LIGHT SCATTERING INVESTIGATION OF ELECTRIC FIELD ALIGNMENT OF PHOSPHOLIPID TUBULES

ZILI LI,* CHARLES ROSENBLATT,* PAUL YAGER,‡ AND PAUL E. SCHOEN§
*Department of Physics, Case Western Reserve University, Cleveland, Ohio 44106; ‡Department of Bioengineering, University of Washington, Seattle, Washington 98195; and §Bio/Molecular Engineering Branch, Naval Research Laboratory, Washington, DC 20375

ABSTRACT The polymerizable diacetylenic phospholipid 1,2-bis(10,12,tricosadiynoyl)-*sn*-glycero-3-phosphocholine ($DC_{23}PC$) forms straight hollow cylinders in water. Using an ac electric field it was possible to achieve significant orientational alignment of the tubules parallel to the field direction, and from light scattering results deduce an effective dielectric susceptibility anisotropy $\Delta\chi_E$. Moreover, we suggest that the alignment arises from an orientational anisotropy of the total electrostatic enthalpy for a dielectric tubule in an electric field, rather than an inherent polarizability anisotropy of the constituent $DC_{23}PC$ molecules, as was the case with magnetic field alignment.

INTRODUCTION

It was recently discovered that the lecithin 1,2-bis(10,12,tricosadiynoyl)-*sn*-glycero-3-phosphocholine ($DC_{23}PC$) forms a microstructure consisting of bilayers which wrap around a hollow core (Yager and Schoen, 1984; Yager et al., 1985; Singh and Schnur, 1985; Georger et al., 1987; Singh et al., 1986). These tubules are generally straight, tens of microns long, ~0.75 μm in diameter, and with wall thicknesses ranging from 1 to 10 bilayers (Yager et al., 1985). The polymerized tubules offer a host of possible applications, and it is therefore important to both understand their interactions and be able to manipulate them in aqueous suspension. Recently, for example, a magnetic birefringence experiment was performed (Rosenblatt et al., 1987), demonstrating near complete orientational alignment in fields of 20–40 kg.

Although the magnetic technique is indeed powerful, in many instances it is more practical to apply electric fields. Nicoli et al. (1981), for example, used transient electric birefringence to study the anisometry of micelles. Similar measurements have been performed on cartilage proteoglycans (Hawkins et al., 1978) and collagen (Bernengo et al., 1974), and electric field–dependent light scattering has been used to orient and study *Escherichia coli* (Morris et al., 1979). In a classic work Pohl (1978) studied electric field–induced colloid aggregation (pearl chaining), and recently workers have used time-dependent field induced rotation to study the electrical properties of cell membranes (Holzapfel et al., 1982). Thus, although electric and magnetic fields often play similar roles in manipulation of microscopic objects, each has advantages lacking in the other. Magnetic fields tend to be noninvasive, whereas electric fields can be switched rapidly and are more easily accessible both inside and outside the laboratory.

The purpose of this paper, then, is to report on the electric field–induced orientation of $DC_{23}PC$ tubules as determined by light scattering. Our central result is that significant alignment is obtained in moderate ac fields at 20–30 V/cm, although the scattering pattern becomes noticably turbulent above this value. We conclude that the alignment arises from the orientational anisotropy of the total electrostatic enthalpy for a dielectric tubule in an electric field, rather than an inherent polarizability anisotropy of the $DC_{23}PC$ molecules, the case with magnetic field alignment (Rosenblatt et al., 1987).

EXPERIMENTAL

The lipid 1,2-bis(10,12-tricosadiynoyl)-*sn*-glycero-3-phosphocholine was synthesized and tubules prepared according to previously published methods (Johnston et al., 1980); our sample comes from the same batch as used earlier in the magnetic measurements (Rosenblatt et al., 1987). The added water was deionized and distilled. Care was taken to minimize the ionic strength to avoid unwanted joule heating and spurious polarization effects (O'Konski and Haltner, 1957). To obtain the distribution of tubule lengths, a small amount of sample was placed between a microscope slide and cover slip. Several photographs were taken using an Olympus BHA microscope (Olympus, Tokyo) in phase-contrast mode. The length distribution was obtained from 103 individual micrographs, were the mean length $\bar{L}$ was found to be 14.8 μm, and the rms deviation 5.9 μm. This value of $\bar{L}$ is smaller than that obtained previously, most likely due to breakage of tubules during shipping. Nevertheless, the wall thickness distribution should be the same as published earlier (Rosenblatt et al., 1987), where the average thickness $t \simeq 0.0125$ μm (2.5 bilayers). The average outer radius b of the tubules is again 0.375 μm, and the inner radius a is, of course, $b - t$.

