectively, as a function of the mole concentration f_2 of *o*-bromoanisole in solution.

According to Eq. (11), from these results and from the measured light refraction index n_{21} of the studied solutions we could calculate their square optical anisotropy δ_{21}^* . The square optical anisotropy dependences on f_2 concentration of the solution are

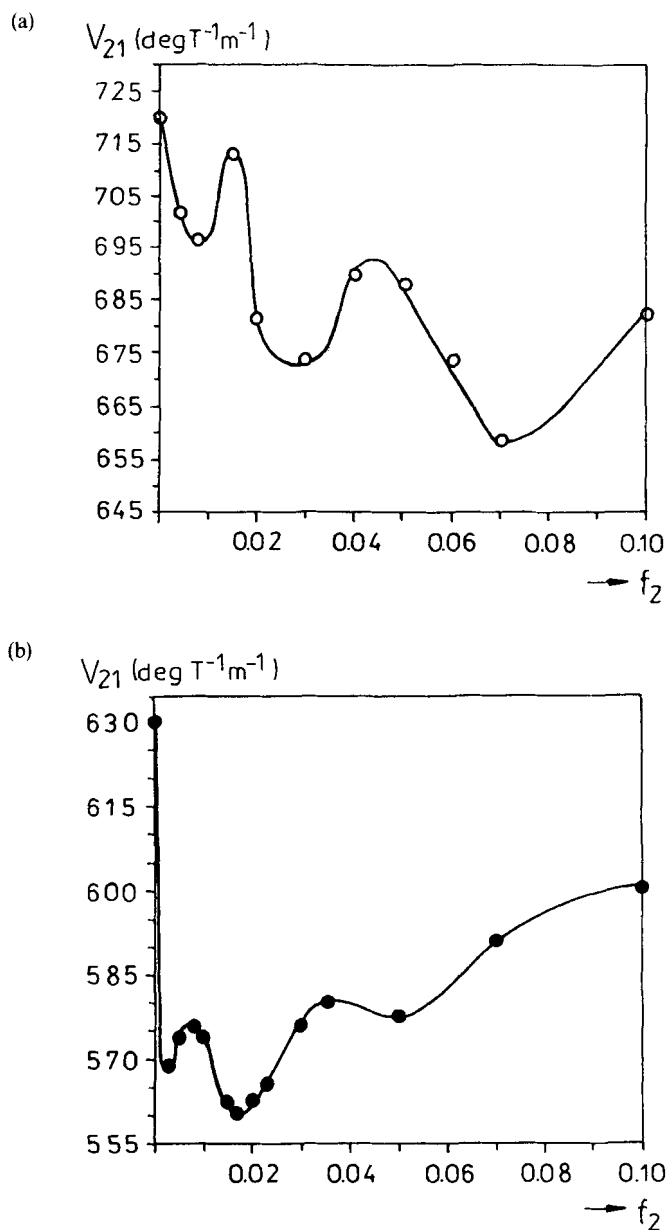


Figure 1 Verdet constant V_{21} of diluted solutions: o -bromoanisole in benzene (a), o -bromoanisole in p -xylene (b).

shown in Figures 2a and 2b for o -bromoanisole in benzene and p -xylene, respectively. For low f_2 concentrations, $f_2 < 0.1$, local extremes of the Verdet constant are observed. These extremes reflect the changes in the square optical anisotropy δ_{21}^* dependent on f_2 . The local extremes in V_{21} (Fig. 1) are seen to correspond to the local extremes in δ_{21}^* .

