The influence of chiral spherical particles on the radiation of optically active molecules

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\textit{New Journal of Physics 14} (2012) 123009 (19pp)
Received 25 July 2012
Published 7 December 2012
Online at \url{http://www.njp.org/}
doi:10.1088/1367-2630/14/12/123009

Abstract. Within the framework of nonrelativistic quantum electrodynamics, a theory of spontaneous emission of a chiral molecule located near a chiral (bi-isotropic) spherical particle is developed. It is shown that the structure of photons in the presence of chiral spherical particles differs significantly from the structure of usual transverse electric and transverse magnetic photons. Exact analytical expressions for a spontaneous emission decay rate of a chiral molecule with arbitrary electric and magnetic dipole moments of transition located near a chiral spherical particle with arbitrary parameters, are obtained and analyzed in detail. Simple asymptotes for the case of a nanoparticle are obtained. Substantial influence of even small chirality on a sphere made from dielectric or double negative metamaterial is found. It is shown that by using chiral spherical particles it is possible to effectively control the radiation of a given enantiomer of optically active molecules.
1. Introduction

Chirality is a geometric property of a three-dimensional body not to coincide with its reflection in a mirror in any shifts and turns. Such a property, for example, belongs to a human hand or a spring. The term ‘chirality’ was proposed by Lord Kelvin in 1873 to explain some special properties of the molecules [1]. Chirality plays an important role in biology and pharmacy, because many complex organic compounds (amino acids, proteins and sugars) have chiral properties. For this reason, a human body can react differently with different enantiomers of the same substance. For example, the same drug, depending on the type of its molecules, may have different taste and smell, or different affects. In physics, there is considerable interest in chiral media [2–6], in which there is a difference in the propagation of left- and right-handed circularly polarized electromagnetic waves. The simplest example of such an optically active medium is an aqueous solution of sugar. At this time, the study of chiral properties is again under special attention because of the possibility of creating metamaterials based on chiral objects [7–12]. Moreover, chiral metamaterials can help one to create negative refraction metamaterials, which have a great number of unusual applications [13].

The spontaneous emission of atoms and molecules in the vicinity of material bodies has been studied in a large number of works. The influence of dielectric microspheres on the spontaneous emission of atoms is considered in [14–17]; the spontaneous emission of an atom located near a microsphere made of double negative (DNG)-metamaterial has been examined in [18]. Optical properties of chiral spherical particles have also been studied quite extensively. At present, the scattering of plane electromagnetic waves on homogeneous and inhomogeneous [3–6, 19, 20] chiral microspheres, including multilayered chiral microspheres [21, 22], has already been considered. The scattering of a Hermite–Gauss laser beam on a chiral microsphere is regarded in [23], the scattering of a plane electromagnetic wave by a chiral sphere located in a chiral medium is investigated in [24]. The radiation pressure force acting on a chiral spherical particle placed in a circularly polarized wave is examined in [25] and the radiation of a point dipole source located inside a chiral spherical particle is described in [26]. Special attention should be paid to the recently developed theory of chiral molecule interaction with a spherical plasmonic nanoparticle [27, 28]. At the same time,
as far as we know, there is no investigation devoted to the influence of chiral spherical particles on the radiation of chiral (optically active) molecules. However, such processes become more and more important since they always appear when different biomolecules and drug molecules are investigated and modified by optical methods. So, in this paper we will present a detailed investigation of this problem.

The goal of this work is to develop an approach to a quantitative description of the influence of a chiral spherical particle made of an arbitrary metamaterial on the spontaneous emission of a chiral molecule. All results will be obtained in the framework of nonrelativistic quantum electrodynamics, under the assumption that the radiative linewidth of the molecule is much smaller than the radiation frequency. The last assumption allows us to use perturbation theory. The rest of the paper is organized as follows. In section 2, the canonical quantization of the electromagnetic field in the presence of a lossless chiral spherical particle with arbitrary permitivity and permeability is performed. In section 3, we obtain general expressions for the spontaneous emission radiative decay rate of a chiral molecule located near a chiral sphere. In section 4, the graphical illustration of the results obtained and their discussion are presented. The geometry of the problem is shown in figure 1.

2. Quantization of the electromagnetic field in the presence of a lossless chiral spherical particle

To solve the problem of quantization of the electromagnetic field in the presence of a lossless chiral spherical particle, let us consider a spherical cavity of infinite radius $\Lambda \rightarrow \infty$ with a perfectly conducting wall and with the chiral particle located at its center. For simplicity, we assume that the nanoparticle is surrounded by a vacuum (see figure 1). The usual procedure of quantization of the electromagnetic field in spherical geometry [29–31] is inapplicable in the case of chiral particles; below we will develop a new approach which is valid in this case.
In the description of classical fields in the presence of chiral spherical particles, we will use the method proposed in [4, 32]. At the same time, to describe the chiral medium, we use the constitutive equations in the Drude–Born–Fedorov form [33–35]

\[
\mathbf{D} = \varepsilon \left( \mathbf{E} + \eta \text{rot} \mathbf{E} \right), \quad \mathbf{B} = \mu \left( \mathbf{H} + \eta \text{rot} \mathbf{H} \right),
\]

where \( \mathbf{D}, \mathbf{E} \) and \( \mathbf{B}, \mathbf{H} \) are the inductions and the strengths of the electric and magnetic fields, correspondingly, \( \varepsilon, \mu \) are the permittivity and the permeability of the chiral medium and \( \eta \) is the dimensional parameter of chirality. The choice of constitutive equations in the form (1) is arbitrary to some extent. Another possibility is to use the so called Boys–Post form:

\[
\mathbf{D} = \varepsilon \mathbf{E} + \frac{\kappa}{c} \mathbf{B}, \quad \mathbf{H} = \frac{1}{\mu} \mathbf{B} + \frac{\kappa}{c} \mathbf{E}.
\]

(2)

There are some indications that for the field quantization the Boys–Post form is even more fundamental than (1) [36]. We will compare QED of particles with the constitutive equations (1) and (2) in a separate publication.