The sample was housed between two microscope slides 2.5 cm wide and 7.5 cm in length, separated by a pair of parallel wires of radius $R = 0.041$ cm running the length of the slides. The wires, which were spaced a distance $D = 0.87$ cm apart, served the dual role of spacers and electrodes.

Address correspondence to Charles Rosenblatt.

For this configuration, the capacitance per length C of the system is given by (Jackson, 1975).

$$C = \epsilon_w \left[2 \cosh^{-1} \left(\frac{D^2 - 2R^2}{2R^2} \right) \right]^{-1} \quad (1)$$

where ϵ_w is the dielectric constant of the medium (water). One can easily show that for an applied potential V (in volts), the resulting field $\mathbf{E}_0$ (in V/cm) midway between the wires is given by $\mathbf{E}_0 = 0.76 \times V$. The voltage was supplied by a General Radio Corp. (Concord, MA) model 1210-C af generator driving a McIntosh (Binghamton, NY) model MC75 audio amplifier at 2 kHz. All voltages and fields used in this paper are rms values.

Light from a He-Ne laser (model 124B, Spectra-Physics Inc., Mountain View, CA) first passed through a light chopper (model 5R-540, Stanford Research Systems, Inc., Palo Alto, CA) operating at 318 Hz, then through the center of the sample midway between the electrodes. The main beam was then intercepted by a beam stop, and the scattered light was detected by means of a photodiode (model HUV1000B, EG & G, Salem, MA) situated an angle 6.25° from the beam axis, such that the scattering wave vector $\mathbf{Q}$ and the applied electric field $\mathbf{E}_0$ were parallel to each other. For this geometry $|\mathbf{Q}| = (10{,}800 \pm 1{,}000)\ \mathrm{cm}^{-1}$, where the spread in $\mathbf{Q}$ arises from the acceptance angle of the detector. The detector output was fed into a PAR model 5101 lock-in amplifier (Princeton, NJ), referenced to the light chopper. The output from the lock-in, proportional to the intensity at the detector, was then read simultaneously by two Keithley Instruments, Inc. (Cleveland, OH) digital volt meters models 192 and 195A, in data logging mode. The two DVMs allowed us to collect data over two different time scales.

The sample holder was first half-filled with water to determine the background scattering intensity due to imperfections in the glass and other unwanted sources. At this point both DVMs were zeroed. The sample holder was then further filled with an aqueous suspension of tubules, so that the final concentration, as determined by serial dilution of an initial stock, was $\rho = 0.25$ mg/ml. After agitating with a thin piece of mylar, the sample was sealed with epoxy.

At zero applied field the output voltage from the lock-in (proportional to the scattered intensity) was recorded for 15 s at 1-s intervals to obtain an average scattering intensity $I(\mathbf{E}_0 = 0)$. The ac field was then switched on, and the second DVM recorded the voltage from the lock-in at 5-s intervals for ~150 s. The first dozen data points, which correspond to the transient intensity as the tubules approached an equilibrium orientational distribution in the applied field, were discarded. The remaining voltages were averaged to obtain $I(\mathbf{E}_0)$. The voltage was then switched off and the tubules allowed to randomize for 5 min (approximately five decay times, typical of what is expected for particles of this size [Rosenblatt et al, 1987]). The process was repeated twice more to obtain $I(\mathbf{E}_0)/I(0)$ at field $\mathbf{E}_0$, and the three values averaged. Here $I(0) = I(\mathbf{E}_0 = 0)$. (Obtaining $I(0)$ just before each measurement of $I(\mathbf{E}_0)$ excludes artifacts due to concentration gradients which vary slowly in time and to sedimentation, which occurs extremely slowly relative to the experiment.) The entire procedure was performed at several values of $\mathbf{E}_0$ up to 57 V/cm, and the results are shown in Fig. 1, where the error bar represents the rms deviation obtained from averaging the three values of $I(\mathbf{E}_0)/I(0)$.