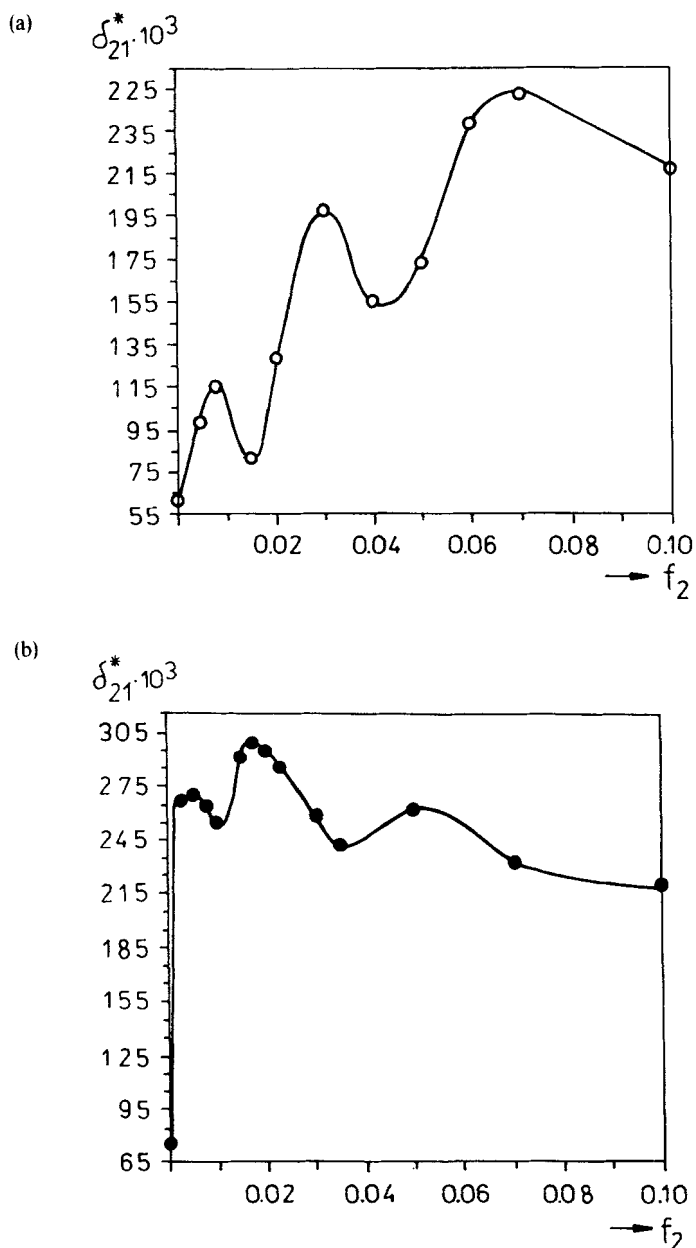


Figure 2 Optical anisotropy δ_{21}^* of complexes: (a)–solutions of *o*-bromoanisole in benzene, (b)–solutions of *o*-bromoanisole in *p*-xylene.

The local extremes in V_{21} appear as a consequence of formation of complexes in dilute solutions of *o*-bromoanisole in nondipolar benzene or *p*-xylene. These complexes include one molecule of *o*-bromoanisole and molecules of benzene or *p*-xylene.

In highly diluted solutions ($f_2 < 0.1$) each *o*-bromoanisole molecule is immersed in a sea of nondipolar solvent molecules. Thus, dipolar molecules of *o*-bromoanisole can be treated as well isolated from each other and in such a liquid solution mainly statistical molecular complexes of a dipolar molecule with a nondipolar one can be formed. The most probable is formation of binary (dipolar molecule–nondipolar molecule) complexes.

A molecular system of coupled molecules is characterized by square optical anisotropy δ_{21}^* . The changes of δ_{21}^* upon changing f_2 , Figures 2a and 2b, indicate the formation of coupled molecules whose configuration changes within the complex as a function of f_2 . If the symmetry of a statistical complex of a dipolar (*o*-bromoanisole) and nondipolar (benzene or *p*-xylene) molecule can be approximated by the axial one, we can put $b_{11}^* = b_{22}^*$ and write:

$$\delta_{21}^* = 2(b_{11}^* - b_{33}^*)^2 / (2b_{11}^* + b_{22}^*)^2. \quad (12)$$

The value of $(2b_{11}^* + b_{33}^*)$ from Eq. (12) is found from the results of molar refraction R^m measurements for the studied solutions ($R^m = (n^2 - 1)M / (n^2 + 2)\rho$, ρ – density, M – molecular mass of the solutions).

