

# Effect of Extinction on Separation of Nanoparticle Enantiomers With Chiral Optical Forces

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**Abstract:** Separation of enantiomers of chiral inorganic nanoparticles can be performed using enantioselective optical forces that are strong enough to make the ordered drift of nanoparticles faster than their movement down a concentration gradient. Here, we solve the problem of nanoparticle diffusion in a bounded domain in the presence of an exponentially decaying driving force, which can represent a chiral force exerted on nanoparticle enantiomers by a circularly polarized light beam exhibiting either scattering, absorption, or both. We analyze the steady state spatial distributions of two basic purity measures of chiral mixtures, showing that extinction puts a fundamental limit on the degree of enantiopurification achievable with optical forces. Our solution can be used to model resolution of racemates of any kind of chiral inorganic nanoparticles that strongly interact with light.

**Index Terms:** Absorption, scattering, optomechanics, enantiomorphism.

## 1. Introduction

The chirality of the building blocks of life results in significantly different biological properties of opposite enantiomers of chiral drug molecules and in their profoundly different impacts on the human body [1]. Drug regulating authorities require separate testing of each enantiomer for its therapeutic efficacy and safety before it comes to the market. This demands versatile, cheap, and reliable methods of separation of molecular enantiomers, the absence of which is currently among the major factors which are slowing down the progress of pharmaceutical industry. The physical properties of molecular enantiomers are almost completely identical, and interactions with other chiral objects are needed to distinguish between them [2], [3].

Great prospects of the development of new chiral resolution methods are nowadays associated with chiral inorganic nanoparticles [4]–[7], such as semiconductor nanocrystals with surface [8]–[12]

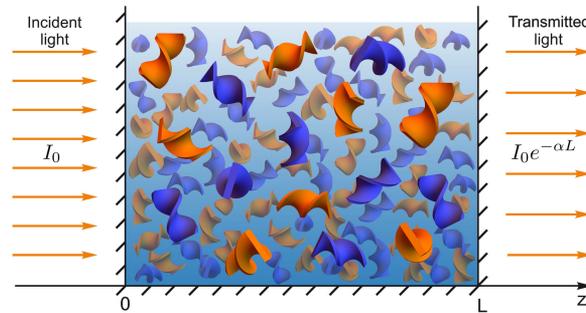


Fig. 1. Left (blue) and right (yellow) enantiomers of randomly oriented inorganic nanoparticles immersed in water and exposed to an exponentially decaying chiral optical force. The nanoparticles are contained inside a cuvette with impenetrable boundaries at  $z = 0$  and  $z = L$ .

or bulk defects [13], [14]. This is owing to the much stronger optical activity of chiral nanoparticles as compared to small chiral molecules and their selective interaction with biological tissues [15] and chiral biomolecules [16], [17]. These unique features have stimulated many theoretical studies aiming to suggest simple and efficient methods of separation of nanoparticle enantiomers using chiral light fields [18]–[24]. In this paper, we continue these studies by presenting an exact solution to the problem of diffusion in the presence of an exponentially decaying driving force, which may occur due to extinction of enantioselective circularly polarised light beam. The diffusion is assumed to take place in a bounded domain, modeling the geometry that is likely to be used for optomechanical resolution of nanoparticle racemates. We believe that the derived solution will eventually prove useful in the advancement of chiral analytical and separation techniques.

## 2. Formulation of the Problem

Consider an ensemble of weakly interacting nanoparticles in the form of a colloidal solution of concentration  $\rho_0$  and temperature  $T$  (see Fig. 1). Let at  $t = 0$  the nanoparticles be exposed to an optical driving force produced by an exponentially decaying light beam. If the beam is propagating in the positive direction of the  $z$  axis, and the decay of its intensity is characterized by a linear-loss (extinction) coefficient  $\alpha$ , then the force exerted on the nanoparticles can be written in the form  $\mathbf{F}(z) = \mathbf{e}_z F_0 e^{-\alpha z}$ , where  $\mathbf{e}_z$  is the unit vector and  $F_0$  scales in proportion to the beam intensity at  $z = 0$ . The spatiotemporal evolution of the nanoparticle concentration  $\rho(z, t)$  due to the force-induced diffusion of the nanoparticles is governed by the modified Fokker–Planck equation of the form [25]