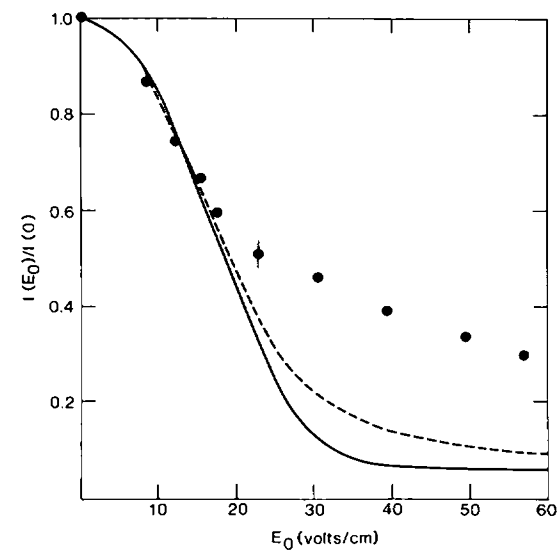

FIGURE 1 The experimental ratio $I(\mathbf{E}_0)/I(0)$ vs. $\mathbf{E}_0$. Error bar represents deviation from the average of three measurements at each field. Solid line represents a monodisperse fit to the data using the first four data points and Eq. 9. Broken line represents a polydisperse fit using the first four data points. Eq. 9, and the measured length distribution.

RESULTS AND DISCUSSION

Before proceeding with the full analysis, we must first determine whether the concentration ρ was, in fact, in the dilute regime (in which the tubules are orientationally uncorrelated), or whether excluded volume effects need be considered. Based upon an Onsager (1949) model for hard right circular cylinders taken to the level of the second virial coefficient, we find that the dilute approximation is valid (Photinos and Saupe, 1985; Rosenblatt and Zolty, 1985) if

$$n\pi\bar{L}^2 b/16 \ll 1,$$

where n is the number density of tubules. For the values at hand, $n = 5.7 \times 10^8\ \mathrm{cm}^{-3}$, and the left side is equal to 0.018; thus, the dilute approximation is, indeed, valid.

In the earlier magnetic field problem, it was shown (Rosenblatt et al., 1987) that tubules could be oriented in fields of order 20 to 30 kG. The magnetic susceptibility anisotropies of the individual molecules are additive, and the total magnetic anisotropy of the tubule $V\Delta\chi_M$ could be sufficiently large such that $\frac{1}{2}V\Delta\chi_M H^2 > k_B T$. Here, V is the volume of lipid in the tubule, $\Delta\chi_M$ the volumetric magnetic susceptibility anisotropy, H the applied field, k_B Boltzmann's constant, and T the temperature. As an example, for tubules of volume $V = 4.4 \times 10^{-13}\ \mathrm{cm}^3$ (i.e., 14.8 μm long) and $\Delta\chi_M = 7 \times 10^{-9}$ cgs (Rosenblatt et al., 1987) in a field of 10^4 G, we find the magnetic energy term equals 1.5×10^{-13} ergs, which is several times the thermal energy. Thus significant alignment is readily achieved by coupling a magnetic field to the susceptibility anisotropy.

In an electric field, however, the mechanism is different. Values for the anisotropy of the local dielectric susceptibility are, unfortunately, not available in the literature, although we can liberally assume a value $\Delta\chi_E = \Delta\epsilon/4\pi \sim 0.25$. Here $\Delta\epsilon$ is the dielectric tensor anisotropy. (Note that smaller values would result in even less alignment.) Then, comparing the quantity $\frac{1}{2}V\Delta\chi_E \mathbf{E}_0^2$ with $k_B T$ for a typical electric field of 0.1 statvolt/cm (30 V/cm), we find the electrostatic term to be some two orders of magnitude smaller than thermal energies, and therefore insufficient to produce the degree of order observed in the experiment. Instead we invoke another mechanism in which tubules of isotropic dielectric constant ϵ are suspended in water of dielectric constant $\epsilon_W = 79$ and are oriented either parallel ($\|$) to an external electric field or perpendicular ($\perp$). The relevant thermodynamic potential, the electrostatic enthalpy H (Callen, 1960), is calculated from Laplace's equation for the two configurations, and the enthalpy associated with an arbitrary orientation of tubules at angle θ relative to the field can be obtained by superposition. This is the classic "depolarization" problem of dielectrics in an applied field (Jackson, 1975) in which the orientation obtains from the anisotropy of the electrostatic enthalpy of an anisometric but otherwise isotropic dielectric.

