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Orientational Ratchets and Angular Momentum Balance in the Janossy Effect

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In the Janossy effect, the threshold intensity for the optical Freedericksz transition in nematic liquid crystals is dramatically reduced by the addition of a small amount of dichroic dye to the sample. We examine the source of torques in the system and the balance of angular momentum. We show that, due to the presence of the dye, the system is an optically pumped orientational ratchet.

Keywords: nonlinear optics; liquid crystals; dye; orientational ratchet; Brownian motor; Janossy effect; angular momentum balance

INTRODUCTION

In the optical Freedericksz transition, liquid crystals undergo a configurational transition when irradiated with polarized light[1]. The torque $\mathbf{\Gamma} = \mathbf{D} \times \mathbf{E}$ exerted by the light on the nematic liquid crystal overcomes the restoring elastic torque if the intensity is sufficiently high, and a director deformation takes place. It has been shown by Janossy and coworkers that when a small amount of dichroic dye is dissolved in the liquid crystal, the threshold intensity for the optical Freedericksz transition is reduced by some two orders of magnitude[2]. Since in this case the direct optical torque exerted by the radiation field on the liquid crystal is not sufficient to overcome the elastic restoring torque, it is interesting to identify the

mechanism from which the torque which distorts the liquid crystal arises. A model for the process has been proposed by Janossy[3], and it has been further considered by Marrucci and Paparo[4]. In this paper we concentrate on aspects not fully discussed in Ref.[3], and examine the details of angular momentum balance in the system. We argue that the system is an optically pumped orientational ratchet.

DIRECT OPTICAL FORCES AND TORQUES

We first consider the direct optical forces and torques from the radiation field which act on the liquid crystal. In addition to considering the usual torque density in anisotropic transparent media, we ask if direct contributions to these arising from absorption by the dye are significant.

Body forces and torques acting on matter can be obtained from the stress tensor. Although the appropriate form of the stress tensor in condensed matter due to electromagnetic radiation is still the subject of some discussion[6, 8], we use the Minkowski stress

$$\sigma_{\alpha\beta} = -E_\alpha D_\beta - H_\alpha B_\beta + \frac{1}{2}\delta_{\alpha\beta}(\mathbf{E}\cdot\mathbf{D} + \mathbf{H}\cdot\mathbf{B})$$

and the momentum density

$$\mathbf{g} = \mathbf{D}\times\mathbf{B} \tag{1}$$

which is widely used[6], and which is believed to be valid in absorbing media if the dielectric tensor is symmetric[7]. The body force is

$$f_\alpha = -\frac{\partial\sigma_{\alpha\beta}}{\partial x_\beta} - \frac{\partial g_\alpha}{\partial t} \tag{2}$$

and substitution gives

$$\mathbf{f} = -\nabla\cdot\frac{1}{2}(\mathbf{E}\cdot\mathbf{D} + \mathbf{B}\cdot\mathbf{H}) \tag{3}$$

since for optical fields the term $-\frac{\partial g_\alpha}{\partial t}$ averages to zero. To lowest order, the bulk torque density in the sample is the antisymmetric part of the stress tensor; that is,

$$\Gamma_\alpha = \varepsilon_{\alpha\beta\gamma}\sigma_{\beta\gamma} \tag{4}$$

and substitution gives for the direct optical torque

$$\mathbf{\Gamma}_{opt} = \mathbf{D} \times \mathbf{E} \tag{5}$$

for nonmagnetic materials.

In the optical Freedericksz transition of transparent nematics, this direct optical torque is balanced by the elastic torque

$$\mathbf{\Gamma}_{el} = K\hat{\mathbf{n}} \times \nabla^2 \mathbf{n} \tag{6}$$

where K is an elastic constant, and $\hat{\mathbf{n}}$ is the nematic director. A simple schematic of the optical Freedericksz transition is shown below (Fig. 1).

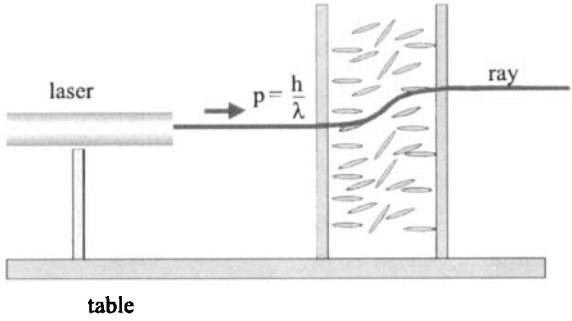

FIGURE 1 The optical Freedericksz transition in non-absorbing nematics. Due to anisotropy, **D** is not parallel to **E**, and the ray is deflected. Angular momentum is transferred from the light to the nematic; no energy is absorbed.

