



Electro-optical effects in disperse systems in strong electric fields of arbitrary shape



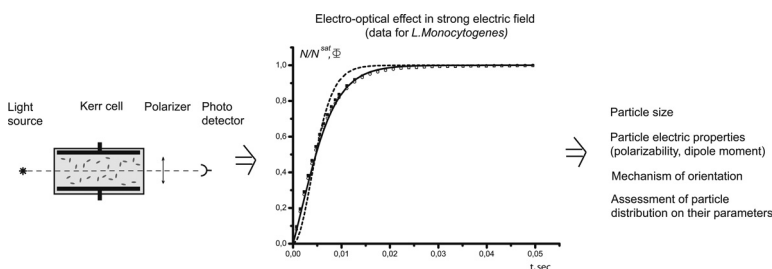
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HIGHLIGHTS

- Formulae describing electro-optical effects in strong electric fields were derived.
- Different particle orientation mechanisms and field shapes are considered.
- Obtained formulae were verified experimentally for *L. Monocytogenes* suspension.
- Particle orientation mechanism for *L. Monocytogenes* was studied.
- The proposed technique allows to assess polydispersity of the solution.

GRAPHICAL ABSTRACT



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ABSTRACT

Electro-optical techniques currently used to study disperse systems allow to determine different geometrical and electrical particle parameters in these systems. To reliably determine some of these parameters a long electric impulse or series of impulses has to be applied, which can alter some of the systems under study. To address this issue and allow to study disperse systems that cannot undergo lengthy application of electric field in our previous work we have proposed a technique to apply short pulses of electric field to the system under study which still allows determining particle parameters just like regular electro-optical techniques. In this article we further develop this technique by generalization of equations to the case of arbitrary field shape and particle orientation mechanism. The obtained experimental data for *L. Monocytogenes* verifies the developed equations for big particles.

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1. Introduction

The well-elaborated electro-optical techniques [1] allow one to determine such particle parameters in colloidal systems as polarizability anisotropy γ , particle dipole moment μ , rotary diffusion coefficient D , size r and others. If the system is polydisperse and/or polymorphic the distribution of particle parameters in the

disperse system can be either assumed or determined by applying some numerical techniques on the experimental data [2,3]. Several experimental techniques can be used to determine particle distribution function on rotary diffusion coefficient D , which is directly linked to particle size and shape. The first and most widespread is studying electro-optical effect relaxation from the state of saturated orientation of all particles in polydisperse system [4,5] (this technique has the benefit of having easily observable effects, but the drawback of having to apply substantial electric fields for a long time). The other involves studying disperse system behavior in weak sine-shaped electric field at high frequency with sine

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modulated amplitude varied at lower frequencies [6] (this technique has the drawback of having to deal with weak experimental signals, but in most cases does not alter the system under study because weak fields are used). Studying electro-optical effects during the application of electric field also allows to obtain particle electrical parameters, such as polarizability anisotropy γ or permanent dipole moment μ , which are, besides particle geometry, significantly influenced by particle nature [7–9]. The nature of electric polarizability of bacteria was studied in [10,11]. Numerical techniques [12] can be used to determine particle distribution function from the processing of experimental data obtained from these experiments.

In this article we present further development of the technique [13–15] related to the application of short pulses of strong electric field to colloidal systems. In [13] we derived the formulae that describe electro-optical effects in strong single sinusoidal electric field pulses only. At the same time it is often hard to obtain strong pulses of electric field of ideal shape: the obtained field shape is usually some approximation to a rectangle, which complicates the analysis of the obtained experimental data. Thus the reduction of electric field pulse length makes it necessary to consider the influence of its fronts on the observed electro-optical effect.

The formulae that allow to interpret experimental data in strong fields of arbitrary shape and obtain the same data from the experiment with short pulses of electric field as from the experiments with long (or repeating) pulses of electric field are presented in this article. We also extend the formulae to the case of particle orientation caused by permanent dipole moment, which allows to judge which particle orientation mechanism (due to permanent or induced dipole moment) is prevailing for a given system. The formulae are also generalized to the case of polydisperse systems and allow to easily assess the presence of distribution of particles in the suspension on electrical properties.

At last, we present the experimental data for bacterial suspension of *L. Monocytogenes* which was studied using the proposed technique, and conclude on the applicability of this technique to bacterial solutions.

2. Theoretical discussion

2.1. Particle orientation function in strong electric fields of arbitrary shape

The density function of angular distribution of dispersed particles W satisfies the rotary diffusion equation with force term:

$$\Delta W + \frac{1}{kT} \text{div}(W \cdot \vec{M}) = \frac{1}{D} \frac{\partial W}{\partial t}, \quad (1)$$

where \vec{M} is the rotary moment of force applied to particles in the suspension. If the moment of orienting force is big, the first term in (1) related to Brownian motion of the particles can be neglected. In this case we consider the electric field causing particle orientation strong.