We first consider a tubule of length L ($L \gg b$) placed parallel to an external field $\mathbf{E}_0 z$. Because the depolarization factor for a long hollow pipe is near zero in this geometry, the field $\mathbf{E}$ inside the tubule is parallel and equal to $\mathbf{E}_0$. In general, the enthalpy H due to a dielectric in an external field $\mathbf{E}_0$ is given by (Jackson, 1975; Callen, 1960).

$$H = -\frac{1}{8\pi}\int_V (\epsilon - \epsilon_W)\mathbf{E} \cdot \mathbf{E}_0 \, dV, \tag{2}$$

where the integral is over the volume of the tubule only. Thus, for this geometry

$$H_\| = -\tfrac{1}{8}(\epsilon - \epsilon_w)\mathbf{E}_0^2 L(b^2 - a^2). \tag{3}$$

For a tubule oriented parallel to the $\hat{y}$-axis and perpendicular to the external field $\mathbf{E}_0 z$, Laplace's equation is used to calculate the field in all space. If we assume that $L \gg b$ and therefore neglect end effects, we find within the tubule

$$\mathbf{E} = B\hat{z} - \frac{Ca^2}{r^2}(\hat{z}\cos 2\phi - \hat{x}\sin 2\phi), \tag{4a}$$

where r and ϕ are polar coordinates in the xz plane,

$$B = \mathbf{E}_0 \frac{2}{1 + \epsilon'}\frac{1}{\Delta} \tag{4b}$$

and

$$C = \mathbf{E}_0 \frac{2(\epsilon' - 1)}{(\epsilon' + 1)^2}\frac{1}{\Delta}. \tag{4c}$$

The quantity ϵ' is defined as ϵ/ϵ_w and

$$\Delta = 1 - \left(\frac{a}{b}\frac{1 - \epsilon'}{1 + \epsilon'}\right)^2. \tag{4d}$$

H can easily be calculated from Eq. 2 and is given by

$$H_\perp = -\frac{1}{8}(\epsilon - \epsilon_w)\mathbf{E}_0^2 L(b^2 - a^2)\frac{2}{1 + \epsilon'}\frac{1}{\Delta}. \tag{5}$$

Thus, neglecting end effects, the difference in enthalpy ΔH for the two configurations can be obtained by subtracting Eq. 5 from Eq. 3. Assuming $\mathbf{E}_0 = 0.1$ statvolt/cm, $L = \bar{L} = 14.8$ μm, and $\epsilon = 3$, we find $\Delta H \simeq -1.2 \times 10^{-13}$ ergs. This value is negative, which means that the perpendicular orientation is more "costly," and thus the tubules align parallel to the field. Moreover, ΔH is several times $k_B T$, which means a signficant degree of orientational order can be expected from this mechanism. It should be noted that ΔH is very insensitive to the actual value of ϵ (up to $\epsilon \sim 25$) owing to the large value of ϵ_w. Thus, even if the tubules were extremely polar, these and subsequent results would still be reasonably quantitatively correct. Nevertheless, it's unlikely that even the polar head groups could result in values of ϵ of order 25.

For a tubule oriented at some angle θ relative to the external field, the enthalpy is given by $H = H_\| \cos^2\theta + H_\perp \sin^2\theta = H_\perp + (H_\| - H_\perp)\cos^2\theta$. Then, in analogy to the magnetic field case, we can write the orientational part of H in the form $\Delta H = -\tfrac{1}{2} V \Delta\chi_E \mathbf{E}_0^2 \cos^2\theta$, where $\Delta\chi_E$ can be equated with (cf. Eqs. 3 and 5)

$$\Delta\chi_E = \frac{-1}{4\pi}(\epsilon - \epsilon_w)\left(\frac{2}{1 + \epsilon'}\frac{1}{\Delta} - 1\right). \tag{6}$$

This, of course, neglects end effects; if end effects were included, $\Delta\chi_E$ would be expected to exhibit some length dependence, especially for the shorter tubules. Nevertheless, Eq. 6 is an instructive form for comparison with magnetic fields. The remainder of this paper, then, will describe a light scattering experiment from which we extract the quantity $\Delta\chi_E$.

