Electromagnetic Torque and Force in Axially Symmetric Liquid Crystal Droplets

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Circularly polarized light exerts torque on birefringent objects. In the case of axially symmetric particles, however, the moment of radiation force balances the direct optical torque. This explains the observation that radial liquid crystal droplets, in contrast to planar droplets, do not spin in circularly polarized light. The conclusion is in agreement with considerations based on the angular momentum conservation of light (Marrucci et al. Phys. Rev. **96**, 163905, 2006).

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It is well known that light can carry angular momentum [1]. One part of the angular momentum is associated with the ellipticity of the polarization state of the light beam ("spin" angular momentum), while the second part is the orbital angular momentum. Circularly polarized waves carry a spin angular momentum of $\pm\hbar$ per photon. This part of the angular momentum was first demonstrated experimentally by Beth [2], who exposed a suspended birefringent object to polarized light and detected the torque on it. A more direct evidence of the transfer of angular momentum to matter is found in droplets of anisotropic materials, like liquid crystals, immersed in liquids. When such droplets are irradiated with circularly polarized light, they are set into

continuous rotation [3-5]. There are two ways to describe the latter phenomenon. The first method is to analyze the optical torque exerted by the electromagnetic field on the material; the second one is to compare the polarization state of the incoming light beam with that of the exiting beam.

In order to illustrate the two methods, we consider a plan-parallel slab of a liquid crystal with thickness d and area A, illuminated by a homogenous beam. For uniaxial media, such as nematic or smectic A phases the optical torque density, $\mathbf{\Gamma}$, can be written as [6]

$$\mathbf{\Gamma} = \langle \mathbf{P} \times \mathbf{E} \rangle = \varepsilon_0 (\varepsilon_{\parallel} - \varepsilon_{\perp}) \langle (\mathbf{n} \cdot \mathbf{E})(\mathbf{n} \times \mathbf{E}) \rangle \tag{1}$$

where $\mathbf{P}$ is the dielectric polarization, induced in the substance by the electric field, $\mathbf{E}$; $\varepsilon_{\perp} = n_o^2$, $\varepsilon_{\parallel} = n_e^2$ are the perpendicular and parallel components of the optical dielectric tensor; n_o and n_e are the ordinary and extraordinary refractive indices respectively, $\mathbf{n}$ is the liquid crystal director, the bracket < > denotes time averaging. In a planar droplet ($\mathbf{n}$ homogenous and perpendicular to the light propagation direction, z) the optical torque has only a z component, which is given as

$$\Gamma_z = \frac{1}{2} \varepsilon_0 (\varepsilon_{\parallel} - \varepsilon_{\perp}) E_{\parallel} E_{\perp} \cos \Delta\varphi . \tag{2}$$

Here $E_{\parallel}$ and $E_{\perp}$ are the amplitudes of the electric field components parallel and perpendicular to the director respectively; $\Delta\varphi$ is the phase difference between them. In the geometrical optics approximation [7] $\Delta\varphi(z) = k_0 (n_e - n_o) z + \Delta\varphi_0$, where k_0 is the wave vector in vacuum and $\Delta\varphi_0$ is the phase difference between the two polarization components at the entrance face of the slab ($z = 0$). The total optical torque, $\mathbf{M}$, acting on the slab has also only a z component:

$$M_z = A\int_0^d \Gamma_z dz = \frac{A}{2}\varepsilon_0(\varepsilon_\parallel - \varepsilon_\perp)E_\parallel E_\perp \int_0^d \cos(k_0(n_e - n_o)z + \Delta\varphi_0)dz$$
$$= A\varepsilon_0 \frac{n_e + n_o}{2k_0} E_\parallel E_\perp \left[\sin\Delta\varphi(d) - \sin\Delta\varphi_0\right]. \quad (3)$$

On the other hand, as shown in [7], in the geometrical optics limit inside a birefringent medium, the quantity $L_z(z) = \varepsilon_0 \frac{n_e + n_o}{2k_0} E_\parallel E_\perp \sin\Delta\varphi(z)$ can be regarded as the spin angular momentum density of the beam. Therefore the total torque can be written as $M_z = A[L_z(d) - L_z(0)]$, i.e. the torque is equal to the change of the angular momentum of the light beam across the sample.

In this letter we investigate the case of axially symmetric droplets. It was pointed out by Marrucci et al. [8] that a slab of nematic liquid crystal oriented radial around the symmetry axis of the slab, converts the spin angular momentum of light into orbital angular momentum. They demonstrated this by considering the example of a half-wave plate. When a (say) left circularly polarized beam passes through the slab, beside becoming right circularly polarized, it becomes also helical, i.e. the phase of the exiting beam changes by $2\pi m$ going around the optical axis. m is an integer; in the case considered $m = 2$ [8]. The change of the spin angular momentum ($m\hbar = 2\hbar$ per photon) is compensated by the change in orbital angular momentum, associated with the helicity of the beam ($-2\hbar$ per photon). The net change of the angular momentum of the light beam is therefore zero. From this fact it follows that the net electromagnetic torque, exerted by the optical field on the sample should vanish too. However, the direct optical torque, as determined above for planar cells, does not vanish in general for radial droplets. In fact, in the geometrical optics approximation, the torque is the same for the radial director configuration as for the uniform one.

