Negative scattering asymmetry parameter for dipolar particles: Unusual reduction of the transport mean free path and radiation pressure

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Lossless dielectric nanospheres (made of nonmagnetic materials) with relatively low refraction index may present strong electric and magnetic dipolar resonances. We establish a relationship between the optical force from a plane wave on small electric and magnetic dipolar particles, the transport cross section, and the scattering asymmetry parameter $g$. In this way we predict negative $g$ (that minimize the transport mean free path below values of the scattering mean free path) for a dilute suspension of both perfectly reflecting spheres as well as of lossless dielectric nanospheres made of moderate permittivity materials, e.g., silicon or germanium nanospheres in the infrared region. Lossless dielectric Mie spheres of relatively low refraction index (as low as 2.2) are shown to present negative $g$ in specific spectral ranges.

The unusual observation of negative-$g$ factors has been limited to systems with appropriate short-range correlation between scatters. While it is frequently argued that the scattering from Mie spherical particles leads to $g > 1$, in this work we show that nonabsorbing Mie spheres of relatively low refraction index $m$ present negative-$g$ factors in specific spectral ranges. Previous numerical work [20] reported calculated negative-$g$ factors for dielectric spheres having refractive index $m$ larger than 3.1. However, the physical origin of these results was not discussed. Interestingly, as we will see, they represent a specific example of small particles, whose scattering may be completely described by a dipolar response to both the electric and magnetic fields. In contrast, we show that relatively large, nondipolar, dielectric spheres may lead to $g < 0$ for $m$ as low as 2.2.

As we will see, there is a close relationship between transport parameters of a dilute suspension of dipolar particles and the theory of optical forces on magnetodielectric small particles [10,11,21], in which it has been shown that, in addition to the force due to the electric and magnetic induced dipoles, there is an additional component due to the interaction between both of them, which was associated with the angular distribution of scattered intensity [10–12]. Maxima and (negative) minima of the $g$ factor are obtained for

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the so-called Kerker conditions [11,22,23] of zero-backward or almost zero-forward differential scattering cross sections (DSCSs). These conditions can be satisfied by hypothetical magnetic particles [8,22,23] as well as by small dielectric particles of high refractive index (e.g., of Si or Ge), which have recently been shown to behave as magnetodielectric [9,12], i.e., whose scattering is effectively dipolar, being well characterized by the Mie coefficients \( a_1 \) and \( b_1 \), both being of comparable strength. A dilute suspension of such particles near the almost-zero-forward scattering condition, will minimize the transport mean free path below the scattering mean free path.

The asymmetry factor \( g = \langle \cos \theta \rangle \) is defined [6,7] as the average of the cosine of the scattering angle \( \theta \) over the particle differential scattering-cross-section distribution \( d\sigma_s/d\Omega \):

\[
g = \langle \cos \theta \rangle = \frac{\int \frac{d\sigma_s}{d\Omega} \cos \theta \, d\Omega}{\int \frac{d\sigma_s}{d\Omega} \, d\Omega} = \frac{\int \frac{d\sigma_s}{d\Omega} \cos \theta \, d\Omega}{\sigma_s},
\]

where \( \sigma_s \) is the scattering cross section. Let us compute the \( g \) factor for a dielectric dipolar sphere of radius \( a \) and real refractive index \( m_p \) immersed in an arbitrary lossless medium with relative dielectric permittivity \( \epsilon \) and magnetic permeability \( \mu \). For spherical particles, the \( g \) factor does not depend on the polarization of the incident light and can be expressed in terms of the Mie coefficients \( a_0 \) and \( b_0 \) (see Sec. 4.5 in Ref. [7]). In Fig. 1(a) we show the \( g \) factor map for a nonabsorbing Mie sphere as a function of the relative refractive index \( m = m_p/\sqrt{\epsilon \mu} \) and the size parameter \( y = m(2\pi a/\lambda) \) calculated from the full Mie expansion. Since usually nonabsorbing materials present low refractive index \((m \lesssim 1.5)\) in the infrared and visible frequency ranges, negative-\( g \) factors in Mie particles were not expected. However, as it can be seen in Fig. 1(a), the \( g \) map shows regions of negative \( g \) for relatively low refraction index \((m \gtrsim 2)\) relevant for semiconductor particles made of silicon \((m \approx 3.5)\) or germanium \((m \approx 4)\) in the infrared and telecom wavelengths. This is one of the main results of the present work. The corresponding scattering-cross-section map for the same spheres (as calculated in Ref. [9]) is plotted in Fig. 1(b). For \( m \) values larger than approximately 2, the region between the magnetic [green in Fig. 1(b)] and electric [red in Fig. 1(b)] dipolar resonances presents a well-defined region of negative asymmetry parameter while the scattering is perfectly described by the first two dipolar terms in the Mie expansion [9].

