Enantioselective manipulation of chiral nanoparticles using optical tweezers

R. Ali, F. A. Pinheiro, F. S. S. Rosa, and P. A. Maia Neto
Instituto de Física, Universidade Federal do Rio de Janeiro,
Caixa Postal 68528, Rio de Janeiro, RJ, 21941-972, Brasil

R. S. Dutra
LISComp-IFRJ, Instituto Federal de Educação, Ciência e Tecnologia,
Rua Sebastião de Lacerda, Paracambi, RJ, 26600-000, Brasil

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We put forward a twofold enantioselective method for chiral nanoparticles using optical tweezers. First, we demonstrate that the optical trapping force in a typical, realistic optical tweezing setup with circularly-polarized trapping beams is sensitive to the chirality of core-shell nanoparticles, allowing for efficient enantioselection. Second, we propose another enantioselective method based on the rotation of core-shell chiral nanoparticles’ equilibrium position under the effect of a transverse Stokes drag force. In this case, the chirality of the particle shell gives rise to an additional twist, thus leading to a strong enhancement of the optical torque driving the rotation. Both chiral resolution methods are shown to be robust against the variation of the size of the system and material parameters, suggesting it could be applied to a wide range of experimental situations, particularly those of biological interest for which optical tweezing is widely used. Our results provide alternative enantioselective mechanisms and pave the way for all-optical manipulation and enantiopure synthesis of chiral nanoparticles. In addition, they can also be applied in the characterization of chirality for testing existing methods of enantioselection.

The concept of chirality, introduced by Lord Kelvin to designate any geometrical object or ensemble of points that lack mirror symmetry, is ubiquitous in Nature. The DNA double-helix structure and the related long-standing enigma of the origin of homochirality of life are just some examples of this fundamental concept in science [1]. In addition to its scientific importance, the concept of chirality is of utmost technological relevance. Indeed, molecular chirality plays a pivotal role in the pharmaceutical and food-processing industries. For instance, enantiopure drugs are known to be more effective and to produce less side effects than their racemic counterparts [2]. Molecular chirality is also essential in several nanotechnological applications, including molecular electronics, DNA nanotechnology, and chiral fullerenes and nanotubes [3].

In the vast majority of these applications, separation of enantiomers is crucial so that developing efficient strategies of chiral resolution is a very active research field. Existing chemical methods of enantioselectivity, such as chiral chromatography, usually produce unwanted side products and have low efficiency [4]. To circumvent these limitations, optical enantioselective mechanisms have emerged as promising strategies to achieve contactless chiral resolution. Indeed, fostered by progresses in the field of plasmonics and metamaterials, many theoretical optical chiral resolution methods have been proposed [5–20], and some of them have been experimentally realized [21–23]. However, due to the weak magnitude of optical forces, optical enantioselective methods are typically limited to particles larger than proteins and molecules of biological and pharmaceutical interest.

Plasmonic tweezers and metasurfaces can increase the magnitude of optical forces and for this reason they were applied to achieve enantioselection of nanometer-sized particles [27, 28]. Optical forces on chiral objects can also lead to interesting effects such as pulling forces [29, 30] and left-handed torques [29]. The closely related topics of optical enantiomeric recognition [31], characterization of chirality of molecules [32] and disordered systems [33] have also been recently investigated.

Here we put forward alternative chiral resolution methods using optical tweezers (OT) [34] with a circularly-polarized trapping beam. Our method allows for enantioselection of sub-100 nm particles and their all-optical manipulation, as well as for the quantitative characterization of chirality. It applies to realistic systems of biological interest, namely chiral molecules in solution. As common in applications of optical tweezers in molecular biology [35], we propose to trap silica nanospheres which are used as handles. In our proposal, outlined in Fig. 1(a), a circularly-polarized Gaussian laser beam propagating along the z-direction reaches an oil immersion high-NA objective that focuses the beam into a diffraction-limited spot in the sample beyond the coverslip. The sample contains an aqueous solution of chiral molecules that will eventually attach to silica nanospheres building a layer of chiral material that we model as an effective chiral shell. We show that optical trapping can only occur for chiral shells with the handedness selected by the choice of the trapping beam circular polarization, thus allowing for enantioselectivity.