Modeling the tubule as a hollow cyclinder, we find in the Rayleigh-Gans approximation (van de Hulst, 1981) that the form factor is given by (Rosenblatt et al., 1982).

$$S(\mathbf{Q}, \theta) = \left\{\frac{\sin\left(\frac{\mathbf{Q}L\cos\theta}{2}\right)[bJ_1(\mathbf{Q}b\sin\theta) - aJ_1(\mathbf{Q}a\sin\theta)]}{\mathbf{Q}L\cos\theta\,\mathbf{Q}(b^2 - a^2)\sin\theta}\right\}^2, \tag{7}$$

where J_1 is the Bessel function of order 1 and θ is the angle between the tubule axis and $\mathbf{Q}$. The Rayleigh-Gans approximation consists of two conditions, $|m - 1| \ll 1$ and $4\pi\delta|m - 1|/\lambda \ll 1$. Here m is the ratio of the refractive indices, λ the wavelength of light, and δ a characteristic dimension of the particle through which the light passes. Although a numerical value for m is unavailable, for most organic and biological materials we can take its value as $1 \leq m \leq 1.2$, and thus the first condition holds reasonably, although not extremely well. For the second condition, we note that the light passes through the tubule walls, this being the dimension used for δ. Taking $\delta \sim 300$ A, we find that $4\pi\delta|m - 1|/\lambda \sim 0.1$, again small, although not totally insignificant with respect to 1. Thus, although the Rayleigh-Gans approximation is used in the analysis, better results would obtain if a full Mie calculation were performed (van de Hulst, 1981). To our knowledge, however, such a calculation has never been done for this geometry.

For an orientational-dependent enthalpy $-\frac{1}{2}V\Delta\chi_E E_0^2 \cos^2\theta$, the orientational distribution function $f(\mathbf{E}_0, \theta)$ is given by

$$f(\mathbf{E}_0, \theta) = \frac{\exp(\frac{1}{2}V\Delta\chi_E \mathbf{E}_0^2 \cos^2\theta / k_B T)}{\int_{-1}^{1} \exp(\frac{1}{2}V\Delta\chi_E \mathbf{E}_0^2 \cos^2\theta / k_B T)\, d(\cos\theta)}. \quad (8)$$

Note that since $\mathbf{Q}$ and $\mathbf{E}_0$ were chosen to be parallel to each other, the angle θ is used not only in the form factor (Eq. 7), but in the distribution function (Eq. 8) as well. As a result, the intensity thus measured at $\mathbf{Q}$ is simply

$$I(\mathbf{E}_0, \mathbf{Q}) \propto \int S(\mathbf{Q}, \theta) f(\mathbf{E}_0, \theta)\, d(\cos\theta). \quad (9)$$

Because the tubules are not monodisperse in size, Eq. 9 also involves an integral over the length distribution (cf. Eqs. 7 and 8). We will thus analyze the data in two steps. First, we'll substitute the average tubule length $\bar{L} = 14.8$ μm for L, and the average volume $\bar{V} = 4.4 \times 10^{-13}$ cm^3 for V into the preceding equations, thereby fitting the data to a monodisperse size distribution. Eq. 9 will then be modified to include the tubule length distribution to obtain what should be a more accurate value of $\Delta\chi_E$ (Rosenblatt et al., 1987; Lewis et al., 1985). Note, however, that $\Delta\chi_E$ is not really a constant, but owing to end effects $\Delta\chi_E$ actually depends on the length to diameter ratio. Because the tubules are relatively long, however, we will neglect end contributions and assume that the orientational enthalpy ΔH scales as $V\Delta\chi_E$, where $\Delta\chi_E$ is constant.

