



Dynamic dielectric and magnetic properties of a spontaneously polarized single layer of the polar dielectric



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ABSTRACT

In the framework of the Debye–Smoluchowski theory and in local electric field approximation, an expression of the dielectric susceptibility tensor is derived for a single layer of spontaneously polarized polar dielectric in the field of a monochromatic radiation. It is shown that the AC radiation also generates or induces magnetic moments with a density that is expressed by the components of the dielectric susceptibility tensor of the layer.

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1. Introduction

In equilibrium, the polar groups of phospholipid molecules, located on opposite sides of the cell membrane, are oriented mirror asymmetrically to each other [1–3]. By the method of minimization of phospholipid bilayers free energy, it was shown the existence in [1] of a finite inclination angle and lower and upper limits of its variation. Further, it was shown in [4] that the inclination angle of the dipoles strongly depends of the ratio of water to phospholipid concentrations.

The existence of spontaneously aligned rigid dipoles makes the system a strongly anisotropic biaxial material with electromagnetic properties completely different from the properties of the nematic liquid crystals [3]. In present theories of dielectric permittivity of a lipid bilayer [5], the contributions of the inclination of dipoles in the electromagnetic properties of cell membrane are ignored. This paper aims to investigate the problem of the influence of the spontaneous polarization on the dynamic, dielectric properties of the cell membrane in the field of a homogeneous AC radiation and so to eliminate the drawback in existing modern theories.

This paper is arranged as follows. It contains the theoretical part, which includes Sec. 2, of the mathematical description of the spontaneous polarization of a single layer of polar dipoles. In Sec. 3, we solve the Debye–Smoluchowski kinetic equation in the

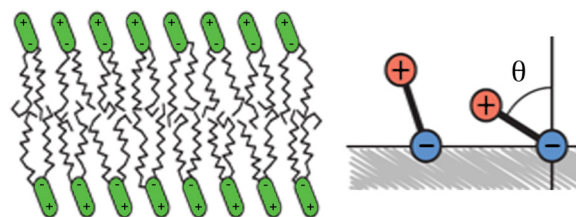


Fig. 1. Mirror asymmetrically arrangement of the phospholipid molecules and single rigid dipole with inclination angle θ .

presence of an AC external field. The polarization tensor of a single rigid dipole is found in Sec. 4, which is used in Sec. 5 to calculate the dielectric susceptibility in the local field approximation. In Sec. 6, we show a method of calculating the magnetic moment associated with the bounded dipole layer.

2. Spontaneous polarization

Because of the mirror symmetry arrangement of phospholipid molecules as seen in Fig. 1, we will consider single layers of spontaneously inclined dipoles.

The polarization of a rigid dipole is defined only by its direction \mathbf{n}

$$\mathbf{p} = p\mathbf{n}. \quad (1)$$

To handle mathematically the spontaneous alignment $\mathbf{p}^0 = p\mathbf{n}^0$ of the dipole parallel to the vector $\mathbf{n}^0 = (\sin\theta_0, 0, \cos\theta_0)$ in the

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plane xoz , we introduce a static, auxiliary electric field \mathbf{E}^0 acting in that direction

$$\mathbf{E}^0 = E^0 \mathbf{n}^0. \quad (2)$$

The dipole–field interaction U_0 is given by

$$U_0 = -\mathbf{p} \mathbf{E}^0. \quad (3)$$

The Boltzmann distribution of dipole moments in equilibrium at temperature T is

$$f^0 = C e^{-w_0(\mathbf{n}^0 \mathbf{n})}, \quad w_0 = pE^0/T, \quad (4)$$

where the coefficient C is defined from the normalization condition

$$\int f^0 d\mathbf{n} = 1 \quad (5a)$$

and it is expressed via a hyperbolic sine function

$$C = w_0/(4\pi \sinh w_0). \quad (5b)$$

To assure the correctness of the description of the spontaneous polarization, we consider the limit $T \rightarrow 0$ of expression (4) for the distribution function f^0 . Using the limit representation of the Dirac function

$$\delta(x) = \frac{1}{2} \lim_{\varepsilon \rightarrow \infty} \varepsilon e^{-\varepsilon|x|}$$

we obtain

$$\lim_{T \rightarrow 0} f^0 = \frac{1}{\pi} \delta(1 - \mathbf{n}^0 \mathbf{n}), \quad (6)$$

which indicates that at temperature $T = 0$, the dipoles are “frozen” parallel to \mathbf{n}^0 .