We also calculate the optical force when the nanospheres are displaced off-axis by the Stokes drag...
Circularly Displacement the OAM associated to the circular polarization of the force $F$ component $F_z$ plane is rotated by an angle $\alpha$. The resulting Stokes drag force produces the trapped nanosphere along the same direction with a constant speed, producing a Stokes drag force $F_{\text{st}}$ enantioselectivity method based on the optical torque. The latter accounts for the OAM transferred to the nanosphere. The equilibrium position is rotated around the $z$-axis by the angle $\alpha$.

We drive the sample along the $x$-direction with a constant speed, producing a Stokes drag force $F_s$ that would displace the trapped nanosphere along the same direction. However, the resulting equilibrium position on the $xy$ plane is rotated by an angle $\alpha$ around the laser beam axis $z$. The optical force is written as a multipole series in terms of the coefficients $C_{\ell m}^{(a)}(\rho, z)$. These are functions of the sphere position (with respect to the paraxial focus) in cylindrical coordinates $(\rho, z)$, with $\ell, m$ representing the multipole order. Explicit expressions for the cylindrical components of $Q$ are given in the supplementary material [49]. We take $\sigma = \pm 1$ for...
left-handed/right-handed circular polarizations. The integration variable θ represents the angle between each incident propagation direction in the glass medium (index \( n_g \)) and the z-axis. The integration limit is defined by the objective numerical aperture (NA): \( \theta_0 = \arcsin(NA/n_g) \). The parameter γ represents the ratio of the objective focal length to the Gaussian beam waist at the objective entrance port (recalling that the focused beam is not described by a paraxial Gaussian model). \( T(\theta) \) is the Fresnel transmission amplitude for refraction at the interface between the glass coverslip and the aqueous solution. Such refraction also introduces the spherical aberration phase correction \( \Phi_\omega(\theta) \). Explicit expressions are given in [19]. \( d_{m,w}^\omega(\theta) \) are the Wigner rotation matrix elements evaluated at the angle \( \theta_w = \arcsin(n_g \sin \theta/n_w) \) in the aqueous solution and \( J_m \) are the cylindrical Bessel functions of integer order \( m \) [3]. Finally, \( k = 2\pi n_g/\lambda_0 \) and \( k_w = n_w k/n_g \) are the wavenumbers in the glass and aqueous solution, respectively, where \( \lambda_0 \) is the vacuum wavelength.

For all numerical examples discussed below, we take the typical values \([22]\) \( \lambda_0 = 1064\, \text{nm}, \ n_w = 1.332, \ n_g = 1.51, \ \gamma = 1.226 \) and \( \text{NA} = 1.3 \). We consider a silica core (index 1.45) of radius \( a = 500\, \text{nm} \) covered with a layer of a chiral material with \( \varepsilon = 2.89 \) and several realistic values for the chirality parameter \( \kappa \).

We begin by analyzing the variation of the axial optical force efficiency \( Q_z \) with the sphere position \( z/r \) (in units of the outer radius \( r \)) along the beam symmetry axis, in order to show that trapping strongly depends on the sign of \( \kappa \). This is demonstrated in Fig. 2 for the case of right-handed circular polarization \( \sigma = -1 \). For a coating of thickness \( t = 175\, \text{nm} \) (main plot), negative (backward) forces are achieved for positive moderate, realistic values of \( \kappa \). This provides a stable trapping near the focus (\( z = 0 \)), while nanospheres coated with the opposite handedness are pushed away from the focal region by radiation pressure. A more complete picture is provided by the inset of Fig. 2, which presents a density plot showing the negative values of \( Q_z \) as a function of \( \kappa \) and the shell thickness \( t \). Enantioselectivity is possible in the range \( 0.1\, \mu\text{m} < t < 0.2\, \mu\text{m} \), whereas spheres coated with thin layers (\( t < 0.1\, \mu\text{m} \)) are trapped regardless of the sign of \( \kappa \), recovering the results for achiral homogeneous nanospheres [37] [38] as \( t \to 0 \).