In accordance with the general rules of electromagnetic field quantization, its expansion in the complete system of eigenfunctions of the classical problem (standing spherical waves) can be presented as follows:

\[
\hat{\mathbf{E}}(\mathbf{r}) = i \sum_s a_s \mathbf{e}(s, \mathbf{r}) - a_s^\dagger \mathbf{e}^*(s, \mathbf{r}), \quad \hat{\mathbf{H}}(\mathbf{r}) = \sum_s a_s \mathbf{h}(s, \mathbf{r}) + a_s^\dagger \mathbf{h}^*(s, \mathbf{r}),
\]

where \( a_s \) and \( a_s^\dagger \) are the operators of annihilation and creation of a photon in \( s \)th mode respectively, with usual rules of commutation \([a_s, a_s^\dagger] = 1\); \( \mathbf{e}(s, \mathbf{r}) \) and \( \mathbf{h}(s, \mathbf{r}) \), the modes of the electric and magnetic fields with definite energies; the vector index \( s = (n, m, \nu) \) denotes a chosen set of quantum numbers: the orbital \((n)\), azimuthal \((m)\) and radial \((\nu)\); the asterisk denotes the operation of complex conjugation.

With the help of Bohren’s transformation [32], the eigenmodes inside the chiral sphere can be written as

\[
\begin{align*}
\mathbf{e}^{in}(s, \mathbf{r}) &= A^{(L)}_{mn} \mathbf{Q}^{(L)}_{mn} + A^{(R)}_{mn} \mathbf{Q}^{(R)}_{mn}, \\
\mathbf{h}^{in}(s, \mathbf{r}) &= -\frac{\sqrt{\varepsilon \mu}}{\mu} \left( A^{(L)}_{mn} \mathbf{Q}^{(L)}_{mn} - A^{(R)}_{mn} \mathbf{Q}^{(R)}_{mn} \right),
\end{align*}
\]

where the left \( \mathbf{Q}^{(L)}_{mn} \) and right \( \mathbf{Q}^{(R)}_{mn} \) contributions correspond to the propagation of the left and right circularly polarized waves with wavenumbers \( k_\text{L} = \frac{k_0}{1 - i \chi} \) and \( k_\text{R} = \frac{k_0}{1 + i \chi} \), where \( k_0 = \omega / c \) is the wavenumber in a vacuum; \( \omega \), the frequency; \( c \), the speed of light in a vacuum and \( \chi = k_0 \eta \) is the dimensionless parameter of chirality. In (4) \( A^{(L)}_{mn} \) and \( A^{(R)}_{mn} \) are the coefficients of expansion. In (3) and everywhere further, the time dependence \( \exp(-i \omega t) \) is assumed. The vector functions \( \mathbf{Q}^{(L)}_{mn} \) and \( \mathbf{Q}^{(R)}_{mn} \) can be written in the following form:

\[
\begin{align*}
\mathbf{Q}^{(L)}_{mn} &= N \psi^{(L)}_{mn} + M \psi^{(L)*}_{mn}, \\
\mathbf{Q}^{(R)}_{mn} &= N \psi^{(R)}_{mn} - M \psi^{(R)*}_{mn},
\end{align*}
\]

(5)

Explicit expressions for spherical vector harmonics \( N \psi^{(J)}_{mn} \) and \( M \psi^{(J)}_{mn} \) (where \( J = \text{L, R} \)) are given in appendix A.

From (5), it follows that \( \mathbf{Q}^{(L)}_{mn} \) and \( \mathbf{Q}^{(R)}_{mn} \) are expressed in terms of the vector spherical harmonics \( N \psi^{(L)}_{mn} + M \psi^{(L)*}_{mn} \) and \( N \psi^{(R)}_{mn} - M \psi^{(R)*}_{mn} \), which always have a nonzero component along the radius (see appendix A) and, therefore, can never be reduced to the usual transverse electric (TE) and transverse magnetic (TM) fields.

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Outside the chiral particle, all the waves have the same propagation velocity and can be represented as

\[ \mathbf{e}^{\text{out}}(s, \mathbf{r}) = C_{mn}^{(1)} \mathbf{N} \xi_{mn}^{(1)} + D_{mn}^{(1)} \mathbf{M} \xi_{mn}^{(1)} + C_{mn}^{(2)} \mathbf{N} \xi_{mn}^{(2)} + D_{mn}^{(2)} \mathbf{M} \xi_{mn}^{(2)}, \]

\[ \mathbf{h}^{\text{out}}(s, \mathbf{r}) = - (C_{mn}^{(1)} \mathbf{M} \xi_{mn}^{(1)} + D_{mn}^{(1)} \mathbf{N} \xi_{mn}^{(1)} + C_{mn}^{(2)} \mathbf{M} \xi_{mn}^{(2)} + D_{mn}^{(2)} \mathbf{N} \xi_{mn}^{(2)}), \] (6)

where \( C_{mn}^{(1)}, C_{mn}^{(2)}, \) and \( D_{mn}^{(1)}, D_{mn}^{(2)} \) are some coefficients; expressions for the spherical vector harmonics \( \mathbf{N} \xi_{mn}^{(j)} \) and \( \mathbf{M} \xi_{mn}^{(j)} \) (where \( j = 1, 2 \)) are given in appendix A.

To find the coefficients in (4) and (6), one should use the condition of continuity of tangential components of the electric and magnetic fields on the surface of a chiral spherical particle having radius \( a \) and the normalization of photon energy in the mode to \( \hbar \omega_n \), where \( \omega_n \) is the mode eigenfrequency. Since the molecule is situated outside the particle, we do not need to know explicit values of the coefficients \( A_{mn}^{(1)} \) and \( A_{mn}^{(2)} \) (see (4)). For the coefficients that describe the fields outside the sphere, one can obtain the following relations (\( n = 1, 2, 3, \ldots; m = 0, \pm 1, \pm 2, \ldots, \pm n \)):

\[ C_{mn}^{(1)} = \alpha_n C_{mn}^{(2)} + \beta_n D_{mn}^{(2)}, \quad D_{mn}^{(1)} = \beta_n C_{mn}^{(2)} + \gamma_n D_{mn}^{(2)}, \]

\[ \frac{D_{mn}^{(2)}}{C_{mn}^{(2)}} = - \frac{1}{2\beta_n} \left( \alpha_n + \gamma_n + \sqrt{(\alpha_n + \gamma_n)^2 - 4\beta_n^2} \right) = (O_n)^{\pm 1}, \]

\[ \left| C_{mn}^{(2)} \right|^2 + \left| D_{mn}^{(2)} \right|^2 = \frac{\hbar \omega_s^3}{2\Lambda c^2} \frac{(2n + 1)}{n(n+1)(n+m)!}, \] (7)

where the coefficients \( \alpha_n, \beta_n, \) and \( \gamma_n \) are given in appendix B.