In equilibrium at the finite temperature T , the dipole (1) fluctuates around, \mathbf{n}^0 . We can find, using expression (4) for distribution function, that the average polarization $\langle \mathbf{p} \rangle_0$

$$\langle \mathbf{p} \rangle_0 = \mathbf{p}^0 L(w_0), \quad (7a)$$

where L is the Langevin function

$$L(x) = \coth x - 1/x. \quad (7b)$$

Our result (7) is consistent with the mean electric moment calculated in [6].

3. The Debye–Smoluchowski equation

We first introduce the interaction V of the dipoles with a homogeneous and monochromatic electric field with the amplitude \mathbf{E} and frequency ω

$$V = -\mathbf{E} \mathbf{d} e^{i\omega t}. \quad (8)$$

Then total field–dipole interaction U is given by

$$U = U_0 + V. \quad (9)$$

The distribution function of rigid dipoles f obeys the Debye–Smoluchowski equation [7,8]

$$\partial_t f - D \Delta f - \zeta^{-1} [\nabla f \cdot \nabla U + f \Delta U] = 0, \quad (10)$$

where D , ζ are the diffusion and the friction coefficients, respectively.

In the absence of a time dependent electric field E , the system is in equilibrium and $D = T/\zeta$ and the distribution function f is given by f^0 in (4). In the presence of the field E , we describe

the distortions from the equilibrium by introducing a function F , defined by the product,

$$f = F f^0. \quad (11)$$

We restrict our consideration in domains of small external electric fields and then, in the linear approximation of V , substitution of (11) in the Debye–Smoluchowski equation (10) gives us the equation for F

$$\zeta \partial_t F + \nabla U_0 \cdot \nabla F - T \Delta F = \Delta V - T^{-1} \nabla U_0 \cdot \nabla V. \quad (12)$$

Expanding F in the basis of spherical functions

$$F = 1 + e^{i\omega t} \sum_{l,m} a_{l,m} Y_{l,m}(\mathbf{n}) \quad (13)$$

we obtain a set of nonhomogeneous algebraic equations for the amplitudes $a_{l,m}$. Because of the strong oscillating character of higher harmonics of $Y_{l,m}$, we may consider the contributions of only the first four harmonics $Y_{0,0}$ and $Y_{1,m}$ and so we obtain the following expression for F of (12)

$$F = 1 + \frac{p(\mathbf{n} \mathbf{E})}{T(1 + i\omega\tau)} e^{i\omega t}. \quad (14)$$

In the absence of an initial polarization, that is, $w_0 = 0$, the response of the system to an external electric field is isotropic and that is equal to the corresponding Debye’s expression [7].

4. Polarization tensor

Consider the distribution function

$$f = f^0 + f^1. \quad (15)$$

For any quantity g , we introduce the notation $\langle g \rangle_{0,1}$ for the averages of g over f^0 and f^1 , respectively. We suppose f^0 to be normalized, i.e., $\langle 1 \rangle_0 = 1$ while the normalization integral of f^1 is $\langle 1 \rangle_1$, that is different of 1. In the linear approximation of f^1 , the average of g in the state (15) $\langle g \rangle$ is given by

$$\langle g \rangle = \langle g \rangle_1 - \langle g \rangle_0 \langle 1 \rangle_1. \quad (16)$$

Now, if we suppose the correction of f^1 to be proportional to f^0 (see (11) and (14)), then $f^1 = \lambda f^0$ and so

$$\langle g \rangle = \langle \lambda g \rangle_0 - \langle \lambda \rangle_0 \langle g \rangle_0 \quad (17)$$

for the average value of quantity g in linear approximation. In (17) $\langle \dots \rangle_0$ is the average in the 0-order distribution f^0 .