We have verified that the results for the axial force efficiency \( Q_z \) shown in Fig. 2 are invariant if we switch from right-handed to left-handed circular polarization and also change the sign of the chirality parameter \( \kappa \). Thus, one can select the handedness of the coating material of the spheres which are optically trapped by simply choosing the appropriate polarization of the incident laser beam.

Another enantioselectivity mechanism is obtained by driving the sample laterally so as to displace the trapped particle away from the beam axis, as discussed in connection with Fig. 1b). Due to the transfer of OAM, the equilibrium position is rotated by an angle \( \alpha \) which depends strongly on both \( \kappa \) and \( t \). In Fig. 3 we plot \( \alpha \) as a function of the shell thickness \( t \), with the silica core radius fixed at \( a = 500\, \text{nm} \) for three different values of \( \kappa \). In the case of very thin shells, \( t \lesssim 50\, \text{nm} \), the curves corresponding to different values of \( \kappa \) approach each other and recover the result for a homogeneous achiral sphere as \( t \to 0 \), as expected. For homogenous achiral spheres, the rotation angle \( \alpha \) is usually opposite to the polariza-

![FIG. 2: Dimensionless axial force efficiency \( Q_z \) as a function of position (in units of the sphere outer radius \( r \)) along the laser axis. We take a left-handed circularly polarized laser beam and consider a silica core of radius \( a = 500\, \text{nm} \) coated with a shell of thickness \( t = 175\, \text{nm} \) and chirality parameter \( \kappa = -0.05 \) (dotted line), \( \kappa = 0 \) (dashed line), \( \kappa = +0.05 \) (dotted line) and \( \kappa = 0.12 \) (solid line). In the first two cases, the core-shell particle is expelled from the focal region by radiation pressure, whereas in the last two ones the particle is trapped near the paraxial focus (\( z = 0 \)). Inset: density plot showing the range of \( \kappa \) and \( t \) leading to negative axial force efficiencies \( (Q_z < 0) \) required for trapping, at the fixed nanosphere position \( z/r = 1 \).

![FIG. 3: Rotation angle \( \alpha \) of the particle equilibrium position as a function of the shell thickness \( t \), for chirality parameters \( \kappa = 0.12 \) (solid line), \( \kappa = 0 \) (dashed line) and \( \kappa = -0.1 \) (dotted line). For the first two cases, the curves end at their respective thresholds of no trapping, since radiation pressure dominates for larger values of \( t \). The silica core radius is fixed at \( a = 500\, \text{nm} \).]
tion handedness of the trapping beam [30]. Hence we find a positive $\alpha$ in the limit $t \rightarrow 0$ when taking right-handed circular polarization (negative spin angular momentum) as in the example of Fig. 3. In other words, the optical torque is opposite to the spin angular momentum of the trapping beam (so-called negative optical torque [51–56]) in the case of very thin shells.

Fig. 3 shows that the optical torque becomes positive (and hence $\alpha$ goes negative for right-handed circular polarization) as the shell thickness increases. For $50 \text{ nm} \leq t \leq 100 \text{ nm}$, the optical torque allows for a mechanical separation of particles with different handednesses, since the curves for the different values of $\kappa$ shown in Fig. 3 are sufficiently apart in this range. Since the condition for trapping is not enough to ensure enantioselectivity in this intermediate thickness range (see Fig. 2), the method based on optical rotation is particularly useful in this case.

The solid line, corresponding to $\kappa = 0.12$, displays a pronounced dip at the thickness $t = 167.5 \text{ nm}$, corresponding to a rotation angle $\alpha \approx 19^\circ$ and hence larger by a factor $\sim 5$ than in the case of achiral particles of the same size, for which the rotation was already experimentally demonstrated [28]. Such an impressive increase in the rotation angle makes trapped spheres with chiral coatings particularly suitable for experimental demonstrations of optical torque in optical tweezers. Taken together, the results of Fig. 3 suggest that the transfer of OAM in optical tweezers might provide an efficient method for characterizing the thickness and, more importantly, the chirality of the coating.