$$\frac{\partial \rho}{\partial t} = D \frac{\partial^2 \rho}{\partial z^2} - v_0 \frac{\partial (e^{-\alpha z} \rho)}{\partial z} \quad (1)$$

where  $D$  is the diffusion coefficient,  $v_0 = D F_0 / (k_B T)$  is the nanoparticle drift velocity in the absence of diffusion and extinction, and  $k_B$  is the Boltzmann constant. This equation is seen to have two characteristic scales  $t_0 = D / v_0^2$  and  $z_0 = D / v_0$  and assume the following relation between the diffusion flux and the concentration of nanoparticles:

$$\xi(z, t) = -D \frac{\partial \rho}{\partial z} + v_0 e^{-\alpha z} \rho \quad (2)$$

which is the combination of Fick's first law and drift transport. In the forms written, (1) and (2) implicitly assume that the temperature is uniform throughout the solution, and that the diffusion coefficient and drift velocity are independent of  $z$ .

We shall assume that the motion of the nanoparticles is bounded to the domain between planes  $z = 0$  and  $z = L$ , which can represent the walls of a cuvette containing the solution. The density mass flux must vanish at both planes, due to the conservation of the nanoparticle number inside the cuvette

$$\xi(0, t) = \xi(L, t) = 0. \quad (3)$$

Together with the initial condition  $\rho(z, 0) = \rho_0$ , these boundary conditions determine the diffusion problem that is solved in this paper.

### 3. Steady State Solution

The steady state solution of the formulated diffusion problem corresponds to the zero nanoparticle flux  $\xi(z, t) = 0$  and is given by

$$\rho_s(z) = \rho_c \exp[L_{\text{eff}}(z)/z_0] \quad (4)$$

where  $L_{\text{eff}}(z) = (1 - e^{-\alpha z})/\alpha$  is the *effective propagation length* [26]–[28], which is reduced compared to the actual coordinate  $z$  the more the higher the extinction is, and the normalization constant is given by the expression

$$\rho_c = \rho_0 \frac{\alpha L e^{-\kappa}}{\text{Ei}(-\kappa) - \text{Ei}(-\kappa q)} \quad (5)$$

where  $\kappa = (\alpha z_0)^{-1}$ ,  $q = e^{-\alpha L}$ , and  $\text{Ei}(x)$  is the exponential integral.

The impact of extinction on the steady state distribution of nanoparticles can be estimated by comparing the present solution with the one corresponding to a constant optical force. Since  $L_{\text{eff}}(z) \rightarrow z$  and  $\rho_c \rightarrow \rho_0(L/z_0)/(e^{L/z_0} - 1)$  as  $\alpha$  approaches zero, (4) and (5) reduce to the exponential dependence of the nanoparticle concentration on the coordinate,  $\rho_s = \rho_c e^{z/z_0}$ , in full agreement with our previous work [18]. One can see that extinction converts this simple exponential solution to a saturable one, fundamentally limiting the degree of spatial separation of nanoparticle enantiomers that is achievable with optical forces.

The latter conclusion can be illustrated using two common measures of enantiopurity of chiral mixtures, i.e., enantiomeric excess (*ee*) and enantiomeric ratio (*er*), which characterize the excess of one enantiomer in a mixture with respect to the other [18]. Equations (4) and (5) are applicable to both enantiomeric forms of chiral nanoparticles. Suppose that initially there is a racemic mixture of two enantiomers, with equal concentrations of the nanoparticles of opposite handednesses, and that different enantiomers are subjected to oppositely directed optical forces, which have the same strength  $F_0 > 0$  at the domain boundaries. Then  $\rho_s(z)$  is the stationary concentration of the enantiomer that diffuses in the  $+z$  direction and  $\rho_s(L - z)$  is the concentration of the other enantiomer, which diffuses in the  $-z$  direction. The local (stationary) enantiomeric excess and enantiomeric ratio in this chiral mixture are given by