The relations (7) define two types of electromagnetic modes (photons) occurring in the presence of a chiral spherical particle, which are analogous to TM and TE modes in the case of a nonchiral spherical particle. The upper sign \((-\) before the square root in (7) corresponds to the modes that we will call \( A \)-type photons, while the sign \( + \) gives us \( B \)-type photons. If the chirality parameter tends to zero \((\chi \to 0)\), then, as follows from (7), the ratio \( \frac{D_{mn}^{(2)}}{C_{mn}^{(2)}} \) tends to zero or to infinity; this leads to a decoupling between TE and TM modes (vector spherical harmonics \( \mathbf{N} \xi_{mn}^{(2)} \) and \( \mathbf{M} \xi_{mn}^{(2)} \) in (6)) and, as a result, leads to the appearance of the usual TM or TE polarized photons.

Thus, in the case of a chiral spherical particle, we have two types of polarization \((A\) and \(B)\), which differ from the TM and TE polarizations.

The frequencies of these modes are determined by asymptotic expressions \((\Lambda \to \infty)\)

\[ \left( \frac{\omega_s}{c} \right)_A \approx \frac{\pi}{\Lambda} \left( v + \frac{n + 1}{2} + \frac{i}{2\pi} \text{Ln} (\alpha_n + \beta_n O_n) \right) + \cdots, \]

\[ \left( \frac{\omega_s}{c} \right)_B \approx \frac{\pi}{\Lambda} \left( v + \frac{n}{2} + \frac{i}{2\pi} \text{Ln} (\gamma_n + \beta_n O_n) \right) + \cdots, \] (8)

It is very important that these eigenfrequencies are positive for arbitrary parameters of the sphere.

From (8), it follows immediately that the density of the final states for both \( A \)-type and \( B \)-type photons has the same value:

\[ \rho(\omega) = \frac{d\nu}{d(\hbar\omega)} = \frac{\Lambda}{\pi \hbar c}, \] (9)
and does not depend on the presence of a spherical particle of a final volume in the cavity, which is consistent with Courant’s theorem [37].

Substituting $\hat{E}$, $\hat{H}$ from (3) and $\hat{D}$, $\hat{B}$ (see (1)) into the expression for the free field Hamiltonian

$$H = \frac{1}{8\pi} \int_V (\hat{E} \hat{D} + \hat{B} \hat{H}) \, dV$$  \hspace{1cm} (10)

and using the properties of the eigenfunctions $e(s,r)$ and $h(s,r)$, we obtain the canonical expression for the Hamiltonian through the creation and annihilation operators:

$$H = \sum_s \hbar \omega_s \left( a_s^\dagger a_s + \frac{1}{2} \right).$$  \hspace{1cm} (11)

Thus, the expressions (6) and (7), together with the expression for the density of the final states (9), describe fully the quantized electromagnetic field in the presence of a chiral spherical particle; this allows one to describe the interaction of this field with arbitrary atoms and molecules. In the next section, we will apply these expressions to find the radiative decay rate of the spontaneous emission of a chiral molecule.

3. Radiative decay rate of the spontaneous emission of a chiral molecule located near a chiral spherical particle

The radiative decay rate of the spontaneous emission (the radiative linewidth $\gamma$) of a chiral molecule located near a lossless chiral spherical particle can be found by making use of Fermi’s ‘golden rule’ [38]:

$$\gamma = \frac{2\pi}{\hbar} \sum_{\text{final}} \left| \langle \text{initial} | H_{\text{int}} | \text{final} \rangle \right|^2 \rho(\omega),$$  \hspace{1cm} (12)

where the summation is performed over all possible final states, which include both types of photons ($A$ and $B$, see (7)), $\rho(\omega)$ is the density of the final states (9), and $H_{\text{int}}$ is the interaction Hamiltonian of a chiral molecule with the electromagnetic field. In this work, we assume that the Hamiltonian is Hermitian, i.e. there are no losses in the material of a particle. If there are losses, there is a possibility of nonradiative transition from the excited state to the ground state. The total decay rate, which includes the nonradiative contribution, can be found within the framework of a very general QED approach [39, 40], within the classical theory [41] or within the nonstandard quantization scheme [42–45]. In further calculations, for brevity we will consider only one channel of the decay of the initial state of the molecule (transition $e \rightarrow g$), i.e. a two-level molecule. To take into account the possibility of the transition of the molecule into several states, it is enough to sum the partial linewidths.

The Hamiltonian of the interaction of a chiral molecule with the electromagnetic field in (12) can be written down in the following form [46]:

$$H_{\text{int}} = - (\hat{d} \hat{E}(r_0)) - (\hat{m} \hat{H}(r_0)),$$  \hspace{1cm} (13)

where $r_0$ is the radius vector of the molecule position; $\hat{d}$ and $\hat{m}$, the operators of the electric and the magnetic dipole moments of the molecule; the field operators $\hat{E}$ and $\hat{H}$ are defined in (3). We suppose that the initial state corresponds to the vacuum state of the field and to the molecule in the excited state, $\langle \text{initial} \rangle = \langle e \rangle |\text{vac}\rangle$; also that the final field state corresponds to the single
the ‘left’ molecules. Explicit expressions for the component of \( d \) nonzero, and the explicit expressions for the decay rate \( \gamma \) magnetic dipole matrix elements. Let us, for clarity, call the molecules with parallel \( d \) complicated chiral molecules, there can be a different phase relation between the electric and total radiative decay rate of spontaneous emission of the molecule:

\[
\gamma_{eg}^A = \gamma_{eg}^A + \gamma_{eg}^B,
\]

(14)

where, for example, in the case of \( A \)-type photons

\[
\gamma_{eg}^A = \frac{\pi}{\hbar} \rho(\omega) \sum_{n,m} \left| \left( \mathbf{d}_n \mathbf{e}_{A}^{\text{at}}(n, m, v, \mathbf{r}_0) - \mathbf{m}_0 \mathbf{h}_{A}^{\text{at}}(n, m, v, \mathbf{r}_0) \right) \right|^2,
\]

(15)

where \( \langle \psi_0 | \mathbf{d} | g \rangle = \mathbf{d}_0 \) and \( \langle \psi | \mathbf{m} | g \rangle = -i \mathbf{m}_0 \) are the dipole moments of the considered transition of the molecule at the frequency \( \omega \approx \omega_0 \) (see (8)). The chosen phase difference between the electric and magnetic dipoles is due to the fact that the magnetic moment operator is purely imaginary. This definition also agrees with the adopted spiral model of a molecule. For more complicated chiral molecules, there can be a different phase relation between the electric and magnetic dipole matrix elements. Let us, for clarity, call the molecules with parallel \( \mathbf{d}_0 \) and \( \mathbf{m}_0 \) the ‘right’ molecules, while the molecules with antiparallel \( \mathbf{d}_0 \) and \( \mathbf{m}_0 \) will be referred to as the ‘left’ molecules. Explicit expressions for the component of (14) for the mode of photons of \( B \)-type can be obtained from (15) by replacing the index \( A \to B \).