We pursue the proposal of chirality characterization in Fig. 3, where we plot the rotation angle $\alpha$ as a function of the chirality parameter $\kappa$ for a coating of thickness $t = 167.5 \text{ nm}$. When employing a right-handed circularly polarized trapping beam (solid line), $\alpha$ displays a strong, approximately linear dependence in the range $0 \leq \kappa \leq 0.1$, allowing for a good resolution in the determination of $\kappa$ in this important range. Given a typical precision $\delta \alpha \sim 0.2^\circ$ in the determination of the rotation angle [30], we estimate a chirality resolution $\delta \kappa \sim 10^{-3}$ from Fig. 4. As discussed in connection with Fig. 2, it is also possible to trap nanospheres coated with the opposite handedness by switching to left-handed circular polarization. In this case, the sign of the rotation angle is reversed, and the resulting variation (dotted line) allows for the characterization of negative values of $\kappa$.

In conclusion, we show that optical tweezing of chiral core-shell nanospheres is enantioselective, since the sign of the chiral parameter allowing for trapping can be selected by choosing the appropriate polarization (left or right circular polarization) of the laser beam, as long as the chiral shell thickness exceeds $\sim 100 \text{ nm}$. Nanospheres with the opposite handedness are simply pushed away from the focal region by radiation pressure. In addition, we put forward another chiral resolution method, where enantiorecognition is achieved by analyzing the rotation of the trapped particle equilibrium position when it is displaced off-axis by driving the sample laterally. By employing this additional method, enantioselection is also achieved for thinner coatings, in the range $50 \text{ nm} \leq t \leq 100 \text{ nm}$. We generally find large rotation angles which should be easily measurable. The angle is strongly dependent on the chirality parameter, thus providing a good resolution for its characterization in the range $0 \leq \kappa \leq 0.1$. Altogether our results demonstrate a twofold alternative enantiorecognition mechanism, based on either optical trapping and optical rotation, which paves the way for all-optical manipulation and enantiopure synthesis of chiral nanoparticles.

Acknowledgments

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Supplemental Materials: Enantioselective manipulation of chiral nanoparticles using optical tweezers

This supplement contains an expanded theoretical description of optical tweezers of particles with a chiral shell.

We consider a circularly-polarized Gaussian beam at the objective entrance port, with $\sigma = \pm 1$ denoting left-handed/right-handed polarization. The dimensionless optical force efficiency (see main text for definition in terms of the optical force) is written as

$$Q(\rho, \phi, z) = Q_s(\rho, \phi, z) + Q_\ell(\rho, \phi, z).$$

(4)

The extinction term $Q_s$ accounts for the rate of momentum removal from the incident beam, while the scattering contribution $Q_\ell$ corresponds to the negative of the rate of momentum carried away by the scattered field. The explicit expressions for their cylindrical components are given below as sums over multipoles of the form

$$\sum_{\ell m} (...) \equiv \sum_{\ell=1}^{\infty} \sum_{m=-\ell}^{\ell} (...)$$

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- Scattering axial component

$$Q_{sz}(\rho, \phi, z) = \frac{8\gamma^2}{AN} \sum_{\ell m} \frac{\sqrt{\ell(\ell+2)(\ell+m+1)(\ell-m+1)}}{\ell+1} \left[ (A_{\ell}A_{\ell+1}^* + B_{\ell}B_{\ell+1}^*)G_{\ell,m}^{\sigma}G_{\ell+1,m}^{\sigma*} \right]$$

$$- \frac{8\gamma^2}{AN} \sigma \sum_{\ell m} \frac{(2\ell+1)}{\ell(\ell+1)} mA_{\ell}B_{\ell}^* |G_{\ell,m}^{\sigma}|^2$$