In the Janossy effect in dyed nematics, the elastic torque remains essentially the same, since the elastic constant K is not significantly effected by the presence of a small amount of dye[3]. However, since the threshold intensity for the transition is reduced by some two orders of magnitude, the direct optical torque is correspondingly reduced. Since the material is absorbing, the fields are different from those in transparent materials. Writing the optical torque in terms of complex field amplitudes gives

$$\mathbf{\Gamma}_{opt} = \frac{1}{2}\mathrm{Re}[e^{-i2\mathbf{k}_i \cdot \mathbf{r}}(\mathbf{D}_o \times \mathbf{E}_o^*)] \tag{7}$$

where $\mathbf{k}_i$ is the imaginary part of the wave vector. We note that, in addition to the usual contribution from the real part of the dielectric tensor, there will be an additional contribution due to the imaginary part arising from absorption by the dye. The relative magnitude of this contribution to the torque is on the order of $(k_i/k_r)^2$, where k_r is the real part of the wave vector, which, for a weakly absorbing sample, is negligible. Thus the direct optical torque, even including the effects of absorption, is not sufficient to bring about the observed transition.

In addition to the torque, there will be a direct body force acting on the sample due to absorption, given by

$$\mathbf{f}_{opt} = \mathbf{k}_i \frac{In}{2c} \tag{8}$$

where I is the intensity and c/n is the speed of energy propagation. In principle, this force can give rise to flow, which could result in a distortion of the director field due to shear alignment. For a deformation to take place, the shear torque $\Gamma_{shear} \simeq f_{opt} w$, where w is the relevant length for shear, must overcome the restoring elastic torque Γ_{el}. Taking w to be the beam waist, comparing these gives

$$\frac{\Gamma_{shear}}{\Gamma_{el}} = \frac{k_i P n L^2}{2cKw} \tag{9}$$

where $P \simeq Iw^2$ is the power and L the sample thickness. For typical experimental values[5] ($P \simeq 10^{-3}W, w \simeq 10^{-5}m, L \simeq 10^{-5}m, k_i \simeq 10m^{-1}, K \simeq 10^{-11}N$) this ratio is $<< 1$; hence this effect is negligible.

We conclude therefore, in agreement with Janossy[2], that direct optical effects are not sufficient to explain the observed transition; a different source of torque must exist. In the subsequent discussion, we ignore momentum transfer from the radiation field to the system.

RATCHETS

In thermal equilibrium, the Brownian motion of particles is symmetric even in an anisotropic medium, hence the net translational or rota-

tional displacement of particles averages to zero. To illustrate this point, Feynman[9] discussed the possibility of constructing a microscopic 'ratchet-and-pawl' motor. However, if the system is driven from equilibrium by external action, biased Brownian motion[10] and net transport[11] are possible[12]. Prost *et al.*[11] showed that in a periodically pumped two-level system, translational current of particles against a viscous restoring force can be maintained by transferring only energy and not momentum to the system. Translational ratchets, characterized by asymmetric and spatially periodic potentials which may be turned ON and OFF, give rise to net currents, and '*motion without a force*'[11].An optical thermal ratchet of this type has been realized recently[13]. Such transport is important in biological systems, and in a variety of technologies ranging from particle separation to signal processing and the production of microscopic machines[12]. We argue here that the orientational transport of dyes is *'rotation without a torque'*, and that the Janossy effect is one, perhaps the first, example of an orientational ratchet.

In his model of the absorption induced director reorientation, Janossy[3] proposed that the torque on the liquid crystal originates in the interaction between the liquid crystal and the photoexcited dye molecules. He argued that the strength of the mean field acting on the dye molecules is different in the ground and excited states, and proposed two coupled rate equations to describe the time evolution of the orientational distribution functions[3]. His equations correspond to the coupled Fokker-Planck equations of Prost[11] to describe the time evolution of the positional probability distributions of a particle in one of two states. Although Janossy allows only optical excitations to the excited state, whereas Prost considers more general transition rates, the two models are nearly equivalent. Prost's two state model, where the particle alternates between two states and experiences two different potentials, is similar to the one state ratchet model where a potential is turned ON and OFF[12]. Both in Janossy's and Prost's models, a net current is produced as a result of asymmetry.