According to the relation:

$$R^m = (3\varepsilon_0)^{-1} N_A (2b_{11}^* + b_{33}^*) / 3 \quad (13)$$

where N_A is the Avogadro number and ε_0 is the permittivity of free space, we get:

$$(2b_{11}^* + b_{33}^*) = 9\varepsilon_0 R^m / N_A, \quad (14)$$

and after Eqs. (12) and (13) the anisotropy of optical polarizability of a molecule or a complex can be expressed as:

$$(b_{11}^* - b_{33}^*) = 3\bar{b}(\delta^*/2)^{1/2}, \quad (15)$$

where $\bar{b} = (2b_{11}^* + b_{33}^*)/3$ is the mean optical polarizability of the complex. From Eq. (14) and the results of R^m (Figs. 3a, 3b), measurements data³ of n_{21} , M and ρ we can get \bar{b} , whereas the value of $(\delta^*/2)^{1/2}$ can be found from Eq. (11). According to the latter equation, $\delta_{21}^*/2$, can be determined from measurements of:

- 1) the Verdet constant V_{21} of the studied solutions of concentration f_2 ,
- 2) light refraction index n_{21} of the studied solution,
- 3) the Verdet constants V_1 and V_2 , light refraction indices n_1 and n_2 , square optical anisotropies δ_1^* and δ_2^* of pure liquid components of the solution.

So, to get the square optical anisotropy of the complexes it is enough to measure V_{21} , n_{21} as a function of f_2 , V_1 , V_2 , n_1 and n_2 . Knowing δ_{21}^* (Fig. 2) we can find from Eq. (15) anisotropy of optical polarizability of a complex in solution, $(b_{11}^* - b_{33}^*)$. Changes of $(b_{11}^* - b_{33}^*)$ as a function of f_2 are caused by changes of geometrical configuration of molecules in a complex. The obtained dependences of $(b_{11}^* - b_{33}^*)$ on f_2 , for the studied complexes, are shown in Figures 4a and 4b. The curve for solutions of *o*-bromoanisole in benzene, Figure 4a, reveals a local maximum of $(b_{11}^* - b_{33}^*)$;

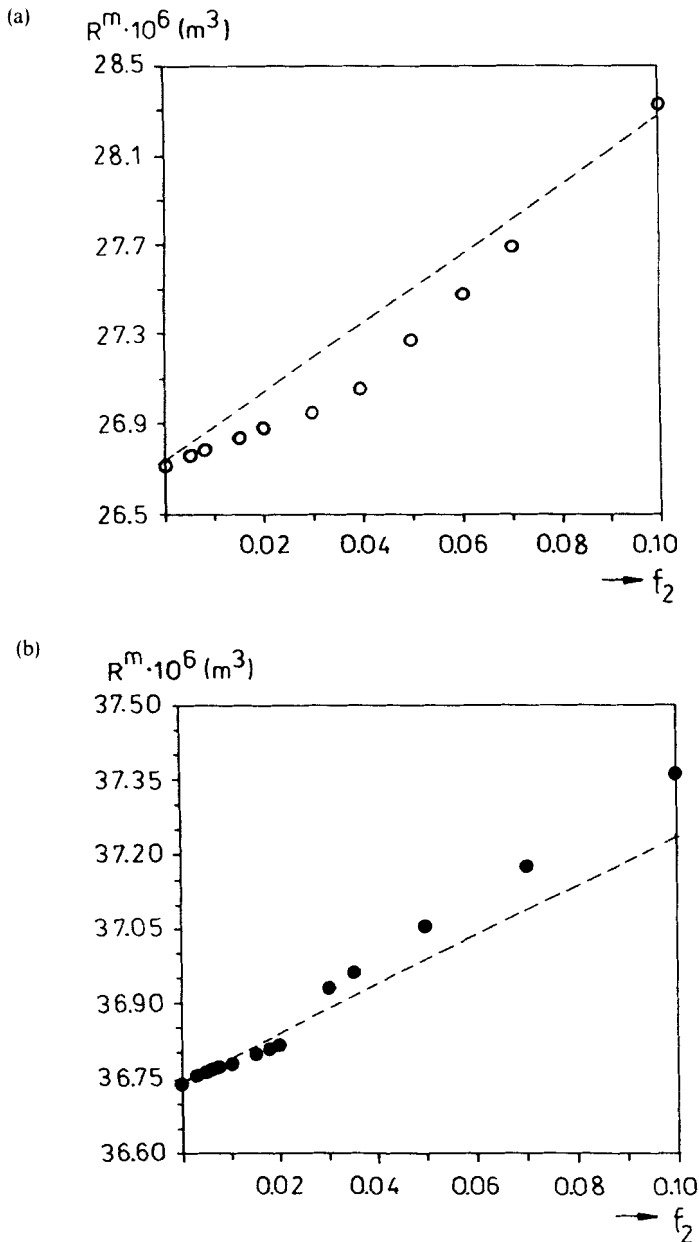


Figure 3 Molar refraction R^m of solutions: *o*-bromoanisole in benzene (a), *o*-bromoanisole in *p*-xylene (b) (dashed line—additivity relation for molar refraction).