- Scattering radial component

$$Q_{sr}(\rho, \phi, z) = \frac{4\gamma^2}{AN} \sum_{\ell m} \frac{\sqrt{\ell(\ell+2)(\ell+m+1)(\ell+m+2)}}{\ell+1} \text{Im}\left\{ (A_{\ell}A_{\ell+1}^* + B_{\ell}B_{\ell+1}^*) \right\}$$

$$\left[ G_{\ell,m}^{\sigma}G_{\ell+1,m+1}^{\sigma} + G_{\ell,-m}^{\sigma}G_{\ell+1,-(m+1)}^{\sigma} \right]$$

$$- \frac{8\gamma^2}{AN} \sigma \sum_{\ell m} \frac{(2\ell+1)}{\ell(\ell+1)} \sqrt{(\ell-m)(\ell+m+1)} \left[ \text{Re}(A_{\ell}B_{\ell}) \text{Im}(G_{\ell,m}^{\sigma}G_{\ell+1,m}^{\sigma*}) \right]$$

- Scattering azimuthal component

$$Q_{s\phi}(\rho, \phi, z) = -\frac{4\gamma^2}{AN} \sum_{\ell m} \frac{\sqrt{\ell(\ell+2)(\ell+m+1)(\ell+m+2)}}{\ell+1} \text{Re}\left\{ (A_{\ell}A_{\ell+1}^* + B_{\ell}B_{\ell+1}^*) \right\}$$

$$\left[ G_{\ell,m}^{\sigma}G_{\ell+1,m+1}^{\sigma} - G_{\ell,-m}^{\sigma}G_{\ell+1,-(m+1)}^{\sigma} \right]$$

$$+ \frac{8\gamma^2}{AN} \sigma \sum_{\ell m} \frac{(2\ell+1)}{\ell(\ell+1)} \sqrt{(\ell-m)(\ell+m+1)} \times \text{Re}(A_{\ell}B_{\ell}) \text{Re}(G_{\ell,m}^{\sigma}G_{\ell+1,m}^{\sigma*})$$

- Extinction axial component

$$Q_{ez}(\rho, \phi, z) = \frac{4\gamma^2}{AN} \sum_{\ell m} (2\ell+1)G_{\ell,m}^{\sigma} \left[ (A_{\ell} + B_{\ell})G_{\ell,m}^{\sigma*} \right],$$

(5)

- Extinction radial component

$$Q_{er}(\rho, \phi, z) = \frac{2\gamma^2}{AN} \text{Im} \sum_{\ell m} (2\ell+1)G_{\ell,m}^{\sigma} \left[ (A_{\ell} + B_{\ell}) \left( G_{\ell,m+1}^{\sigma*} - G_{\ell,m-1}^{\sigma*} \right) \right]$$
• Extinction azimuthal component

\[ Q_{\epsilon \phi}(\rho, \phi, z) = -\frac{2\gamma^2}{AN} \sum_{\ell m} (2\ell + 1) G^{(\sigma)}_{\ell, m} \left[ (A_{\ell} + B_{\ell}) \left( G_{\ell, m-1}^{(\sigma)} + G_{\ell, m+1}^{(\sigma)} \right)^* \right] \]

In addition to the multipole coefficients \( G^{(\sigma)}_{\ell, m}(\rho, \phi, z) \) defined in Eq. (3) of the main letter, we also define

\[ G^{(\sigma)\pm}_{\ell, m}(\rho, \phi, z) = \int_0^{\theta_0} d\theta \sin \theta \cos \theta_w \sqrt{\cos \theta} T(\theta) e^{-\gamma^2 \sin^2 \theta \rho^2_{m,\sigma}(\theta_w)} J_{m-\sigma} (k\rho \sin \theta) e^{i[\Phi_w(\theta) + k_w \cos \theta_w z]}, \]

\[ G^{(\sigma)\pm}_{\ell, m}(\rho, \phi, z) = \int_0^{\theta_0} d\theta \sin \theta \cos \theta_w \sqrt{\cos \theta} T(\theta) e^{-\gamma^2 \sin^2 \theta \rho^2_{m,\sigma}(\theta_w)} (\Phi_w(\theta) + k_w \cos \theta_w z], \]

with \( \theta_w = \sin^{-1}(\sin \theta/N) \) and \( N = n_w/n_g \). The phase \( \Phi_w \) accounts for the spherical aberration arising from the refractive index mismatch at the glass-water interface:

\[ \Phi_w(\theta) = k (-L/N \cos \theta + NL \cos \theta_w), \]

where \( L \) represents the distance between the paraxial focal plane and the glass slide. We also take \( T(\theta) \) as the Fresnel transmission amplitude (neglecting polarization dependence since \( N \approx 1 \)).