Defining the time dependent part of the dipole moment by

$$\langle \mathbf{p} \rangle(t) = \langle \mathbf{p}(\omega) \rangle e^{i\omega t},$$

where $\langle \mathbf{p}(\omega) \rangle$ is related to the polarization tensor α_{ij} by

$$\langle p_i(\omega) \rangle = \alpha_{ij}(\omega) E_j \quad (18)$$

and using (17) and (18), we find for the polarization tensor in the perturbed state (14) that

$$\alpha_{ij}(\omega) = \alpha(\omega) [\delta_{ij} + 2\beta n_i^0 n_j^0], \quad (19)$$

where

$$\alpha(\omega) = \frac{d^2 L}{w_0 T(1 + i\omega\tau)}, \quad (20)$$

$$2\beta = 3 + (w_0/L)(L^2 - 1) \quad (21)$$

and L is Langevin function (7b). In the limit $w_0 \rightarrow 0$, with $w_0^{-1} L \rightarrow 1/3$, we obtain

$$\alpha(\omega) \rightarrow \frac{d^2}{3T(1+i\omega\tau)} \quad \text{and} \quad \beta \rightarrow 0. \quad (22a)$$

The polarization tensor (19) of the free polar dipoles [7,8] becomes

$$\alpha_{ij}(\omega) \rightarrow \frac{d^2}{3T(1+i\omega\tau)} \delta_{ij}. \quad (22b)$$

On the other hand, in the limit of saturation, that is, $w_0 \rightarrow \infty$, $L \sim 1 - w_0^{-1}$ and $2\beta \rightarrow 1$, then $\alpha_{ij}(\omega) \rightarrow 0$ as

$$\alpha_{ij}(\omega) \sim \frac{d^2 w_0^{-1}}{T(1+i\omega\tau)} [\delta_{ij} + n_i^0 n_j^0]$$

i.e., no polarization in this limit (frozen dipoles).

5. Dielectric susceptibility

We consider a system of point rigid dipoles located on the plane xoy . We suppose the dipoles arranged in the form of a square lattice with lattice constant a and surface density N . The polarization is given by

$$P_i(\omega) = N\alpha_{ij}(\omega) E_j^{loc}(\omega), \quad (23)$$

where E^{loc} is the field of all other dipoles acting on a single dipole located at the origin of coordinates

$$E^{loc} = \mathbf{E} + \sum_{n,m \neq 0,0} \mathbf{E}_{n,m}. \quad (24)$$

The second term in (24) is calculated in [9] for square, triangular and honeycomb lattices. For the square lattice, in the nonretarded limit of the electromagnetic interaction, the x and y components of the scattered field are given by

$$\sum_{n,m \neq 0,0} (\mathbf{E}_{n,m})_{x,y} = 2gd_{x,y}(\omega), \quad (25)$$

where g is related to the lattice spacing a as follows

$$g = 4.51681a^{-3}. \quad (26)$$

Using the expressions of the local field (24), (25) and its connection with the polarization (23), we obtain

$$\hat{A}_{ij} E_j^{loc} = E_i, \quad (27)$$

where the matrix \hat{A} is given by

$$\hat{A} = \begin{bmatrix} 1 - 2g\alpha_{xx} & 0 & -g\alpha_{zx} \\ 0 & 1 - 2\alpha_{yy} & 0 \\ -g\alpha_{zx} & 0 & 1 + g\alpha_{zz} \end{bmatrix}. \quad (28)$$

Solving equation (27), we find the local electric field acting on a dipole in the lattice. Then, using definition (23), we find the following nonzero diagonal components for the electric susceptibility χ_{ij}

$$\begin{aligned} \chi_{yy} &= N\alpha(1 - 2\alpha g)^{-1} \\ \chi_{xx} &= \chi [(\gamma - \cos 2\theta_0)(z + \gamma + \cos 2\theta_0) - \sin^2 2\theta_0] \\ \chi_{zz} &= \chi [(\gamma + \cos 2\theta_0)(z - 2\gamma + 2\cos 2\theta_0) - \sin^2 2\theta_0]. \end{aligned} \quad (29a)$$