The moment of force depends on the nature of particle orientation, angle Θ between the direction of force applied to a particle and its axis of orientation, and can change if the field strength is not constant. Here we derive formulae that describe electro-optical effects for two different cases of particle orientation in strong fields of arbitrary shape: the one caused by the presence of anisotropy of particle polarizability γ and the one due to particle permanent dipole moment μ .

The shape of electric field pulse front considerably influences the observed electro-optical effects only when the electric field is strong, since the particle torque moment is proportional to either the first (the case of orientation due to particle permanent dipole moment μ) or the second (the case of orientation due to particle

anisotropy of polarizability γ) power of electric field strength. If one divides the fronts of electric field applied to the system under study into elementary subintervals, then one can only consider the contribution of subintervals where the field is strong to particle orientation. This peculiarity was used here to solve (1).

Assuming Eq. (1) is solved and particle angular distribution function W is found particle orientation degree Φ can be obtained by integrating light intensity scattered by the particle in forward direction with $W(u, t)$ ($u = \cos \Theta$). In the generic case numerical techniques can be used to calculate the scattered light intensity [16], but in a number of practical cases the use of simple analytical scattering theories is sufficient. In the case of Rayleigh scattering [8] particle orientation degree Φ can be found by integrating W with the second Legendre polynomial $P_2(u)$ [17]:

$$\Phi(t) = \frac{\int_{-1}^1 P_2(u)W(u, t)du}{\int_{-1}^1 W(u, t)du}, \quad (2)$$

For small particles this formula is theoretically justified, however, experiments showed that for optical effects caused by light dichroism this formula is also applicable for big particles with size from 0.1 of incident light wave length to several light wave lengths, if incident light is white [2] (this is the case for the studied bacterial suspensions).

In the case of weak electric fields particle angular velocity, as well as the time of electro-optical effect rise, is also influenced by the Brownian motion and can considerably differ for different particles in a polydisperse suspension.

2.2. Orientation caused by anisotropy of polarizability

If particle orientation happens because of the presence of anisotropy of polarizability the moment of force $M = \gamma E^2(t) \sin 2\Theta/2$, where $\gamma = \gamma_{\parallel} - \gamma_{\perp}$ (γ_{\parallel} is the particle polarizability anisotropy along the main axis of orientation, γ_{\perp} is the particle polarizability anisotropy across the main axis of orientation) and, provided $\gamma E^2/kT \gg 1$ (1) can be expanded into

$$(3u^2 - 1)W(u, \tau_{\gamma}) + u(u^2 - 1) \frac{\partial W(u, \tau_{\gamma})}{\partial u} = \frac{\partial W(u, \tau_{\gamma})}{\partial \tau_{\gamma}}, \quad (3)$$

where

$$\tau_{\gamma} = \frac{\gamma \overline{DE^2}(t)t}{kT}, \quad \overline{E^2}(t) = \frac{1}{t} \int_0^t E^2(t')dt'. \quad (4)$$

Eq. (3) can be solved analytically, assuming normalization of $W(u, \tau)$ such that for chaotic particle orientation $W(u, 0) = 1$:

$$W(u, \tau_{\gamma}) = e^{2\tau_{\gamma}}(u^2 + (1 - u^2)e^{2\tau_{\gamma}})^{-3/2}. \quad (5)$$

which, upon substituting W into (2) gives the particle orientation function in strong electric field of arbitrary shape due to the presence of polarizability anisotropy:

$$\Phi(\tau_{\gamma}) = \frac{1}{2} \left(\frac{3e^{2\tau_{\gamma}}}{e^{2\tau_{\gamma}} - 1} - \frac{3e^{2\tau_{\gamma}} \text{ArcTan} \sqrt{e^{2\tau_{\gamma}} - 1}}{(e^{2\tau_{\gamma}} - 1)^{3/2}} - 1 \right). \quad (6)$$

Dependence $\Phi(\tau_{\gamma})$ is presented in Fig. 1.

2.3. Orientation caused by permanent dipole moment

If particle orientation happens because of the presence of permanent dipole moment the moment of force $M = \mu E(t) \sin \Theta$ and, provided $\mu E(t)/kT \gg 1$ (1) expands into

$$2uW(u, \tau_{\mu}) + (u^2 - 1) \frac{\partial W(u, \tau_{\mu})}{\partial u} = \frac{\partial W(u, \tau_{\mu})}{\partial \tau_{\mu}}, \quad (7)$$

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