$$ee(z) = \frac{\rho_s(z) - \rho_s(L - z)}{\rho_s(z) + \rho_s(L - z)} = \tanh\left(\frac{L_{\text{eff}}(z) - L_{\text{eff}}(L - z)}{2z_0}\right) \quad (6)$$

$$er(z) = \frac{\rho_s(z)}{\rho_s(L - z)} = \exp\left(\frac{L_{\text{eff}}(z) - L_{\text{eff}}(L - z)}{z_0}\right) \quad (7)$$

and are seen to be the functions of parameters  $\alpha$  and  $z_0$ . The variations of the enantiopurity measures occur within ranges  $|ee(z)| \leq \tanh[L_{\text{eff}}(L)/(2z_0)]$  and  $|\ln er(z)| \leq L_{\text{eff}}(L)/z_0$ . As  $\alpha$  or  $z_0$  is increased, both ranges shrink due to the reduction of ratio of the effective domain length  $L_{\text{eff}}(L)$  to the characteristic diffusion scale  $z_0$ . For  $z_0 \gg L_{\text{eff}}(L)$  and  $\alpha \gg 1/L$ , the widths of these ranges become directly proportional to  $\kappa$ , reducing as  $|ee(z)| \lesssim \kappa/2$  and  $|\ln er(z)| \lesssim \kappa$ .

Fig. 2 shows the steady state concentration of nanoparticles and the two measures of enantiopurity defined in (6) and (7) for different values of  $\alpha L$  and  $L/z_0$ . The distribution of nanoparticles is seen to gradually change from a simple exponential for  $\alpha L = 0$  to an almost uniform one for  $\alpha L \gg 1$ . The comparison of the upper two panels shows that this change is more pronounced for larger ratios  $L/z_0$ . The spatial variations of enantiomeric excess and enantiomeric ratio, shown in panels (c) and (d), follow the same trend, being the most pronounced in the absence of extinction and gradually approaching their values in a racemic mixture when extinction becomes high and the driving force cease penetrating into the solution.

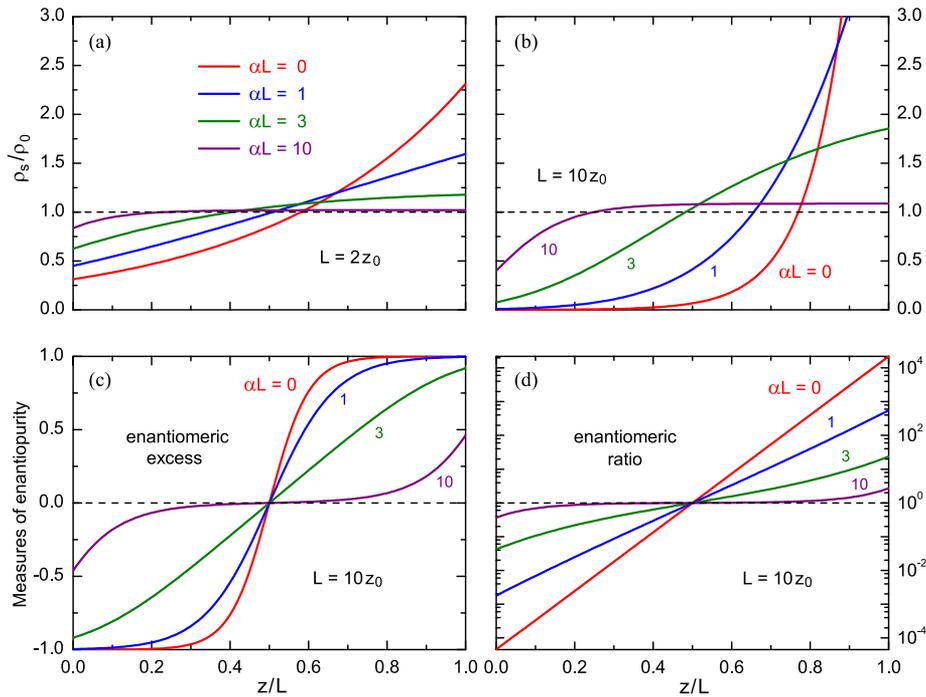


Fig. 2. (a) and (b) Steady state concentration of nanoparticles diffusing in the presence of exponentially decaying optical force and (c) and (d) steady state measures of enantiopurity in an initially racemic mixture of chiral nanoparticles, whose two enantiomeric forms are subjected to oppositely directed optical forces of the same strengths at  $z = 0$  and  $z = L$ .