Let us assume for definiteness that the molecule is located on the \( z \)-axis of a Cartesian coordinate system at the point \( r_0 > a \). In this case, only the components with \( m = 0, \pm 1 \) are nonzero, and the explicit expressions for the decay rate (15) will have the following form:

\[
\gamma_{eg}^A = \gamma_{eg}^{A,-1} + \gamma_{eg}^{A,1} + \gamma_{eg}^{A,0},
\]

(16)

where

\[
\gamma_{eg}^{A,-1} = \frac{k_0}{2\hbar r_0^2} \sum_{n=1}^{\infty} \frac{2n+1}{1+|O_n|^2} |(d_{0x} - i d_{0y})(\psi_n^0(k_0 r_0) + T_n^{A \xi_n^{(1)}}(k_0 r_0))
- O_n(d_{0y} + i d_{0x})(\psi_n^0(k_0 r_0) + L_n^{A \xi_n^{(1)}}(k_0 r_0))
+ O_n(m_{0x} - i m_{0y})(\psi_n^0(k_0 r_0) + L_n^{A \xi_n^{(1)}}(k_0 r_0))
-(m_{0y} + i m_{0x})(\psi_n^0(k_0 r_0) + T_n^{A \xi_n^{(1)}}(k_0 r_0))|^2,
\]

(17)

\[
\gamma_{eg}^{A,1} = \frac{k_0}{2\hbar r_0^2} \sum_{n=1}^{\infty} \frac{2n+1}{1+|O_n|^2} |O_n(d_{0y} - i d_{0x})(\psi_n^0(k_0 r_0) + L_n^{A \xi_n^{(1)}}(k_0 r_0))
-(d_{0x} + i d_{0y})(\psi_n^0(k_0 r_0) + T_n^{A \xi_n^{(1)}}(k_0 r_0))
+(m_{0x} - i m_{0y})(\psi_n^0(k_0 r_0) + T_n^{A \xi_n^{(1)}}(k_0 r_0))
-O_n(m_{0y} + i m_{0x})(\psi_n^0(k_0 r_0) + L_n^{A \xi_n^{(1)}}(k_0 r_0))|^2,
\]

(18)

and

\[
\gamma_{eg}^{A,0} = \frac{2}{\hbar k_0 r_0^2} \sum_{n=1}^{\infty} \frac{(2n+1)n(n+1)}{1+|O_n|^2} |d_{0z}(\psi_n^0(k_0 r_0) + T_n^{A \xi_n^{(1)}}(k_0 r_0))
+O_n m_{0z}(\psi_n^0(k_0 r_0) + T_n^{A \xi_n^{(1)}}(k_0 r_0))|^2.
\]

(19)

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In the case of the photon mode of $B$-type, one can obtain

\[ \gamma_{eg}^B = \gamma_{eg}^{B,-1} + \gamma_{eg}^{B,1} + \gamma_{eg}^{B,0}, \]

where

\[ \gamma_{eg}^{B,-1} = \frac{k_0}{2 \hbar r_0} \sum_{n=1}^{\infty} \frac{2n + 1}{1 + |O_n|^2} \left| O_n(d_{0x} - id_{0y})(\psi_n(k_0r_0) + T^{B\,\xi^{(1)}_n}(k_0r_0)) ight. \\
- (d_{0y} + id_{0x})(\psi_n(k_0r_0) + T^{B\,\xi^{(1)}_n}(k_0r_0)) \\
+ (m_{0x} - im_{0y})(\psi_n(k_0r_0) + T^{B\,\xi^{(1)}_n}(k_0r_0)) \\
- O_n(m_{0y} + im_{0x})(\psi_n(k_0r_0) + T^{B\,\xi^{(1)}_n}(k_0r_0)) |^2. \]

\[ \gamma_{eg}^{B,1} = \frac{k_0}{2 \hbar r_0} \sum_{n=1}^{\infty} \frac{2n + 1}{1 + |O_n|^2} \left| (d_{0y} - id_{0x})(\psi'_n(k_0r_0) + L^{B\,\xi^{(1)}_n}(k_0r_0)) ight. \\
- O_n(d_{0x} + id_{0y})(\psi'_n(k_0r_0) + T^{B\,\xi^{(1)}_n}(k_0r_0)) \\
+ O_n(m_{0y} - im_{0x})(\psi_n(k_0r_0) + T^{B\,\xi^{(1)}_n}(k_0r_0)) \\
- (m_{0x} + im_{0y})(\psi'_n(k_0r_0) + L^{B\,\xi^{(1)}_n}(k_0r_0)) |^2, \]

and

\[ \gamma_{eg}^{B,0} = \frac{2}{\hbar k_0 r_0^4} \sum_{n=1}^{\infty} \frac{(2n + 1)^2}{O_n^2} \left| O_n d_{0z} \left( \psi_n(k_0r_0) + T^{B\,\xi^{(1)}_n}(k_0r_0) \right) ight. \\
+ m_{0z} \left( \psi_n(k_0r_0) + L^{B\,\xi^{(1)}_n}(k_0r_0) \right) |^2. \]

In (17)–(19), (21)–(23), the coefficient $O_n$ is defined in (7), the functions $\psi_n(k_0r_0)$ and $\xi^{(1)}_n(k_0r_0)$ are defined in appendix A, the prime to a function means the derivative with respect to its argument; we have used the following notations:

\[ T^A_n = \frac{1}{4} \left( \alpha_n - \gamma_n + \sqrt{(\alpha_n + \gamma_n)^2 - 4\beta^2_n} - 2 \right), \]

\[ L^A_n = \frac{1}{4} \left( \gamma_n - \alpha_n - \sqrt{(\alpha_n + \gamma_n)^2 - 4\beta^2_n} - 2 \right), \]

\[ T^B_n = \frac{1}{4} \left( \alpha_n - \gamma_n - \sqrt{(\alpha_n + \gamma_n)^2 - 4\beta^2_n} - 2 \right), \]

\[ L^B_n = \frac{1}{4} \left( \gamma_n - \alpha_n + \sqrt{(\alpha_n + \gamma_n)^2 - 4\beta^2_n} - 2 \right). \]