We simplify further Janossy's model to gain insight into the essential

physics. The dye is regarded as an anisotropic molecule, with orientation specified by the unit vector $\hat{\mathbf{s}}$, which in the ground state does not interact with the nematic liquid crystal, but in the excited state interacts as another 'nematic' molecule. The probability of excitation is $\sim (\mathbf{E} \cdot \hat{\mathbf{s}})^2$, where $\mathbf{E}$ is the optical field and the lifetime of the excited state is τ. The interaction energy of the dye and the nematic with director $\hat{\mathbf{n}}$, is $U_1 = 0$ in the ground state, and is of the of the form $U_2 \sim (\hat{\mathbf{n}} \cdot \hat{\mathbf{s}})^2$ in the excited state. In the ground state, the dye molecule undergoes unbiased orientational diffusion. As a result of the irradiation, a dye molecule is excited into the excited state, where it undergoes directed orientational Brownian motion under the influence of the nematic potential, and eventually decays into the ground state. Since in this process the ground state dye molecules are not symmetrically distributed around the director, they can exert a net torque on the director.

Symmetry Considerations

We next examine the symmetry requirements for such a net torque to exist. For simplicity, we consider a $2-d$ situation, where the orientation of the director $\hat{\mathbf{n}}$ and the dye $\hat{\mathbf{s}}$ can be described by the angles θ_n and θ_d respectively. The Fokker-Planck equations for the evolution of the densities of the ground and excited state molecules become

$$\partial_t \rho_1 = D_1 \partial_{\theta_d} \{\partial_{\theta_d} \rho_1 + \rho_1 \partial_{\theta_d} U_1\} - \rho_1 f_1 + \rho_2 f_2 \tag{10}$$

and

$$\partial_t \rho_2 = D_2 \partial_{\theta_d} \{\partial_{\theta_d} \rho_2 + \rho_2 \partial_{\theta_d} U_2\} - \rho_2 f_2 + \rho_1 f_1 \tag{11}$$

where D and f are the diffusivity and transition rate for dye molecules; the subscripts 1 and 2 denote, respectively, the ground and excited states. In the simplified example above, $U_1 = 0$. The torque exerted by the nematic field on a dye molecule in the ground state is $-\partial_{\theta_d} U_1$ and similarly in the excited state. The average torque exerted by the dye on the nematic is therefore

$$\Gamma_1 + \Gamma_2 = < \rho_1 \partial_{\theta_d} U_1 >_{\theta_d} + < \rho_2 \partial_{\theta_d} U_2 >_{\theta_d} \tag{12}$$

where $<>_{\theta_d}$ denotes orientational averaging; contribution to the torque is thus proportional to the orientational particle current in each state. We now examine how the torque depends on the symmetry properties of the potentials U and inverse lifetimes f. We assume the orientational dependence is weak; that is, of order ε. We then write

$$U_1(\theta_d) = u_{01} + \varepsilon u_1(\theta_d) \tag{13}$$

and

$$f_1(\theta_d) = \overline{\nu}_1 + \varepsilon \nu_1(\theta_d) \tag{14}$$

where $< \nu_1(\theta_d) >_{\theta_d} = < u_1(\theta_d) >_{\theta_d} = 0$, and similarly for particles in state 2. Averaging over time and expanding in Fourier series gives

$$< \rho_1(\theta_d,t) >_t = \overline{\rho}_1 + \varepsilon \sum_q \tilde{\rho}_1(q) e^{iq\theta_d} \tag{15}$$

and we obtain, to $O(1)$,

$$0 = \overline{\rho}_1 \overline{\nu}_1 - \overline{\rho}_2 \overline{\nu}_2 \tag{16}$$

If the total average dye density is ρ_{TOT}, this gives

$$\overline{\rho}_1 = \frac{\rho_{TOT}}{1 + \overline{\nu}_1/\overline{\nu}_2} \tag{17}$$

and

$$\overline{\rho}_2 = \frac{\rho_{TOT}}{1 + \overline{\nu}_2/\overline{\nu}_1} \quad . \tag{18}$$