#### 4. General Solution

In order to find the concentration of nanoparticles at an arbitrary time  $t$ , we use the ansatz  $x = e^{-\alpha z}$  to rewrite the diffusion equation and the boundary conditions in the form

$$\frac{\partial \rho}{\partial t} = D \alpha^2 x^2 \frac{\partial^2 \rho}{\partial x^2} + \alpha x (\alpha D + v_0 x) \frac{\partial \rho}{\partial x} + \alpha v_0 x \rho \quad (8)$$

$$\left. \frac{\partial \rho}{\partial x} \right|_{x=1} = -\kappa \rho(1, t), \quad \left. \frac{\partial \rho}{\partial x} \right|_{x=q} = -\kappa \rho(q, t). \quad (9)$$

We next separate the variables by representing the nanoparticle concentration in the form

$$\rho(x, t) = [1 + f(x)e^{-\mu^2 \alpha^2 D t}] \rho_s(x) \quad (10)$$

where  $\mu$  is a real parameter and  $\rho_s(x) = \rho_c e^{\alpha(1-x)}$  is the steady state solution given in (4). This form of general solution ensures that it approaches the steady state distribution of nanoparticles as time grows indefinitely large.

The unknown function  $f(x)$  obeys the ordinary differential equation

$$x^2 f'' + x(1 - \kappa x) f' + \mu^2 f = 0 \quad (11)$$

and the Neumann boundary conditions  $f'(1) = f'(q) = 0$ . The real solution to this equation can be written in the form

$$f(x) = x^{i\mu} [C_1 U(i\mu, 2i\mu + 1, \kappa x) + C_2 L_{-i\mu}^{2i\mu}(\kappa x)] + \text{c.c.} \quad (12)$$

where  $U(a, b, x)$  and  $L_n^k(x)$  are the confluent hypergeometric function of the second kind and the associated Laguerre polynomial [29],  $C_1$  and  $C_2$  are the real integration constants, and 'c.c.' stands for complex conjugate.

The boundary conditions lead to the dispersion equation

$$A(1)B(q) = A(q)B(1) \quad (13)$$

where

$$A(q) = i\mu q^{i\mu-1} U(i\mu, 2i\mu + 1, \kappa q) - \frac{i\mu}{\sqrt{\pi q}} e^{\kappa q/2} \kappa^{-i\mu+1/2} K_{i\mu+1/2} \left( \frac{\kappa q}{2} \right) + \text{c.c.} \quad (14)$$

$$B(q) = i\mu q^{i\mu-1} L_{-i\mu}^{2i\mu}(\kappa q) - \kappa q^{i\mu} L_{-1-i\mu}^{1+2i\mu}(\kappa q) + \text{c.c.} \quad (15)$$

and  $K_n(x)$  is the modified Bessel function of the second kind. This equation determines the spectrum of parameter  $\mu$ ,  $\mu_n = \{ \dots, \mu_{-2}, \mu_{-1}, \mu_0, \mu_1, \mu_2, \dots \}$ . The form of the dispersion equation suggests that  $\mu_{-n} = -\mu_n$  and  $\mu_0 = 0$ .