When the field was switched on the tubules aligned preferentially along $\mathbf{E}_0$; the scattering thus changed from a symmetric pattern to a long, narrow pattern oriented perpendicular to $\mathbf{E}_0$ (Fig. 2). At the detector the intensity decreased, as evidenced from the data points in Fig. 1. Moreover, at higher fields ($\mathbf{E}_0 \geq 20$ V/cm) the scattering, as observed visually using a white screen, exhibited more and more turbulence. At this time it's not clear whether

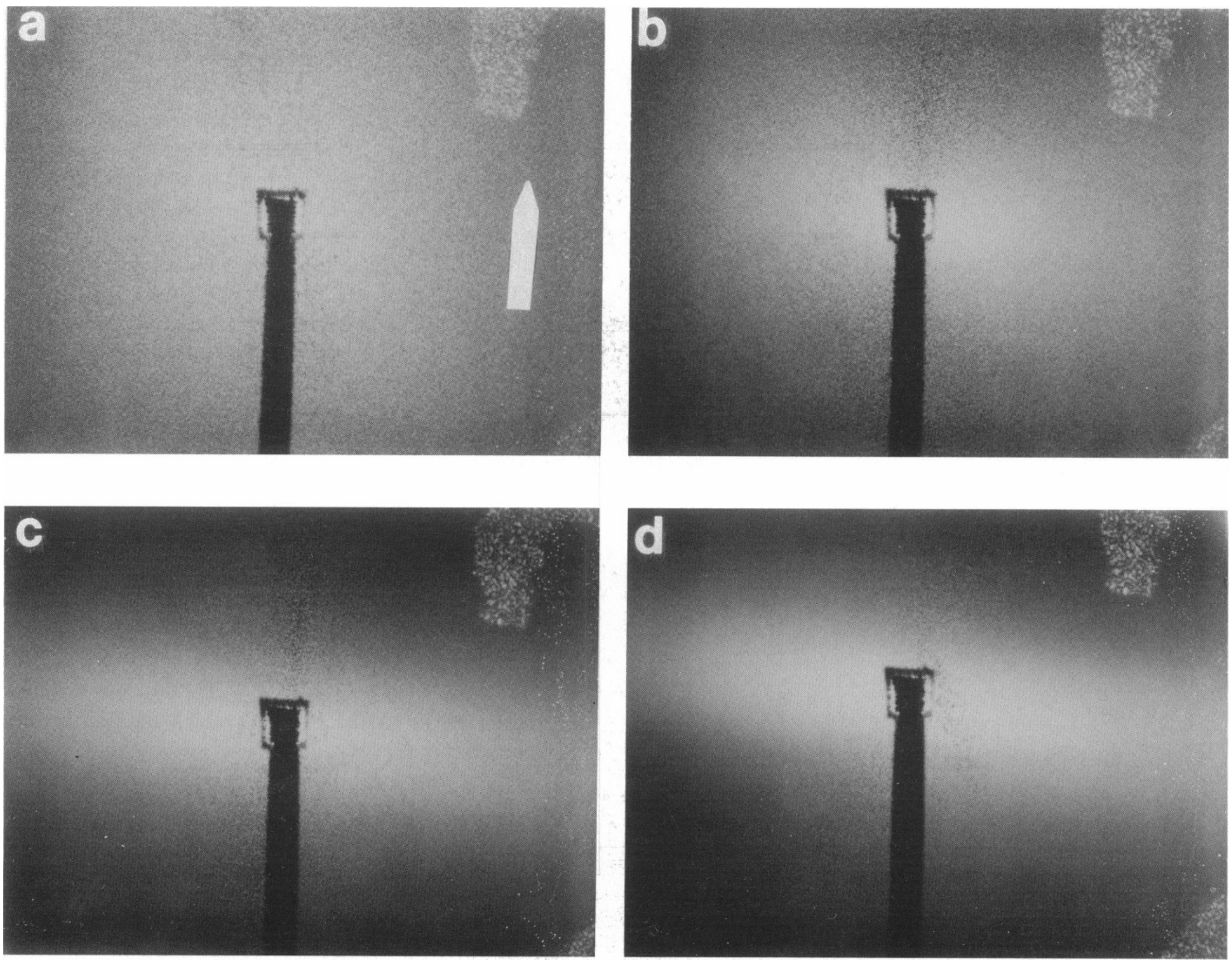

FIGURE 2 Photographs of the scattering pattern at four different fields: (*a*) $\mathbf{E}_0 = 0$, (*b*) $\mathbf{E}_0 = 19.0$ V/cm, (*c*) $\mathbf{E}_0 = 38.0$ V/cm, and (*d*) $\mathbf{E}_0 = 57.0$ V/cm. Direction of arrow indicates the direction of both the applied field $\mathbf{E}_0$ and scattering wavevector $\mathbf{Q}$, and length of arrow corresponds to a magnitude $|\mathbf{Q}| = 10{,}000$ cm^{-1}. The dark central spot is the beam stop.