$(b_{11}^* - b_{33}^*)_{\max} = 8.445 \times 10^{-40} \text{ Fm}^2$ for $f_2 \approx 0.008$, $11.209 \times 10^{-40} \text{ Fm}^2$ for $f_2 \approx 0.03$ and $12.906 \times 10^{-40} \text{ Fm}^2$ for $f_2 \approx 0.07$. For the solutions of mole concentration $f_2 \approx 0.015$ and $f_2 \approx 0.04$ the anisotropy of optical polarizability reaches a local minimum. The minimum value $(b_{11}^* - b_{33}^*)_{\min} = 7.112 \times 10^{-40} \text{ Fm}^2$ and $9.978 \times 10^{-40} \text{ Fm}^2$,

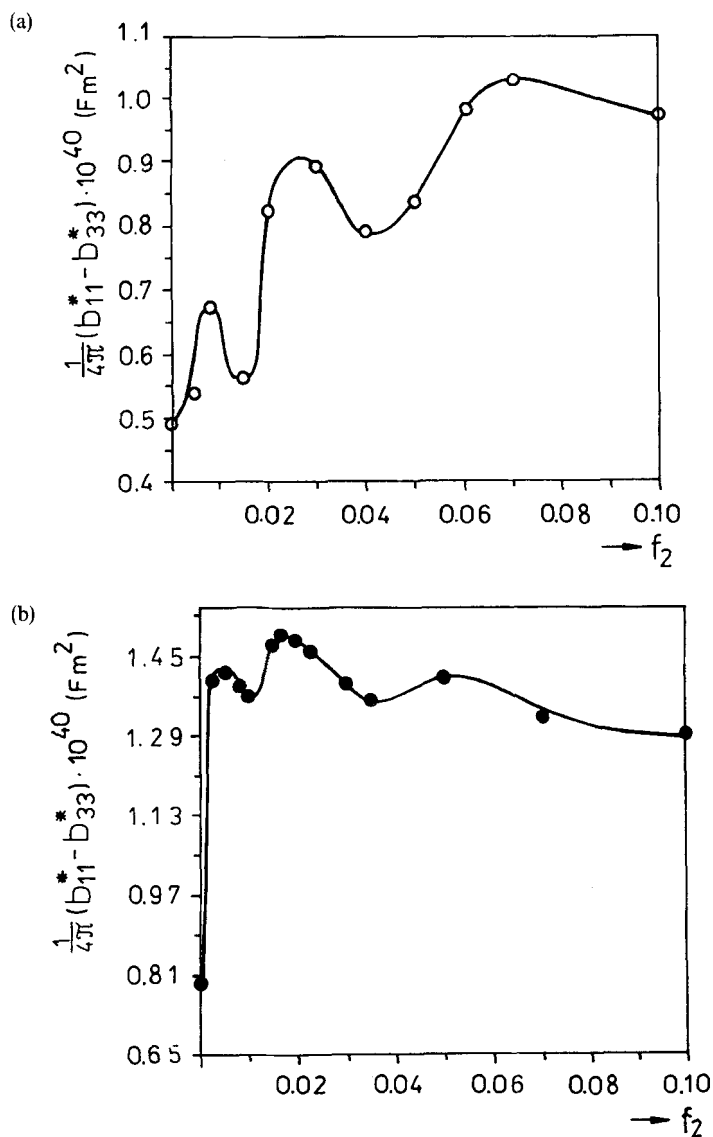


Figure 4 Anisotropy of optical polarizability ($b_{11}^* - b_{33}^*$) of complexes: (a)–solutions of *o*-bromoanisole in benzene, (b)–solutions of *o*-bromoanisole in *p*-xylene.