The factor

\[ A = 16\gamma^2 \int_0^{\theta_0} ds \exp(-2\gamma^2 s^2) \frac{\sqrt{(1-s^2)(N^2-s^2)}}{(\sqrt{1-s^2} + \sqrt{N^2-s^2})^2} \]

is the fraction of the beam power transmitted into the sample chamber, with \( \theta_0 = NA/N_g \).

The optical force components also depend on the effective external Mie coefficients \( A_{\ell} \) and \( B_{\ell} \) which we derive as follows:

\[ A_{\ell} = a_{\ell} + i\sigma d_{\ell} \]
\[ B_{\ell} = b_{\ell} - i\sigma c_{\ell} \]

(6)

\( A_{\ell} \) and \( B_{\ell} \) are convenient for a number of applications involving circularly-polarized fields scattered by chiral media. The coefficients \( a_{\ell}, b_{\ell}, c_{\ell} \) and \( d_{\ell} \) are the effective Mie coefficients for a sphere made of chiral material [1].

For a core-shell nanosphere, their explicit expressions [2] are given in terms of the size parameters \( a = k_w a \) and \( v = k_w r \) corresponding to the core and outer radii \( a \) and \( r \). The refractive indexes of the chiral shell (with respect to the host) are \( N_{L/R} = (\sqrt{\epsilon} \pm \kappa)/n_w \), where \( \kappa \) is the chirality parameter (see main text). We also need the relative refractive index of the core \( N_i \) with respect to the host medium of index \( n_w \). Finally, \( N_{HI} = (N_L + N_R)/2 \) is the average relative index of the chiral shell.

\[ a_{\ell} = -\Delta_{\ell}^{-1}(A_{\ell} W_{L\ell} + A_{\ell} W_{R\ell}) \]
\[ b_{\ell} = -\Delta_{\ell}^{-1}(B_{\ell} W_{L\ell} + B_{\ell} W_{R\ell}) \]
\[ c_{\ell} = i\Delta_{\ell}^{-1}(A_{\ell} V_{L\ell} + A_{\ell} V_{R\ell}) \]
\[ d_{\ell} = i\Delta_{\ell}^{-1}(B_{\ell} V_{L\ell} - B_{\ell} V_{R\ell}) \]

with

\[ \Delta_{\ell} = W_L V_{R\ell} + B_{\ell} W_{R\ell} \]
\[ A_{R\ell} = X_{R\ell}(-\eta_{\ell}^{(1)}(v) - N_{HI} U_{R\ell}(+) \eta_{\ell}(v)) \]
\[ A_{L\ell} = X_{L\ell}(+\eta_{\ell}^{(1)}(v) - N_{HI} U_{L\ell}(+) \eta_{\ell}(v)) \]
\[ B_{L\ell} = X_{L\ell}(-\eta_{\ell}^{(1)}(v) - U_{L\ell}(+) \eta_{\ell}(v)) \]
\[ B_{R\ell} = X_{R\ell}(+) N_{HI} \eta_{\ell}^{(1)}(v) - U_{R\ell}(+) \eta_{\ell}(v) \]
\[ V_{L\ell} = X_{L\ell}(+\eta_{\ell}^{(2)}(v) - N_{HI} U_{L\ell}(+) \eta_{\ell}(v)) \]
\[ V_{R\ell} = X_{R\ell}(+) N_{HI} \eta_{\ell}^{(2)}(v) - U_{R\ell}(+) \eta_{\ell}(v) \]
\[ W_{L\ell} = X_{L\ell}(+\eta_{\ell}^{(3)}(v) - N_{HI} U_{L\ell}(+) \eta_{\ell}(v)) \]
\[ W_{R\ell} = X_{R\ell}(+) N_{HI} \eta_{\ell}^{(3)}(v) - U_{R\ell}(+) \eta_{\ell}(v) \]