this turbulence was due to electrohydrodynamic instabilities, joule heating of the sample, or dielectrophoresis. Nevertheless, at higher fields the concomitant stirring of the sample competed with the field-induced alignment, resulting in a smaller degree of orientational order than would otherwise be expected. (The possibility of tubules pinned to the walls with random orientations would also increase $I(\mathbf{E}_0)/I(0)$, although not enough to be significant, because the sample thickness $2R = 0.082$ cm $\gg L$). We thus decided to perform a least squares fit of the experimental data for $I(\mathbf{E}_0)/I(0)$ to the theoretical form in Eq. 9 using a range shrinking technique. The first three values of field were used in the fit, then the first four, then five, and so on. Using the monodisperse fit we found an evolution in $\Delta\chi_E$ from 100 cgs to 59, and chose the value $\Delta\chi_E = 93$ cgs obtained using the first four data points as the "correct" value. (This should be compared with a theoretical value of ~60 cgs obtained from Eq. 6.) The resulting fit is shown by the solid line in Fig. 1. Note that even in this case a small systematic deviation between theory and experiment is present, due in part to the presence of a small amount of turbulence even at low fields, and in part to the monodisperse approximation used in the fit. Moreover, note that unlike the birefringence results, the intensity ratio is not expected to scale as $\mathbf{E}_0^2$, except at extremely low fields, such that $\Delta H \ll k_B T$. We then refitted the data to Eq. 9 by including the length distribution, where the intensity was weighted by the fraction of tubules having length L and volume V. The range of $\Delta\chi_E$ values was shifted upward, where we found $\Delta\chi_E = 121$ cgs from the first four data points. Using this as the "correct" value, the polydisperse fit to Eq. 9 is shown by the broken line in Fig. 1.

It's instructive to use the experimental value of $\Delta\chi_E = 121$ cgs to calculate the orientational enthalpy $-\frac{1}{2}V\Delta\chi_E\mathbf{E}_0^2$ at a field $\mathbf{E}_0 = 0.1$ statvolts/cm and volume $V = 4.4 \times 10^{-13}$ cm^3. We find a value -2.6×10^{-13} ergs. When compared with the theoretical calculation using Eqs. 3 and 5, we find that they differ by a factor of approximately two. Given some of the approximations used in both the theoretical calculation (neglect of end effects and turbulence) and the fit (the Rayleigh-Gans approximation), and given that the theory was intended solely as a plausibility argument for the dielectric mechanism, the agreement is not at all unreasonable. This is especially true because the inherent polarizability anisotropy mechanism (as in the magnetic case) was shown to result in an enthalpy some two orders of magnitude smaller.

We can take the experimental result for $\Delta\chi_E$ one step further. The orientational order parameter S is normally defined as $\langle \frac{3}{2}\cos^2\theta - \frac{1}{2} \rangle$, where the brackets represent a thermal average. For $\mathbf{E}_0 = 0.05$ statvolts/cm (15 V/cm), we calculate $S = 0.24$ using a value $\Delta\chi_E = 121$ cgs. For $\mathbf{E}_0 = 0.1$ statvolt/cm, the calculated value of S is 0.73, although clearly the experimental value is smaller than the calculated value due to the presence of a turbulence at higher fields.

We had previously used another method, namely birefringence (Rosenblatt et al., 1987), to determine the orientational order parameters. This method requires either a reliable form for the distribution function up to nearly saturated order, which we do not have owing to turbulence, or an experimental value of the saturated birefringence Δn_0. We can obtain Δn_0 from the magnetic measurements, and then measure $S = \Delta n(\mathbf{E}_0)/\Delta n_0$ directly. This work, which nicely complements the light scattering data, is currently underway.

To summarize, we have performed electric field alignment of lipid tubules, finding that significant alignment ($S \sim 0.5$) is attainable at reasonably small fields. As the field is increased much above 20 V/cm, however, turbulence becomes important and reduces the alignment from what would be expected from the field alone. Undoubtedly, even higher electric fields would yield alignments comparable with that of magnetic fields in the neighborhood of several tens of kilogauss, giving us another method by which we can manipulate this system.

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