Note, that despite the fact that this derivation has been performed with the assumption of the absence of losses in the material of the particle, the expressions obtained will also describe exactly the radiative losses for nanoparticles made of real materials, i.e. for complex $\varepsilon$ and $\mu$. This follows from the fact that classical and quantum calculations give the same analytical expressions for the radiative decay rate of spontaneous emission through Green’s function of a classical problem [39, 40]. Since the classical approach is valid in the case of both absorbing and nonabsorbing bodies, this means that for the generalization of the expressions obtained for
the case of a real (absorbing) material of a spherical particle, it is only necessary to substitute
the appropriate complex values of dielectric permittivity and magnetic permeability into the
obtained expressions.

It should be noted that our QED expressions for the radiative decay rates in the lossless
case coincide with our calculations of the total decay rate, based on the approach described
in [39, 40].

To calculate the relative radiative decay rate of the spontaneous emission of chiral
molecules located near a chiral spherical particle, one should normalize the expression (14) to
the value of the radiative decay rate of the spontaneous emission of the molecule in the absence
of the particle

\[ \gamma_{0, eg} = \frac{4k_0^3}{3\hbar} |d_0|^2 + \frac{4k_0^3}{3\hbar} |m_0|^2. \]

(25)

4. Analysis of the results obtained and illustrations

From the cumbersome formulas obtained in the previous section, it is difficult to discover all
the features of the spontaneous decay of a chiral molecule in the system under consideration. In
this section, we will explore the expressions found in some typical cases.

To find an expression for the spontaneous emission radiative decay rate in the case of a
nanoparticle, one should expand \( n = 1 \) terms in (17)–(19), (21)–(23) into a series of \( k_0 a \rightarrow 0 \)
and keep only the principal terms. As a result, one has

\[
\gamma_{eg} = \frac{4k_0^3}{3\hbar} \left| d_0 \right|^2 \left( 1 - \frac{\alpha_{EE}}{r_0^3} \right) + \frac{4k_0^3}{3\hbar} \left| m_0 \right|^2 \left( 1 - \frac{\alpha_{HH}}{r_0^3} \right) - \frac{2i}{3\hbar} \chi \left( \frac{\epsilon - 1}{(\epsilon + 2)(\mu + 2)} - 4\chi^2 \frac{\epsilon \mu}{(\epsilon + 2)(\mu + 2)} \right) \left( 1 + \frac{2\alpha_{HE}}{r_0^3} \right) + \frac{2i}{3\hbar} \chi \left( \frac{\epsilon - 1}{(\epsilon + 2)(\mu + 2)} - 4\chi^2 \frac{\epsilon \mu}{(\epsilon + 2)(\mu + 2)} \right) \left( 1 + \frac{2\alpha_{HE}}{r_0^3} \right) \left( 1 + \frac{2\alpha_{HE}}{r_0^3} \right),
\]

(26)

where we have used the abbreviations

\[
\alpha_{EE} = a^3 \frac{(\epsilon - 1)(\mu + 2) + 2\chi^2 \epsilon \mu}{(\epsilon + 2)(\mu + 2) - 4\chi^2 \epsilon \mu},
\]

\[
\alpha_{HH} = a^3 \frac{(\mu - 1)(\epsilon + 2) + 2\chi^2 \epsilon \mu}{(\epsilon + 2)(\mu + 2) - 4\chi^2 \epsilon \mu},
\]

\[
\alpha_{HE} = -\alpha_{HE} = a^3 \frac{3\chi \epsilon \mu}{(\epsilon + 2)(\mu + 2) - 4\chi^2 \epsilon \mu},
\]

(27)

which, as can be shown, coincide with the electromagnetic polarizabilities of a chiral spherical
particle in the quasistatic limit [47, 48].

To give an additional physical insight into (26), let us derive an expression for the radiative
decay rate of a chiral nanoparticle within the quasistatic classical approach suggested in [47].

If the nanoparticle and the molecule are placed close enough, then their total radiation will be
determined by their total electric and magnetic dipole moments:

\[
\gamma_{eg} = \frac{4k_0^3}{3\hbar} |d_0 + \delta d|^2 + \frac{4k_0^3}{3\hbar} |-im_0 + \delta m|^2,
\]

(28)
where $\delta d$ and $\delta m$ are the electric and the magnetic dipole moments induced in the nanoparticle, while $d_0$ and $-im_0$ are the oscillating dipole moments of the molecule. Explicit expressions for induced dipole momenta have the following form:

$$\delta d = \alpha_{EE} E_0 (r_0) + \alpha_{EH} H_0 (r_0),$$

$$\delta m = \alpha_{HE} E_0 (r_0) + \alpha_{HH} H_0 (r_0),$$

where the electromagnetic polarizabilities of a chiral spherical nanoparticle in a homogeneous field are given by (27). The electric and magnetic near fields from the radiating molecule also have well known expressions:

$$E_0 (r) = \frac{3n (nd_0) - d_0}{r^3}, \quad H_0 (r) = -i \frac{3n (nm_0) - m_0}{r^3},$$

where $n$ is the unit vector in the direction from the center of the particle to the observation point.

Substituting (29) and (30) into (28) we get the expression for the radiative decay rate of a chiral molecule near a chiral nanosphere

$$\gamma_{eg} = \frac{4k_0^3}{3\hbar} \left| d_0 + \frac{\alpha_{EE}}{r_0^3} (3n (nd_0) - d_0) - \frac{i\alpha_{EH}}{r_0^3} (3n (nm_0) - m_0) \right|^2$$

$$+ \frac{4k_0^3}{3\hbar} \left| m_0 + \frac{i\alpha_{HE}}{r_0^3} (3n (nd_0) - d_0) + \frac{\alpha_{HH}}{r_0^3} (3n (nm_0) - m_0) \right|^2. \tag{31}$$

It is easy to check that this expression is identical to (26) for the one chosen in (26) for the specific position of the molecule. This coincidence gives us an additional check of our tedious QED calculation. On the other hand, the quasistatic derivation of the decay rate gives us an understanding of the physical processes involved.