The time-dependent concentration of nanoparticles satisfying the boundary conditions can now be written in the form

$$\rho(x, t) = \left( 1 + 2 \sum_{n=0}^{\infty} a_n f_n(x) e^{-\mu_n^2 \alpha^2 D t} \right) \rho_s(x) \quad (16)$$

where

$$f_n(x) = x^{i\mu_n} [U(i\mu_n, 2i\mu_n + 1, \kappa x) + b_n L_{-i\mu_n}^{2i\mu_n}(\kappa x)] + \text{c.c.} \quad (17)$$

$$b_n = -\frac{A_n(1)}{B_n(1)} = -i\mu_n \frac{U(i\mu_n, 2i\mu_n + 1, \kappa) - \pi^{-1/2} e^{\kappa/2} \kappa^{-i\mu_n+1/2} K_{i\mu_n+1/2}(\kappa/2) + \text{c.c.}}{i\mu_n L_{-i\mu_n}^{2i\mu_n}(\kappa) - \kappa L_{-1-i\mu_n}^{1+2i\mu_n}(\kappa) + \text{c.c.}} \quad (18)$$

and we have taken into account that  $f_n(x) = f_{-n}(x)$ .

The initial condition  $\rho(x, 0) = \rho_0$  allows one to find the normalization constant  $a_n$ . In order to do this, we need to know the weight  $w(x)$ , with which the system of functions  $f_n(x)$  is orthogonal on the interval  $q \leq x \leq 1$ . It follows from the definition of  $f_n(x)$  and  $f_m(x)$  that they obey the equations

$$x^2 f_n'' + x(1 - \kappa x) f_n' + \mu_n^2 f_n = 0 \quad (19)$$

$$x^2 f_m'' + x(1 - \kappa x) f_m' + \mu_m^2 f_m = 0. \quad (20)$$

The subtraction of the first equation multiplied by  $f_m$  from the second equation multiplied by  $f_n$ , followed by the integration of both sides of the resulting equation with weight  $w(x)$  from  $q$  to 1, yields

$$\begin{aligned} (\mu_n^2 - \mu_m^2) \int_q^1 w f_n f_m dx &= \int_q^1 w x^2 (f_m' f_n - f_n' f_m)' dx + \int_q^1 w x(1 - \kappa x) (f_m' f_n - f_n' f_m) dx \\ &= w x^2 (f_m' f_n - f_n' f_m) \Big|_q^1 + \int_q^1 [w x(1 - \kappa x) - (w x^2)'] (f_m' f_n - f_n' f_m) dx. \end{aligned} \quad (21)$$

The first term on the right-hand side of this equation is zero due to the boundary conditions whereas the second term vanishes for  $w x(1 - \kappa x) = (w x^2)'$ . The solution to this differential equation (up to a constant factor) is given by  $w(x) = e^{-\kappa x}/x$ . We thus come to the orthogonality relation

$$\int_q^1 w f_n f_m dx = \sigma_n \delta_{nm} \quad (22)$$

where  $\delta_{nm}$  is the Kronecker delta and  $\sigma_n$  is implicitly defined. With this relation, the initial condition gives

$$a_n = \frac{1}{2\sigma_n} \int_q^1 w(x) f_n(x) \left( \frac{\rho_0}{\rho_c} e^{\alpha(x-1)} - 1 \right) dx. \quad (23)$$

Note that  $f_0 = 2$  and  $a_0 = 0$ , so that the summation in (16) can start from  $n = 1$ .

Equations (4), (5), (13)–(15), (16)–(18), (22), and (23) constitute the main results of this paper and are applicable regardless of the nature of the driving force. It is easy to show that in the absence of optical losses the obtained analytical solution is reduced to the well-known result from [18]. This can be inferred by noticing that  $x \rightarrow 1$ ,  $-\alpha \partial/\partial x \rightarrow \partial/\partial z$ , and  $\alpha^2 \partial^2/\partial x^2 \rightarrow \partial^2/\partial z^2$  for  $\alpha \rightarrow 0$  and, hence, (8) in the limit of small  $\alpha$  turns into the diffusion equation (16) presented in [18].

## 5. Conclusion

We have solved a 1-D diffusion problem for nanoparticles which experience a drift motion due to the presence of an exponentially decaying driving force. The motion is assumed to occur between two impenetrable planes, which can represent the walls of a cuvette containing the nanoparticles. It was shown that extinction converts an exponential steady state distribution of nanoparticles to a saturable one, and limits the degree of spatial separation of nanoparticle enantiomers achievable with optical forces. The derived solution can be used for modeling diffusion upon optomechanical resolution of nanoparticle racemates.

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