respectively. For $f_2 > 0.04$ the anisotropy of optical polarizability of the complex increases and for $f_2 = 0.1$ is equal $12.265 \times 10^{-40} \text{ Fm}^2$. For pure *o*-bromoanisole ($f_2 = 1$) a value of $(b_{11}^* - b_{33}^*)$ is $12.956 \times 10^{-40} \text{ Fm}^2$. Analysing the analogous dependences for *o*-bromoanisole in *p*-xylene, Figure 4b, we find a local maximum $(b_{11}^* - b_{33}^*)_{\max} = 17.203 \times 10^{-40} \text{ Fm}^2$ for $f_2 = 0.005$, $18.761 \times 10^{-40} \text{ Fm}^2$ for $f_2 \approx 0.017$ and $17.668 \times 10^{-40} \text{ Fm}^2$ for $f_2 \approx 0.05$. The local minima $(b_{11}^* - b_{33}^*)_{\min} = 17.228 \times 10^{-40} \text{ Fm}^2$, $17.140 \times 10^{-40} \text{ Fm}^2$ are present for $f_2 \approx 0.01$ and $f_2 = 0.035$, respectively.

The presence of local extremes of $(b_{11}^* - b_{33}^*)$ for similar concentrations of the solutions in benzene and *p*-xylene, the maxima at about $f_2 \approx 0.008, 0.03, 0.07$ and the minima $f_2 \approx 0.02$ and 0.04 indicates a similar mechanism of the interactions of *o*-bromoanisole with nondipolar solvent whose essential part is formation of statistical complexes. Local extremes of the effective anisotropy of optical polarizability of a complex of molecules appear as a consequence of changes in configuration of the molecules in the complex upon changing concentration of the solution. In the limiting^{14,15} cases the configuration of statistically coupled molecules is distinguished by the mutually nearly parallel or perpendicular orientation of the axes of maximum optical polarizability of the molecules in the complex. In the case of the mutually nearly parallel orientation of the axes of maximum polarizability of molecules, the anisotropy of the optical polarizability of a complex reaches the maximum value (Fig. 4a, for $f_2 \approx 0.008$; Fig. 4b, for $f_2 \approx 0.005$ and for $f_2 \approx 0.017$) whereas in the case this orientation is nearly perpendicular the anisotropy of the optical polarizability takes the minimum value (Fig. 4a, for $f_2 \approx 0.015$ and $f_2 \approx 0.04$; Fig. 4b, for $f_2 \approx 0.01$ and $f_2 \approx 0.035$). Similar relations between the geometrical configuration of molecules in a binary complex and the values of the anisotropy of optical polarizability have been obtained for dilute solutions of toluene in benzene and *p*-xylene⁷.

The conclusions formulated in this paper concerning the relationship between the observed extremes in the anisotropy of optical polarizability of a complex and the geometrical configuration of the molecules forming the complex are in agreement with theoretical predictions. For molecular complexes whose molecules have mutually nearly perpendicular orientation of their axes of the maximum optical polarizability the theory (J. A. Prins, W. Prins¹⁴ and S. Woźniak¹⁵) predicts the negative sign of the angular correlation factor, J^* , ($J^* < 0$) while for molecules of the nearly parallel orientation of directions of the maximum optical polarizability the angular correlation factor is positive, ($J^* > 0$).

An illustration of the above mentioned theoretical relations between the sign of J^* and geometrical orientation of directions of the maximum optical polarizability of two molecules forming a complex, are the results of J^* measurement reported in¹⁶ for solutions of *o*-bromoanisole in benzene or *p*-xylene. The results of these measurements have fully confirmed the conclusions following from the theoretical analysis.

For dilute solutions of *o*-bromoanisole in benzene and *o*-bromoanisole in *p*-xylene the value of the angular correlation factor J_{21}^* is negative and increase with mole concentration f_2 of *o*-bromoanisole in solutions (Table 1). Experimental results for bimolecular complexes give $J_{22}^* = -0.18$ for benzene⁷, 0.54 for *p*-xylene⁷ and 0.89 for *o*-bromoanisole¹⁶. Positive values of J_{22}^* for *p*-xylene and *o*-bromoanisole indicate a tendency for parallel orientation of these molecules in the bimolecular complex while negative value of J_{22}^* for benzene indicate a perpendicular orientation of benzene molecules in the bimolecular benzene complex. Existence of bimolecular complexes in analysed solutions is established by the data of molar refraction R^m (Fig. 3a) and results of $\Delta R^m > 0$ for $f_2 > 0.09$ (*o*-bromoanisole-benzene solutions). Also, $\Delta R^m > 0$ for *o*-bromoanisole-*p*-xylene solutions for $f_2 > 0.025$ (Fig.3b) where ΔR^m is the difference between experimental result of molar refraction, R^m , and calculated data, R_0^m , from the additivity relation: $R_0^m = R_1^m(1 - f_2) + R_2^m f_2$ (dashed line—Figures 3a, 3b)