Denoting Fourier coefficients with $\tilde{}$, to $O(\varepsilon)$, we obtain

$$\begin{bmatrix} -(q^2 D_1 + \overline{\nu}_1) & \overline{\nu}_2 \\ \overline{\nu}_1 & -(q^2 D_2 + \overline{\nu}_2) \end{bmatrix} \begin{bmatrix} \tilde{\rho}_1(q) \\ \tilde{\rho}_2(q) \end{bmatrix} = \begin{bmatrix} q^2 \overline{\rho}_1 \tilde{u}_1(q) + \overline{\rho}_1 \tilde{\nu}_1(q) - \overline{\rho}_2 \tilde{\nu}_2(q) \\ q^2 \overline{\rho}_2 \tilde{u}_2(q) + \overline{\rho}_2 \tilde{\nu}_2(q) - \overline{\rho}_1 \tilde{\nu}_1(q) \end{bmatrix} \tag{19}$$

which can be solved to give

$$\tilde{\rho}_1(q) = \frac{-(q^2 D_2 + \overline{\nu}_2)\overline{\rho}_1 \tilde{u}_1(q) - \overline{\nu}_2 \overline{\rho}_2 \tilde{u}_2(q) - D_2(\overline{\rho}_1 \tilde{\nu}_1(q) - \overline{\rho}_1 \tilde{\nu}_2(q))}{q^2 D_1 D_2 + D_1 \overline{\nu}_2 + D_2 \overline{\nu}_1} \tag{20}$$

and similarly for $\tilde{\rho}_2(q)$. Now we look at the torques.

$$\Gamma_1 = < \rho_1 \nabla U_1 > = \varepsilon^2 \sum_q \tilde{\rho}_1(q) iq \tilde{u}_1(-q) \tag{21}$$

or

$$\Gamma_1 = \varepsilon^2 \sum_q \frac{-(q^2 D_1 + \overline{\nu}_1)\overline{\rho}_2 \tilde{u}_2(q) iq \tilde{u}_1(-q)}{q^2 D_1 D_2 + D_1 \overline{\nu}_2 + D_2 \overline{\nu}_1} -$$

$$-\varepsilon^2 \sum_q \frac{D_1(\overline{\rho}_2 \tilde{\nu}_2(q) - \overline{\rho}_1 \tilde{\nu}_1(q) iq \tilde{u}_1(-q))}{q^2 D_1 D_2 + D_1 \overline{\nu}_2 + D_2 \overline{\nu}_1} \tag{22}$$

where we have made use of the fact that $\sum_q \tilde{u}_1(q) iq \tilde{u}_1(-q) = 0$. A similar expression holds for Γ_2. Finally, we get for the total torque

$$\Gamma = \varepsilon^2 \sum_q \frac{(aq^2 + b)\tilde{u}_2(q) iq \tilde{u}_1(-q)}{q^2 D_1 D_2 + D_1 \overline{\nu}_2 + D_2 \overline{\nu}_1} +$$

$$+\varepsilon^2 \sum_q \frac{(\overline{\rho}_2 \tilde{\nu}_2(q) - \overline{\rho}_1 \tilde{\nu}_1(q)) iq (D_1 \tilde{u}_1(-q) - D_2 \tilde{u}_2(-q))}{q^2 D_1 D_2 + D_1 \overline{\nu}_2 + D_2 \overline{\nu}_1} \tag{23}$$

where $a = (D_1 \overline{\rho}_2 - D_2 \overline{\rho}_1)$, and $b = (\overline{\nu}_1 \overline{\rho}_2 - \overline{\nu}_2 \overline{\rho}_1)$. Eq. 23 gives the general condition for nonvanishing torque on the nematic. In our example, $U_2 \sim (\hat{\mathbf{n}} \cdot \hat{\mathbf{s}})^2 = \cos^2 \theta_d$ if we chose our coordinate so that $\theta_n = 0$, and $f_1 \sim (\mathbf{E} \cdot \hat{\mathbf{s}})^2$ [†]; $U_1 = 0$, and $f_2 = const.$ Although $U_2(\theta_d)$ is symmetric about 0, f_1 is not (unless $\mathbf{E}$ is parallel to $\hat{\mathbf{n}}$), hence the term $\sum_q \frac{\overline{\rho}_1 \tilde{\nu}_1(q) iq \tilde{u}_2(-q)}{q^2 D_1 + \overline{\nu}_1}$ is non-vanishing, and the dye exerts a net torque on the nematic director[‡].

ANGULAR MOMENTUM BALANCE

In our simple model, the initial orientation of the excited dye molecule is close to that of the exciting field $\mathbf{E}$, and as it exerts a torque on the nematic, it experiences an equal and opposite torque from the nematic

[†] f_1 is also a weak function of $(\hat{\mathbf{n}} \cdot \hat{\mathbf{s}})^2$ due to detailed balance.