The condition that the denominator of (27) is zero one

$$(\varepsilon + 2) (\mu + 2) - 4\chi^2 \varepsilon \mu = 0$$

(32)

corresponds to the condition of a chiral-plasmon resonance occurring in a spherical nanoparticle. The roots of (32) determine the values of the constants $\varepsilon$ and $\mu$, that correspond to the chiral plasmon oscillations. It should be noted that the resonant values of $\varepsilon$ and $\mu$ are not independent; their values differ from the corresponding values in the case of plasmon resonance in a nonchiral particle, where (32) is split into two independent equations, $\varepsilon + 2 = 0$ and $\mu + 2 = 0$ [49].

In practice, the orientation of the molecule can be arbitrary with respect to the nanoparticle surface; therefore, to get an effective radiative decay rate of spontaneous emission, one should average (31) over the orientations of molecules, or, equivalently, over the unit vector $n$. As a result we obtain

$$\gamma_{eg}^{\text{eff}} = \frac{4k_0^3|d_0|^2}{3\hbar} \left\{ 1 + \frac{2}{r_0^2} |\alpha_{EE} - i\xi \alpha_{EH}|^2 + |\xi|^2 \right\} + \frac{2}{r_0^2} |\alpha_{HE} - i\xi \alpha_{HH}|^2, \tag{33}$$

where $\xi$ is defined by the relation $m_0 = \xi d_0$.

As a rule, the magnetic dipole moment of the molecule is much smaller than the electric dipole moment $|m_0| \ll |d_0|$. The chirality parameter is usually small ($\chi \ll 1$) (see, however, [50]). This fact determines that the second term on the right-hand side of (33), corresponding to the induced electric dipole moment, is usually greater than the term corresponding to the induced magnetic dipole moment (fourth term in (33)).

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Thus, effective interference between the electric and magnetic fields is possible only under the following two conditions:

1. In the system under consideration, a chiral-plasmon resonance must be present, i.e. the condition (32) must be satisfied. Under this condition, the contribution of the magnetic radiation increases.

2. The electric dipole moment induced in the nanoparticle should be minimized, i.e. the following condition must be satisfied (see (33)):

$$\alpha_{ee} - i\xi \alpha_{eh} = 0.$$  \hspace{1cm} (34)

The solution of the system of equations (32) and (34) determines the values of the dielectric permittivity and magnetic permeability of the nanoparticle, which correspond to the minimal values of the radiative decay rate of a chiral molecule. This means that the interference between the electric dipole and magnetic dipole radiation becomes maximal and destructive if

$$\mu \rightarrow -\frac{2d_0}{d_0 + 2\chi m_0}, \quad \varepsilon \rightarrow -\frac{2m_0}{m_0 + 2\chi d_0},$$  \hspace{1cm} (35)

where $d_0$ and $m_0$ are the Cartesian projections of the corresponding vectors. From (35), it follows that by changing the sign in $m_0$, i.e. when the chirality of the molecule is changed, the ‘resonant’ magnetic permeability $\mu$ varies only slightly ($\chi \ll 1, |m_0| \ll |d_0|$) and approximately equals $-2$. On the other hand, the ‘resonant’ dielectric permittivity $\varepsilon$ may have different signs for molecules with different chirality. This means that both nanoparticles with simultaneously negative $\varepsilon$ and $\mu$ (DNG-metamaterials) and nanoparticles with negative $\mu$ and positive $\varepsilon$ ($\mu$ negative (MNG)-metamaterials or magnetic plasma) are suitable for the effective control of the radiation of chiral molecules. From the viewpoint of the practical implementation of such nanoparticles, the most suitable are nanoparticles with negative $\mu$ and positive $\varepsilon$, because they can be implemented using the well developed technology of split ring resonators [51, 52]. On the other hand, chiral DNG-metamaterials can be also synthesized [12].

One should note that all the results obtained (including (35)) are valid for any values of the chirality parameter and of the relation between the electric and magnetic dipole moments, not only for the small ones. For a larger chirality parameter (see e.g. [50]), one can expect even more pronounced effects.

The above expressions give a good description of the properties of arbitrary chiral molecule radiation of nearby nanoparticles, i.e. when the retardation can be ignored. When the retardation effects are significant, it is necessary to use the full expression for the decay rates given in the previous section.

In any case, the process of the spontaneous decay of a chiral molecule located near a chiral spherical particle is very complicated; below, for clarity, we will give graphical illustrations of some possible regimes of the interaction of the chiral molecule with nanoparticles of different composition but neglecting the losses in them.

Figure 2 shows the distribution of absolute values of the reflection coefficient $T_A^4$ (see (24)) depending on the dielectric permittivity and the magnetic permeability of the material of nonchiral (see figure 2(a)) and chiral (figure 2(b)) spherical particles for different values of $k_0d$. Large values of this coefficient correspond to a resonance. This figure clearly shows that there is a significant difference between these cases, as reflected in the fact that nonzero chirality of the particle leads both to an increase in the number of observed modes (whispering gallery...
Figure 2. The dependence of the absolute values of the coefficient $T_{4A}$ on the permittivity ($\varepsilon$) and the permeability ($\mu$) of a chiral spherical particle for different values of $k_0a$. (a) The chirality parameter $\chi = 0$. (b) The chirality parameter $\chi = 0.1$. The particle is placed in a vacuum.