Table 1 Angular Correlation Factor J^* for Dilute Solutions of *o*-Bromoanisole in Benzene (J_{21}^*), *p*-Xylene (J_{21}^{**}) and Pure Liquid (J_{22}^*), Verdet Constant V . Optical Anisotropy δ^* of *o*-Bromoanisole, Benzene and *p*-Xylene.

| | | | | | | | | | | |
|-----------------------|------------------------|-------|-------|----------------|-------|-------|------------------|-------|-------|-------|
| f_2 | 0.02 | 0.025 | 0.03 | 0.04 | 0.05 | 0.06 | 0.07 | 0.08 | 0.09 | 0.1 |
| J_{21}^* | -0.72 | -0.55 | -0.44 | -0.36 | -0.32 | -0.20 | -0.19 | -0.19 | -0.18 | -0.18 |
| J_{21}^{**} | -0.81 | -0.70 | -0.24 | -0.22 | -0.20 | -0.18 | -0.15 | -0.12 | -0.10 | -0.08 |
| <i>LIQUID</i> | <i>o</i> -BROMOANISOLE | | | <i>BENZENE</i> | | | <i>p</i> -XYLENE | | | |
| V (deg/Tm) | 809 | | | 720 | | | 630 | | | |
| $\delta^* \cdot 10^3$ | 112 | | | 62 | | | 75 | | | |
| J_{22}^* | 0.89 | | | -0.18 | | | 0.54 | | | |

$R_2^m = 41.62 \times 10^{-6} \text{ m}^3$ for *o*-bromoanisole and $R_1^m = 26.72 \times 10^{-6} \text{ m}^3$, $36.74 \times 10^{-6} \text{ m}^3$ for benzene and *p*-xylene, respectively.

The value of $\Delta R^m > 0$ approve for binary while $\Delta R^m < 0$ for ternary interactions¹.

5 CONCLUSIONS

Measurements of Verdet constant of dilute binary liquid solutions of polar solute in a nondipolar solvent make the basis of the proposed method of determination of square optical anisotropy of complexes forming in the solution. Assuming that the interaction between a dipolar molecule (solute) and nondipolar solvent is dominant in a dilute solution, we achieve a coherent interpretation of measurements of magneto-optical Faraday effect, molar refraction R^m and square optical anisotropy δ^* of complexes in solutions of *o*-bromoanisole in benzene and *p*-xylene. The presented interpretation of complex formation is supported by the presence of local maxima of Verdet constant and molar (ΔR^m) refraction in their dependences on the concentration of the dipolar component in the solution. The observed minima of square optical anisotropy δ_{21}^* and optical anisotropy ($b_{11}^* - b_{33}^*$), at a mole concentration of bromoanisole $f_2 \approx 0.015$ and $f_2 \approx 0.04$, prove that in a complex the molecules have their axes of maximum polarizability arranged in nearly perpendicular configuration. This configuration has been confirmed by results of the Cotton-Mouton effect investigation ($J_{21}^* < 0$). For increasing concentration, at $f_2 \approx 0.025$ (*o*-bromoanisole – benzene solution) and at $f_2 \approx 0.02$ (*o*-bromoanisole – *p*-xylene solution) local maxima in δ_{21}^* and ($b_{11}^* - b_{33}^*$) are observed which testifies to the change in the geometrical configuration of the molecules in a complexes. The results reported in this work indicate the usefulness of Faraday effect measurements for identification of the statistical molecular complexes forming in liquid solutions, determination of the value of anisotropy of optical polarizability as well as configuration of molecules in the complex. Geometrical configuration of the molecules forming the complexes can be analyzed by X-ray method¹⁷. This method is analysed now on the grounds of molecular model resulting from the data of the presented paper.

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