In this letter we prove that the apparent contradiction between the two approaches of deriving the total torque can be resolved by taking into account the electromagnetic force acting on the droplet. As it will be shown, for axially symmetric droplets the z component of the moment of this force balances the direct optical torque. The total torque is zero hence axially symmetric droplets are not set into rotation by light beams.

There are different methods to calculate the radiation force. We first apply the method developed by Gordon [9] for "gaseous" medium, i.e. a medium, where the difference between the macroscopic and local field strengths can be neglected. In this limit, the force density, **f**, can be written as

$$\mathbf{f} = \mathbf{f}^{(E)} + \mathbf{f}^{(B)} \quad \text{with } \mathbf{f}^{(E)} = (\mathbf{P}\cdot\nabla)\mathbf{E} \quad \text{and} \quad \mathbf{f}^{(B)} = \frac{\dot{\mathbf{P}}}{c}\times\mathbf{B} \tag{4}$$

where **B** is the magnetic field strength. The first term on the right-hand-side of the equation represents the force acting on dipoles in an electric field gradient, while the second one gives the Lorentz force exerted by the magnetic field on oscillating dipoles. Introducing cylindrical coordinates ρ, ψ and z, the electric field strength can be written as $\mathbf{E} = E_\rho \mathbf{e}_\rho + E_\psi \mathbf{e}_\psi + E_z \mathbf{e}_z$, where $\mathbf{e}_\rho$, $\mathbf{e}_\psi$ and $\mathbf{e}_z$ are unit vectors in the radial, azimuthal and z directions, respectively. Similar expressions hold for **P**, **f** and ∇. The azimuthal component of the force is

$$f_\psi^{(E)} = \frac{1}{\rho} P_\psi \left(E_\rho + \frac{\partial E_\psi}{\partial \psi}\right) + P_\rho \frac{\partial E_\psi}{\partial \rho} + P_z \frac{\partial E_\psi}{\partial z},$$

$$f_\psi^{(B)} = \dot{P}_\rho \left(\frac{\hat{E}_\psi}{\rho} + \frac{\partial \hat{E}_\psi}{\partial \rho} - \frac{1}{\rho}\frac{\partial \hat{E}_\rho}{\partial \psi}\right) + \dot{P}_z \left(\frac{\partial \hat{E}_\psi}{\partial z} - \frac{1}{\rho}\frac{\partial \hat{E}_z}{\partial \psi}\right). \tag{5}$$

In deriving the first relation, the identity $\frac{\partial}{\partial \psi}\mathbf{e}_\rho = \mathbf{e}_\psi$ was used. For the second relation the Maxwell equation $\nabla \times \mathbf{E} = -\frac{1}{c}\dot{\mathbf{B}}$ was applied; $\widehat{\mathbf{E}} = \int_0^t \mathbf{E}(t')dt' + C$, where C is independent from time (although it depends on spatial coordinates). For a monochromatic wave $\left\langle \dot{P}_i \widehat{E}_j \right\rangle = -\left\langle P_i E_j \right\rangle$ and $\left\langle \dot{P}_i \frac{\partial \widehat{E}_j}{\partial x_k} \right\rangle = -\left\langle P_i \frac{\partial E_j}{\partial x_k} \right\rangle$, where the time averaging is over a number of full periods. The time average of the azimuthal force is therefore

$$\left\langle f_\psi \right\rangle = \frac{1}{\rho}\left\langle P_\psi E_\rho - P_\rho E_\psi \right\rangle + \frac{1}{\rho}\left\langle P_\rho \frac{\partial E_\rho}{\partial \psi} + P_\psi \frac{\partial E_\psi}{\partial \psi} + P_z \frac{\partial E_z}{\partial \psi} \right\rangle. \tag{6}$$