[‡] Although if $\mathbf{E}$ is parallel to $\hat{\mathbf{n}}$ and there is no net torque, the system may not be stable. If $\nu_1(q)$ is large, a small director fluctuation will result in a rapidly increasing torque, and the system can undergo a configurational transition.

field which causes it to rotate towards the nematic director. It eventually decays back to the ground state, diffuses, and gets excited again when it approaches an orientation nearly parallel to the exciting field **E**. In the steady state, the director orientation is fixed, determined by the balance of elastic torques, due to anchoring at the cell walls, and torques coming from the exited dye molecules, whose orientation is not parallel to the director. The dye molecule therefore very much resembles the rotor of a d.c. electric motor, whose winding becomes excited when it reaches a certain orientation. The nematic field acting on the dye plays the role of the field of the permanent magnet of the d.c. motor. The key difference is that conventional electric motors rely on inertia to reorient the rotor, whereas the dye relies on Brownian motion. The dye and the nematic together thus constitute a dissipative molecular motor. The analogy between the molecular and macroscopic motor is useful in considering the angular momentum current flow in the system.

The excited dye molecules rotate, on the average, from the orientation of the exciting field towards the nematic director; while the net orientational current of the dye molecules in the ground state is, on the average, zero. The rotating dye molecules experience, in addition to the reactive torque from the nematic field, a viscous torque from the surrounding nematic fluid. This is implicit in the dissipative dynamics described by the Fokker-Planck Eq. 10 and 11. In addition to exerting a torque on the nematic, the dye molecules are a source of vorticity, and give rise to shear flow. A simple schematic is shown in Fig. 2.

On the average, the torque due to viscous shear exerted by each dye molecule on the nematic fluid is equal and opposite to the torque exerted by the dye molecule on the nematic due to the interaction U_2. The total torque exerted by the dye on the nematic is therefore zero, and it is surprising therefore that there can be an observable response. However, the torque due to the interaction U_2 tends to align the nematic uniformly, whereas the viscous torques give rise to spatially heterogeneous shear induced alignment, as can be seen from Fig. 2. The wavenumber of the

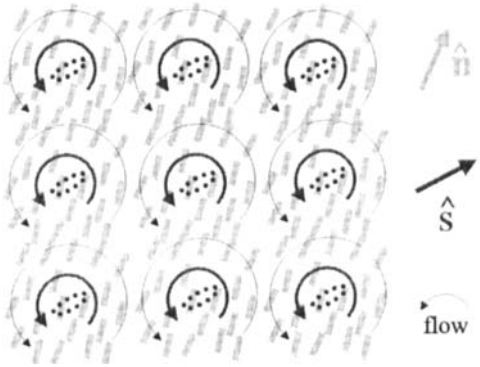

FIGURE 2 The rotating dye molecules, with average orientation shown by a dotted line, give rise to shear flow of the nematic. Shear alignment tends to orient the director so that it makes a fixed angle with the shear.

shear induced deformations is $q \simeq 2\pi/l_d$ where l_d is the mean distance between dye molecules, while the wavenumber of the deformation due to the potential U_2 is $q \simeq 2\pi/L$, where L is the sample thickness. Since the elastic restoring torque is $\Gamma_{el} \sim Kq^2$, the amplitude of director deformations due to shear relative to those caused by interactions via the potential U_2 is $\sim (l_d/L)^2 << 1$, hence shear induced deformations are negligible. Elastic torques essentially prevent the short wavelength distortions due to shear, but not the long wavelength distortions due to the interactions via the potential U_2. Due to the disparity of wavelengths, of the two effects do not cancel, and a net distortion of the nematic can occur.

The main role of shear flow then is to carry angular momentum back to the cell walls, where the torque due to shear flow on the walls balances the elastic torques from the nematic which act on the walls via the anchoring potential. Angular momentum current therefore circulates in the cell, carried from the cell walls by elastic interactions of the nematic, transferred to the dye through interaction via the potential U_2, and back to the walls through shear stress due to shear flow. The current is driven by the orientational ratchet mechanism using energy absorbed from the

radiation field which is in turn dissipated in shear. No external source of angular momentum is required. Fig. 3 shows a simple sketch of the system, useful for considering angular momentum flow, depicting the dye molecules as motors running in a viscous medium, interacting via elastic interactions and shear flow.