The susceptibility tensor is not symmetric and it has only two nonzero off-diagonal elements, viz.

$$\begin{aligned} \chi_{zx} &= \chi z \sin 2\theta_0 \\ \chi_{xz} &= \chi(z - 3\gamma + 3\cos 2\theta_0) \sin 2\theta_0, \end{aligned} \quad (29b)$$

where we have used the definitions

$$\gamma = 1 + \beta^{-1}, \quad z = (\alpha\beta g)^{-1}$$

and

$$\chi = Ng^{-1} [3\cos^2 2\theta_0 + 3z\cos 2\theta_0 + (z - 2\gamma)(z + \gamma) - 1]^{-1}.$$

In the absence of spontaneous polarization, that is, $w_0 \rightarrow 0$, we see that χ_{xx} is equal to χ_{yy} , i.e., the symmetry in the plane xoy is recovered. In the limit of strong saturation, that is, $w_0 \rightarrow \infty$, $\chi_{ij} \rightarrow 0$ as is the case for the polarization tensor α_{ij} (19).

6. Magnetization

There is a restriction in the polar dipole system under consideration, viz., the dipoles are not completely free but one end of the dipole is bound to the surface of the membrane. This fact allows lipid bilayers magnetic properties in the field of microwave radiation: the free end of the dipole acts as a rigid charge rotor, which carries a magnetic moment. The accurate theory of these phenomena should be based on Kapitza's method of solution governing the equations for the rigid rotor in the field of a microwave radiation [10,11]. However, we can find the magnetization of the single layer of the spontaneously polarized polar dielectric directly from the expression for the dielectric susceptibility (28).

To find the trajectory of the free end of the dipole, we go from the limit of point dipole to extended dipole moments having the same dipole moment. Suppose the free end of the dipole carries a charge q , then we can define the trajectory by using the definition of the dipole moment

$$x_i(t) = q^{-1} \text{Re}\{d_i(\omega) e^{i\omega t}\}. \quad (30)$$

We define the vector \mathbf{S} of the trajectory enclosed in an area by the expression

$$S_i = \frac{1}{2} \oint \varepsilon_{ijl} x_j \dot{x}_l, \quad (31)$$

where ε_{ijl} is the three-dimensional Levi-Civita symbol. Then using (30), we find for the z component of \mathbf{S}

$$S_z = \pi q^{-2} \text{Im}[d_x(\omega) d_y^*(\omega)]. \quad (32)$$

The remaining components of \mathbf{S} can be obtained by a cyclic permutation of the indices in (32).

We define the magnetization vector \mathbf{M} in analogy to the set of periodic loop currents with surface density N

$$\mathbf{M} = q\omega N \mathbf{S}. \quad (33)$$

Then, using (28) and (32) we find for M_z

$$M_z = m_{ij} \text{Im}[\chi_{xi} \chi_{yj}^* e^{i\delta_{ij}}], \quad (34)$$

where δ_{ij} is the phase difference of i -th and j -th components of the complex amplitudes E_i and E_j . In (34) we define the tensor

$$m_{ij} = \pi\omega(Nq)^{-1} |\alpha|^2 |E_i E_j|. \quad (35)$$

The remaining components of \mathbf{M} can be obtained from (34) by a cyclic permutation of the indices x , y , and z .

7. Summary

The polar groups of phospholipid molecules, located on opposite sides of the cell membrane, are spontaneously polarized and thus bringing additional electrical and magnetic properties of the membrane in the field of a microwave radiation. Owing to the mirror symmetry of arrangement of phospholipid molecules, the lipid bilayer can be considered as a combination of two independent

single layers of spontaneously inclined dipoles. In the framework of the Debye–Smoluchowski theory and in local electric field approximation, an expression of the dielectric susceptibility tensor is derived for a single layer of spontaneously polarized polar dielectric in the field of monochromatic radiation. It is shown that the AC radiation stimulates also magnetic moments with a density that is expressed by components of the dielectric susceptibility tensor of the layer.

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