Let us consider a circularly polarized beam with an axially symmetric intensity distribution. Before penetrating into the droplet, the electric field components can be written as $E_x = A(\rho, z)\cos\omega t,\ E_y = A(\rho, z)\sin\omega t$. Performing a transformation from rectangular coordinates to cylindrical ones, we obtain $E_\rho = A(\rho, z)\cos(\omega t - \psi),\ E_\psi = A(\rho, z)\sin(\omega t - \psi)$. Within the droplet $E_i = A_i \cos(\omega t + \varphi_i - \psi)$, $i = \rho, \psi, z$, where the amplitude A_i and the phase φ_i depend on the details of light propagation. However, as a consequence of the axial symmetry of the droplet and the intensity distribution of the incoming beam, A_i and φ_i are only a function of ρ and z, but not of ψ. Taking into account this fact and using the linear relation between **P** and **E**, one obtains

$$\left\langle P_\rho \frac{\partial E_\rho}{\partial \psi} + P_\psi \frac{\partial E_\psi}{\partial \psi} + P_z \frac{\partial E_z}{\partial \psi} \right\rangle = 0. \tag{7}$$

Therefore the z component of the moment of electromagnetic force density, m_z, is

$$m_z = \rho\left\langle f_\psi \right\rangle = \left\langle P_\psi E_\rho - P_\rho E_\psi \right\rangle = -\Gamma_z \,. \tag{8}$$

As it can be seen, the moment of the force density compensates the direct optical torque, even locally.

In the general case the electromagnetic force is usually calculated from the Maxwell stress tensor and the Abraham force [9, 10]. In this approach the force density, $\mathbf{f}_{EM}$, is given as

$$\mathbf{f}_{EM} = \mathrm{Div}\,\mathbf{T} + \frac{1}{c}\frac{\partial}{\partial t}\mathbf{E}\times\mathbf{B} \tag{9}$$

where the Maxwell stress tensor is, for non-magnetic materials, $T_{ij} = (F_{em} + \rho_m \frac{\partial F_{em}}{\partial \rho_m})\delta_{ij} + E_i D_j + B_i B_j / \mu_0$, with $F_{em} = -\frac{1}{2}(\mathbf{E}\cdot\mathbf{D} + B^2/\mu_0)$ and ρ_m is the density of the medium. It is shown by Gordon for isotropic dielectrics [9] that $\mathbf{f}$ and $\mathbf{f}_{EM}$ differ by the gradient of a scalar function of the field strengths. To prove that the same holds for anisotropic media, we use the Maxwell equations (assuming no static charges or currents): $\mathrm{div}\,\mathbf{D} = \mathrm{div}\,\mathbf{B} = 0; \nabla\times\mathbf{E} = -\frac{1}{c}\dot{\mathbf{B}}; \nabla\times\mathbf{B} = \frac{1}{c}\dot{\mathbf{D}}$, with $\mathbf{D} = \varepsilon_0\mathbf{E} + \mathbf{P}$. Taking into account the mathematical identities $\mathrm{grad}(\mathbf{E}\cdot\mathbf{D}) = (\mathbf{E}\cdot\nabla)\mathbf{D} + (\mathbf{D}\cdot\nabla)\mathbf{E} + \mathbf{E}\times(\nabla\times\mathbf{D}) + \mathbf{D}\times(\nabla\times\mathbf{E})$ and $\frac{1}{2}\mathrm{grad}\,B^2 = (\mathbf{B}\cdot\nabla)\mathbf{B} + \mathbf{B}\times(\nabla\times\mathbf{B})$ one finds from Eqs (4) and (9):

$$\mathbf{f}_{EM} - \mathbf{f} = \mathrm{grad}(\rho_m \frac{\partial F_{em}}{\partial \rho_m} - \frac{1}{2}\mathbf{E}\cdot\mathbf{P}) \,. \tag{10}$$

As the difference between the forces can be expressed as a gradient of a scalar function, it follows that $\oint_S \langle f_{EM,\psi} - f_\psi \rangle ds = 0$. The integral is taken along a circle in the ρ, ψ plane with the center at the z axis. As a consequence of the axial symmetry, along this path $\langle f_{EM,\psi} - f_\psi \rangle$ must be independent of ψ. For this reason not only the integral, but its argument should vanish as well, i.e. $\langle f_{EM,\psi} \rangle = \langle f_\psi \rangle$. It is thus shown that Eq. (8) holds in the general case too.

From the above considerations it follows that in circularly polarized light the total torque acting on radial liquid crystal droplets vanishes. As a result, these droplets, in contrast to planar ones, do not spin in circularly polarized light. This conclusion is in agreement with experimental observations [11, 12].

The direct optical torque by itself causes director deformation, while the electromagnetic force induces flow. The question arises that how can they compensate each other? To find the answer we note that the initial director configuration is indeed distorted by the optical torque exerted by the light beam. This distortion was directly observed in laser tweezers experiments [11, 12]. Associated with the director deformation, a non-uniform stress field develops in the droplet [13]. In planar configuration, where the moment of radiation force is zero [5], the force arising from the director stress spins the droplet. In radial droplets the two forces are equal in magnitude, but opposite in direction, therefore the droplet is not rotated. This is true regardless form the light-induced deformation of the director, provided that the director distribution remains axially symmetric. It should be noted that this conclusion is not based on the geometrical optics approximation; it is valid for plane waves as well as for tightly focused beams. It also holds for any rheology between the liquid and liquid crystal. It is possible, nevertheless, that above a threshold intensity symmetry-breaking deformation occurs in the director configuration of a

radial droplet. Above the threshold the axial symmetry is lost therefore the moment of the radiation force does not compensate exactly the direct optical torque. In this case droplet rotation takes place. Such observations in radial droplets were reported by Murazawa et al. [11, 12].