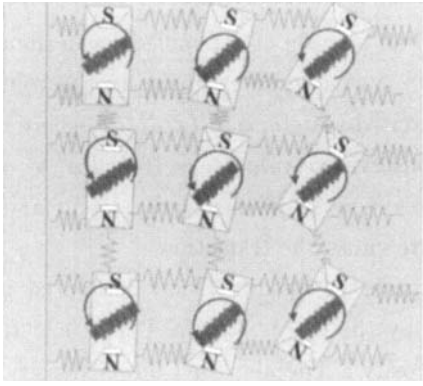

FIGURE 3 Schematic of motor analog where dye molecules are regarded as rotors in viscous medium, moving under optical excitation under the influence of the nematic field.

A torque is exerted on the cell wall on the left by the elastic interaction of nematic represented here by springs. An equal and opposite torque is exerted on the walls by viscous shear due to rotation of dye. The dye molecules, represented by rotors, rotate when in the excited state due to the nematic field. The rotors in turn exert a torque on the nematic director, represented by the stator, which is balanced by elastic interactions.

Energy Considerations

We now briefly consider energy requirements for this mechanism. First, we ask if the interaction energy between the dye and the liquid crystal is sufficient to generate the torque required for the distortion.

We assume the interaction energy of an excited dye molecule and the nematic to be on the order of kT_{NI}, where T_{NI} is the nematic-istropic

transition temperature, and the corresponding energy density is $\mathcal{E}_{dye} = \rho_2 U_2 \simeq \rho_2 kT$. The energy density associated with the elastic distortion is $\mathcal{E}_{el} = K/L^2 \simeq kT/(l_o L^2)$ where l_o is a molecular length. Comparing these, we get

$$\frac{\mathcal{E}_{dye}}{\mathcal{E}_{el}} = \rho_2 l_o L^2 \simeq \phi(\frac{L}{l_o})^2 \tag{24}$$

where ϕ is the volume fraction of dye molecules in the excited state. Typically $(L/l)^2 \sim 10^{10}$, and a crude estimate for the volume fraction of dye molecules in the excited state is $\phi \simeq I\tau\alpha l_o^3/h\nu$ where α is the absorption length. Taking conservative values $I = 10^6 W/m^2, \tau = 10^{-8} s$ $L = 10^{-4} m$ and $\alpha = 1/m$, gives $\phi = 10^{-10}$, hence $\mathcal{E}_{dye} \simeq \mathcal{E}_{el}$ and the interaction is sufficiently strong to cause the transition.

Next, we compare the absorbed and dissipated power. The power density dissipated by the dye is $\mathcal{P}_{diss} \simeq \Gamma^2/(\gamma\phi) = K^2/(\gamma\phi L^4)$ where γ is a rotational viscosity. The absorbed power density is $\mathcal{P}_{abs} \simeq I\alpha$, and comparing these, we get

$$\frac{\mathcal{P}_{diss}}{\mathcal{P}_{abs}} = \frac{K^2}{\gamma\phi L^4 I\alpha} \tag{25}$$

Taking the values $K = 10^{-11} N$, $\gamma = 10^{-2} Pas$, and ϕ, L and α as before, we get $\mathcal{P}_{diss} \simeq \mathcal{P}_{abs}$. Thus the energy absorbed from the radiation field is sufficient to drive the dissipative current

SUMMARY

In this paper we have considered the Janossy effect, where a small amount of dichroic dye dissolved in a nematic liquid crystal results in a reduction of the threshold intensity for the optical Freedericksz transition. We have confirmed that direct optical effects, including those due to absorption, do not provide sufficient torque to distort the nematic hence cannot account for the threshold reduction.

Our main contribution is to point out that the system exhibiting the Janossy effect is an optically pumped orientational ratchet, equivalent to

translational ratchet proposed by Prost[11]. In such a ratchet, the transfer of energy without the transfer of angular momentum from the light is sufficient to create orientational currents in the system. We have examined the symmetry requirements for a net torque on the director arising from orientational currents of the dye, and showed that although both the interaction potential between the nematic and the dye and the excitation rate are symmetric functions of their arguments about their minima, their relative positions can give rise to the asymmetry necessary for a nonzero torque to exist. We have provided a simple physical picture of the dye as a viscous molecular motor.

We have considered the details of torque balance. We pointed out the existence of shear flow in the system, and discussed its role in carrying angular momentum. We have explained how, although no net torque is exerted by the dye on the liquid crystal in the bulk, the interaction between the dye and the nematic results in a distortion of the director field. Finally, we showed that although the dye concentration is small, it can exert a sufficient torque on the director to bring about the transition, and that the power absorbed from the light field is comparable to the viscous dissipation in the system.

Our results are in agreement with the model of Janossy[3]; however, we have provided details here which were not apparent in Ref.[3].

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