It is very interesting that even for a dielectric sphere with a small admixture of chirality, strong and unexpected effects occur. In figure 3, the radiative decay rate of the spontaneous emission of a chiral molecule placed near a chiral spherical particle with positive values of permittivity and permeability is shown as a function of the size parameter. As seen from this figure, the presence of chirality in a spherical particle leads to a shift of the maxima of the spontaneous emission radiative decay rate in comparison with the case of a nonchiral particle (cf peaks in figure 3 associated with different orbital quantum numbers $n$). Indeed, as one knows, a condition that allows us to estimate the position of the resonance is given by...
Figure 3. The radiative decay rate of the spontaneous emission of a chiral molecule located in the close vicinity of the surface of a chiral dielectric spherical particle \((r_0 \to a)\) with \(\varepsilon = 6\) and \(\mu = 1\) as a function of its size, \(k_0 a\). (a) The transition electric dipole moment of the molecule is oriented along the \(x\)-axis \((d_0 = d_0 e_x)\), i.e. it is tangential to the surface of the particle. (b) The transition electric dipole moment of the molecule is oriented along the \(z\)-axis \((d_0 = d_0 e_z)\), i.e. it is normal to the surface of the particle. The solid line corresponds to the transition magnetic dipole moment of a molecule oriented along the \(x\)-axis and the dashed line corresponds to the transition magnetic dipole moment of a molecule oriented along the \(z\)-axis. The value of the transition magnetic dipole moment \(\{m_0x, m_0z\} = 0\). The particle is placed in a vacuum.

\[ k_0 a \sqrt{\varepsilon \mu} \approx n + 1/2. \] In the case of chiral spherical particles, the left- and right-polarized waves exist simultaneously and \(k_{L} a > k_0 a \sqrt{\varepsilon \mu}\) if \(\chi > 0\). Hence, the above condition should be changed to \(k_{L} a \approx n + 1/2\). As a result, this leads to smaller resonant values of \(k_0 a\) than those of a nonchiral particle (see figure 2). An even more interesting feature of ordinary dielectric particles with a small admixture of chirality is a substantial increase in the quality factor of the whispering gallery modes. This figure clearly shows that the corresponding linewidth can be increased by a factor of 9, or even more.

Note, also, that in figure 3, only a slight difference between the radiative decay rates of spontaneous emission corresponding to different orientations of the transition magnetic dipole moment of a chiral molecule at a fixed electric dipole moment is observed. However, one should expect a growth of this difference in the case of a gradual increase of absolute values of the magnetic dipole momentum.

Figure 4 shows the radiative decay rate of the spontaneous emission of a chiral molecule located near chiral spherical nanoparticles \((k_0 a = 1)\) as a function of the chirality \(\chi = k_0 \eta\). As is clearly seen from this figure, the change of the sphere chirality has the greatest impact on the rate of the spontaneous decay of the molecules near dielectric and DNG spherical particles, in which high-Q modes can be excited. When the chirality parameter approaches the critical value \(\chi_{crit} = 1/\sqrt{\varepsilon \mu}\), \(k_{L} \to \infty\) and the number of oscillations in the radiative decay rate increases rapidly. In the case of metallic particles, the dependence of the radiative decay rate of the
Figure 4. The radiative decay rate of the spontaneous emission of a chiral molecule located in the close vicinity of the surface of a chiral spherical particle \((r_0 \to a)\) of size \(k_0 a = 1\), as a function of the chirality parameter \(\chi\). A dielectric sphere has \(\epsilon = 4\) and \(\mu = 1\), a metal sphere has \(\epsilon = -4\) and \(\mu = 1\), a DNG sphere has \(\epsilon = -4\) and \(\mu = -1.11\). (a) The transition electric dipole moment of the molecule is oriented along the \(x\)-axis \((d_0 = d_0 \hat{e}_x)\), i.e. it is tangential to the surface of the particle. (b) The transition electric dipole moment of the molecule is oriented along the \(z\)-axis \((d_0 = d_0 \hat{e}_z)\), i.e. it is normal to the surface of the particle. The solid line corresponds to the transition magnetic dipole moment of the molecule oriented along the \(x\)-axis and the dashed line corresponds to the transition magnetic dipole moment of the molecule oriented along the \(z\)-axis. The value of the transition magnetic dipole moment \(\{m_0x, m_0z\} = 0.1d_0\). The particle is placed in a vacuum.

spontaneous emission on the parameter \(\chi\), which is shown in figure 4, is weak, due to imaginary values of \(\sqrt{\epsilon \mu}\) and the absence of propagating waves.

Chiral molecules play an especially important role in biology and pharmacy. Therefore, it is extremely important to arrange the separation of the ‘right’ and ‘left’ enantiomers of molecules in the racemic mixtures. This can be done in various ways, for example, using the radiation pressure forces in the electromagnetic field of the left- or right-handed circularly polarized electromagnetic waves [25], or using spiral optical beams [53]. In addition, such a selection can be efficiently performed with the help of chiral nanoparticles [47].

Figure 5 shows the radiative decay rate of the spontaneous emission of a chiral molecule located near a DNG chiral spherical nanoparticle as a function of the dielectric constant for a fixed value of the magnetic permeability. It is seen from the figure that for the molecules which differ only in the orientation of the magnetic dipole moment of transition (the ‘left’ molecule with \(m_0 = -0.1d_0\), and the ‘right’ one with \(m_0 = +0.1d_0\), the radiative decay rate takes significantly different values; thus, under such conditions it is possible to control effectively the radiation properties of enantiomers. It is important to note that both the results of the calculations obtained within the framework of the QED theory (this paper) and in the framework of simplified quasistatic expressions presented in the beginning of this section (see also [49]) yield almost identical results, confirming the correctness of both approaches.
Figure 5. The radiative decay rate of the spontaneous emission of a chiral molecule located in the close vicinity of the surface of a chiral DNG spherical nanoparticle ($r_0 \rightarrow a$) with $\varepsilon = \varepsilon' + i 0.1$, $\mu = -1.6$, $\chi = 0.2$, and $\kappa_{0a} = 0.1$. (a) The electric and magnetic dipole moments of the transition of the molecule are oriented along the $x$-axis (tangentially to the particle surface). (b) The electric and magnetic dipole moments of the transition of the molecule are oriented along the $z$-axis (normally to the particle surface). The dashed line shows the asymptotic expression (26). The nanoparticle is placed in a vacuum.

Figure 6 shows the ratio of the effective radiative decay rate of the spontaneous emission of the ‘left’ molecule and the radiative decay rate of the ‘right’ molecule and vice versa. From this figure, it follows that if condition (35) is satisfied, then the decay rates of the ‘left’ and ‘right’ molecules differ by a factor of 25 or 30 or even more, depending on the chirality of the molecule considered as the reference one. In other words, nanoparticles with the parameter given by (35) will enhance the radiation of the ‘right’ molecules and slow down the radiation of the ‘left’ molecules and vice versa. Let us stress that DNG values of $\varepsilon$ and $\mu$ (DNG metamaterial) of a chiral sphere or its negative $\mu$ (MNG metamaterial) are of crucial importance for such a discrimination. Possible applications of the effect of discrimination of the radiation can be found in [47].