We have also introduced the functions

\[ X_{R\ell}(\pm) = j_\ell(N_{R\ell}v) + D_{\ell} \eta_{\ell}^{(2)}(N_{R\ell}v) \pm D_{\ell} \eta_{\ell}^{(1)}(N_{R\ell}v) \]
\[ X_{L\ell}(\pm) = j_\ell(N_{L\ell}v) + D_{\ell} \eta_{\ell}^{(2)}(N_{L\ell}v) \pm D_{\ell} \eta_{\ell}^{(1)}(N_{L\ell}v) \]
\[ U_{R\ell}(\pm) = \eta_{\ell}^{(1)}(N_{R\ell}v) + D_{\ell} \eta_{\ell}^{(2)}(N_{R\ell}v) \pm D_{\ell} \eta_{\ell}^{(1)}(N_{R\ell}v) \]
\[ U_{L\ell}(\pm) = \eta_{\ell}^{(1)}(N_{L\ell}v) + D_{\ell} \eta_{\ell}^{(2)}(N_{L\ell}v) \pm D_{\ell} \eta_{\ell}^{(1)}(N_{L\ell}v) \]

where

\[ D_{\ell} = -\Delta_{\ell}^{-1}[G_{\ell}(N_{R\ell}) H_{\ell}(N_{L\ell}) + F_{\ell}(N_{R\ell}) K_{\ell}(N_{L\ell})] \]
\[ D_{2\ell} = -\Delta_{\ell}^{-1}[F_{\ell}(N_{R\ell}) K_{\ell}(N_{R\ell}) - G_{\ell}(N_{R\ell}) H_{\ell}(N_{R\ell})] \]
\[ D_{3\ell} = -\Delta_{\ell}^{-1}[G_{\ell}(N_{L\ell}) H_{\ell}(N_{L\ell}) - F_{\ell}(N_{L\ell}) K_{\ell}(N_{L\ell})] \]
\[ D_{4\ell} = -\Delta_{\ell}^{-1}[G_{\ell}(N_{L\ell}) H_{\ell}(N_{R\ell}) + F_{\ell}(N_{L\ell}) K_{\ell}(N_{R\ell})] \]

\[ F, G, H, K, \text{ and } \text{K} \text{ are functions of the refractive index variable } N = N_L, N_R \text{ defined as} \]

\[ F_{\ell}(N) = N_{HI} \eta_{\ell}(N_{\alpha}) \eta_{\ell}^{(1)}(N_{\alpha}) - N_{HI} \eta_{\ell}^{(2)}(N_{\alpha}) \eta_{\ell}(N_{\alpha}) \]
\[ G_{\ell}(N) = N_{HI} \eta_{\ell}(N_{\alpha}) \eta_{\ell}^{(1)}(N_{\alpha}) - N_{HI} \eta_{\ell}^{(2)}(N_{\alpha}) \eta_{\ell}(N_{\alpha}) \]
\[ H_{\ell}(N) = N_{HI} \eta_{\ell}(N_{\alpha}) \eta_{\ell}^{(1)}(N_{\alpha}) - N_{HI} \eta_{\ell}^{(2)}(N_{\alpha}) \eta_{\ell}(N_{\alpha}) \]
\[ K_{\ell}(N) = N_{HI} \eta_{\ell}(N_{\alpha}) \eta_{\ell}^{(1)}(N_{\alpha}) - N_{HI} \eta_{\ell}^{(2)}(N_{\alpha}) \eta_{\ell}(N_{\alpha}) \]
$j_\ell(\rho)$, $y_\ell(\rho)$ are the spherical Bessel functions of the first and second kind, respectively; whereas $h^{(1)}_\ell(\rho)$ is the spherical Hankel function of the first kind \cite{3}. We also define

\begin{align*}
\eta^{(1)}_\ell(\rho) &= \frac{1}{\rho} d[\rho j_\ell(\rho)]/d\rho \\
\eta^{(2)}_\ell(\rho) &= \frac{1}{\rho} d[\rho y_\ell(\rho)]/d\rho \\
\eta^{(3)}_\ell(\rho) &= \frac{1}{\rho} d[\rho h^{(1)}_\ell(\rho)]/d\rho
\end{align*}

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