Finally, we discuss two aspects of the above phenomenon, both connected with absorption. First, in the presence of absorption the dielectric polarization, **P**, is phase-shifted with respect to the electric filed, **E**; therefore Eq. (7) is not valid any longer. The exact compensation between the torques does not hold, so droplet rotation can occur. The second consequence of the presence of absorbing dyes is that the optical torque is greatly enhanced [14,15]. However, as shown in [5], in order to fulfill the law of conservation of angular momentum, the *total* torque acting on the droplet, associated with the enhancement, must be zero. Indeed, it was found that dye-doping has no influence on the spinning speed of planar droplets [5, 16]. In the case of radial droplets, dye-induced enhancement of the optical torque can not induce droplet rotation. On the other hand, it can lower the symmetry-breaking threshold, just like in the case of optical Freedericksz transition in dye-doped homeotropic nematic liquid crystal cells [14].

1. J.D. Jackson: *Classical Electrodynamics* , John Wiley & Sons, New York, NY (1999).
2. R.A. Beth, "Mechanical Detection and Measurement of the Angular Momentum of Light" Phys. Rev. **50,** 115 -125 (1936)**.**
3. S. Juodkazis, M. Shikata, T. Takahashi, S. Matsuo and H. Misawa, “Size Dependence of Rotation Frequency of Individual Laser Trapped Liquid Crystal Droplets” Jpn. J. Appl. Phys., Part 2, **38**, L518-L520 (1999).
4. S. Juodkazis, S. Matsuo, N. Murazawa, I. Hasegawa and H. Misawa, “High-efficiency optical transfer of torque to a nematic liquid crystal droplet” Appl. Phys. Lett. **82**, 4657-4659 (2003).

5. C. Manzo, D. Paparo, L. Marrucci and I. Jánossy, "Light-induced rotation of dye-doped liquid crystal droplets" Phys. Rev. E **73**, 05707 1-14 (2006).
6. E. Santamato, B. Danio, M. Romagnoli M. Settembre and Y.R. Shen, "Collective Rotation of Molecules Driven by the Angular Momentum of Light in a Nematic Film" Phys. Rev. Lett. **57**, 2423-2426 (1986).
7. E. Santamato and Y.R. Shen "Pseudo-Stokes parameter representation of light propagation in layered inhomogeneous uniaxial media in the geometric optics approximation" J. Opt. Soc. Am. A **4**, 356 - 359 (1987).
8. L. Marrucci, C. Manzo and D. Paparo, "Optical Spin-to-Orbital Angular Momentum Conversion in Inhomogeneous Anisotropic Media" Phys. Rev. Lett. **96**, 163905 1-4 (2006).
9. J.P. Gordon, "Radiation Forces and Moments in Dielectric Media" Phys. Rev. A **8**, 14-21 (1973).
10. L. Landau and E. Lifschitz, *Electrodynamics of Continuous Media*, Pergamon Press, Oxford (1984).
11. N. Murazawa, S. Juodkazis and H. Misawa, "Characterization of bipolar and radial nematic liquid crystal droplets using laser-tweezers" J. Phys. D: Appl. Phys. **38**, 2923-2927 (2005).
12. E. Brasselet, N. Murazawa, S. Juodkazis and H. Misawa, "Statics and dynamics of radial nematic liquid-crystal droplets manipulated by laser tweezers" Phys. Rev. E **77**, 041704 1-7 (2008).
13. S. Chandrasekhar, *Liquid Crystals*, 2nd ed. (Cambridge University Press, Cambridge, England, 1992), pp 88 – 98.
14. I. Jánossy, "Optical Reorientation In Dye-Doped Liquid Crystals" J. Nonlin.Opt. Phys. Mat. **8**, 361-377 (1999).

15. M. Kreuzer, L. Marrucci and D. Paparo, “Light-induced modification of kinetic molecular properties: enhancement of optical Kerr effect in absorbing liquids, photoinduced torque and molecular motors in dye-doped nematics” J. Nonlin.Opt. Phys. Mat. **9**, 157-182 (2000).
16. C. Manzo, D. Paparo, L. Marrucci and I. Jánossy, “Total optical torque and angular momentum conservation in dye-doped liquid crystal droplets spun by circularly polarized light” Mol. Cryst. Liq. Cryst. **454**, 101-110 (2006).