5. Conclusion

Thus, in this work the analytical expressions for the radiative decay rate of the spontaneous emission of a chiral molecule located near an arbitrary spherical particle with chiral properties are obtained and investigated within the QED theory. Using this approach, the spontaneous emission decay rates of chiral molecules placed near a spherical particle made of different materials (dielectrics, metals, DNG metamaterials, etc) are investigated in detail. It is shown that nonzero chirality of a spherical particle leads to an increase in the number of excited modes in comparison with a nonchiral one due to the coupling between the left- and right-polarized waves. Quasistatic expressions for the radiative decay rate of the spontaneous emission of a chiral molecule located near a chiral nanosphere are obtained and investigated. These expressions are in good agreement with the exact QED results and allow one to estimate the
Figure 6. (a) The ratio of the effective radiative decay rate of the spontaneous emission of ‘left’ molecules with $m_0 = -0.01d_0$ to the effective radiative decay rate of the spontaneous emission of ‘right’ molecules with $m_0 = +0.01d_0$ and (b) vice versa as a function of the dielectric permittivity ($\varepsilon = \varepsilon' + i0.01$) and the magnetic permeability ($\mu = \mu' + i0.01$) of the material from which the chiral spherical nanoparticle is made. The chirality parameter $\chi = 0.2$. The molecule is placed in the close vicinity of the surface of the spherical nanoparticle ($r_0 \to a$). The nanoparticle is placed in a vacuum.

parameters of nanoparticles when the radiation of ‘right’ or ‘left’ molecules is suppressed. It is found that to suppress the radiation of the ‘left’ molecules, the sphere should be made of MNG metamaterials, while to suppress the radiation of the ‘right’ molecules, one should use DNG materials.

The results obtained can be used to calculate the radiative decay rate of the spontaneous emission of chiral molecules in the vicinity of chiral spherical particles, to interpret the experimental data on the interaction of chiral molecules and particles, as well as for the detection and separation of enantiomers of chiral molecules with the help of chiral particles.

Acknowledgments

The authors thank the Belarusian Republican Foundation for Fundamental Research (DG, grant number F12R-006) and the Russian Foundation for Basic Research (VK, grant numbers 11-02-91065, 11-02-92002, 11-02-01272, and 12-02-90014) for partial financial support of this work.

Appendix A. Spherical vector harmonics

Spherical vector harmonics that describe the electromagnetic field inside a chiral spherical particle have the form

$$M\psi^{(j)}_{mn} = \text{rot} (r\psi^{(j)}_{mn}), \quad N\psi^{(j)}_{mn} = \frac{1}{k_f} \text{rot} M\psi^{(j)}_{mn},$$  \hspace{1cm} (A.1)
where $\mathbf{r}$ is the radius vector and $\Psi_{mn}^{(j)}$ is one of the scalar spherical harmonics

$$
\Psi_{mn}^{(j)} = \psi_n (k_J r) P_n^m (\cos \theta) e^{im\varphi},
$$

(A.2)

where $0 \leq r < \infty$, $0 \leq \theta < \pi$ and $0 \leq \varphi < 2\pi$ are spherical coordinates; $\psi_n (k_J r) = (\pi k_J r/2)^{1/2} J_{n+1/2} (k_J r)$, the Riccati–Bessel function [54], where $J_{n+1/2} (k_J r)$ is the Bessel function [54]; $P_n^m (\cos \theta)$, the associated Legendre function [54]; $k_J$, the wavenumber in the chiral medium and the index $J = L, R$.

Spherical vector harmonics that describe the electromagnetic field outside a chiral spherical particle have the following form:

$$
M_{mn}^{(j)} = \text{rot} \left( \mathbf{r} Z_{mn}^{(j)} \right), \quad N_{mn}^{(j)} = \frac{1}{k_0} \text{rot} \, M_{mn}^{(j)},
$$

(A.3)

where the scalar spherical harmonic $Z_{mn}^{(j)}$ is

$$
Z_{mn}^{(j)} = e_n^{(j)} (k_0 r) P_n^m (\cos \theta) e^{im\varphi},
$$

(A.4)

where $e_n^{(j)} (k_0 r) = (\pi k_0 r/2)^{1/2} H_{n+1/2}^{(j)} (k_0 r)$ is the Riccati–Bessel function [54], where $H_{n+1/2}^{(j)} (k_0 r)$ is the Hankel function of the first $(j = 1)$ and second $(j = 2)$ kind, respectively, $k_0$ is the wavenumber in a vacuum.

More information about the properties of spherical vector harmonics can be found, for example, in [55].

**Appendix B. Coefficients $\alpha_n$, $\beta_n$ and $\gamma_n$**

Explicit expressions for the coefficients in (7) have the following form:

$$
\alpha_n = 1 - 2 \left( \frac{A_n (L) V_n (R) + A_n (R) V_n (L)}{W_n (L) V_n (R) + W_n (R) V_n (L)} \right),
$$

$$
\beta_n = 2 \left( \frac{W_n (L) A_n (R) - W_n (R) A_n (L)}{W_n (L) V_n (R) + W_n (R) V_n (L)} \right),
$$

$$
\gamma_n = 1 - 2 \left( \frac{W_n (L) B_n (R) + W_n (R) B_n (L)}{W_n (L) V_n (R) + W_n (R) V_n (L)} \right).
$$

(B.1)

In (B.1) we have used the abbreviations $(J = L, R)$:

$$
W_n (J) = \frac{\sqrt{E \mu \lambda}}{\mu} \psi_n (k_J a) \xi_n^{(1)'} (k_0 a) - \psi_n' (k_J a) \xi_n^{(1)} (k_0 a),
$$

$$
V_n (J) = \psi_n (k_J a) \xi_n^{(1)'} (k_0 a) - \frac{\sqrt{E \mu \lambda}}{\mu} \psi_n' (k_J a) \xi_n^{(1)} (k_0 a),
$$

$$
A_n (J) = \frac{\sqrt{E \mu \lambda}}{\mu} \psi_n (k_J a) \psi_n' (k_0 a) - \psi_n' (k_J a) \psi_n (k_0 a),
$$

$$
B_n (J) = \psi_n (k_J a) \psi_n' (k_0 a) - \frac{\sqrt{E \mu \lambda}}{\mu} \psi_n' (k_J a) \psi_n (k_0 a),
$$

(B.2)

where the functions $\psi_n (k_J a)$ and $\xi_n^{(1)} (k_0 a)$ are defined in appendix A, the prime to a function stands for the derivative with respect to its argument.
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