The asymmetry factor \( g \) and the seemingly unrelated problems of transport mean free path and optical forces are now tied together by the close relation between power and momentum transfer, i.e., by the definition of transport \( \sigma^* \) and radiation pressure \( \sigma^{(y)} \) cross sections [6,7,24]. The transport cross section \( \sigma^* \) of a particle is expressed in terms of \( \sigma_s \) as [1,3]

\[
\sigma^* = \int \frac{d\sigma_s}{d\Omega} (1 - \cos \theta) \, d\Omega = \sigma_s (1 - g).
\]

For a dilute suspension of optically uncorrelated particles with density \( \rho \), the transport MFP \( \ell^* = 1/\rho \sigma^* \) is related to the scattering MFP \( \ell_s = 1/\rho \sigma_s \) through the aforementioned relationship \( \ell^* = \ell_s (1 - g) \). In contrast, the radiation pressure cross section is customarily defined as [6] \( \sigma^{(y)} = \sigma^{(ext)} - \langle \cos \theta \rangle \sigma_s = \sigma_a + \sigma^* \), where \( \sigma^{(ext)} = \sigma_e + \sigma_m \), with \( \sigma_a \) being the absorption cross section. In the absence of absorption, there is no difference between transport and radiation pressure cross sections,

\[
\sigma^* = \sigma^{(y)} = \sigma_s (1 - g).
\]

Hence there is a direct relation between transport quantities and the forces from an incident plane wave on a dielectric sphere.

In order to get deeper physical insight into the influence of the electric-magnetic dipole force in \( \ell^* \), it is interesting to derive the explicit expressions connecting both transport and radiation pressure with the \( g \) factor for the simplest and most important case of dipolar particles. Let us consider such a particle whose dipolar electric \( p \) and magnetic \( m \) moments are related to the external polarizing fields through \( p = \epsilon_0 \alpha_e E \) and \( m = (\alpha_m/\mu_0 k^3) B \). The dynamic polarizabilities \( \alpha_e \) and \( \alpha_m \) that characterize the dipole excitation can be expressed in terms of the Mie coefficients \( a_1 \) and \( b_1 \) as [6,7] \( \alpha_e = ia_1(6\pi/k^3) \) and \( \alpha_m = ib_1(6\pi/k^3) \).
The differential scattering cross section averaged over the incident polarizations is [10–12]

\[
\frac{d\sigma_s(\theta)}{d\Omega} = \frac{k^4}{16\pi} \left( |\alpha_e|^2 + |\alpha_m|^2 \right) \left( 1 + \cos^2 \theta \right) + 2\text{Re}(\alpha_e\alpha_m^*) \cos \theta.
\]

From Eqs. (4) and (1)

\[
g = \frac{\text{Re}(\alpha_e\alpha_m^*)}{|\alpha_e|^2 + |\alpha_m|^2},
\]

which shows that for a dipolar particle, \(|g| \leq 1/2\). Notice also that from Eqs. (4) and (5) this asymmetry factor may be expressed as

\[
g = \frac{1}{2} \left[ \frac{d\sigma_s^{(0)}}{d\Omega}(0^\circ) - \frac{d\sigma_s^{(0)}}{d\Omega}(180^\circ) \right].
\]

In the absence of absorption, the time-averaged force exerted on the dipolar particle by a time harmonic incident plane wave \(E = E_0 e^{i(k\cdot r)}\) is all radiation pressure [10,11] and reads

\[
(F) = (F_e) + (F_m) + (F_{e,m})
\]

\[
= \frac{\epsilon_0\epsilon}{2}|E_0|^2 \left\{ k \text{Im}(\alpha_e + \alpha_m) - \frac{k^4}{6\pi} \text{Re}(\alpha_e\alpha_m^*) \right\}
\]

\[
= \frac{\epsilon_0\epsilon}{2}|E_0|^2 \sigma^{(e)} \frac{k}{k}.
\]

The last term in Eq. (7), \((F_{e,m})\), due to the interaction between electric and magnetic dipoles [10,11,21], was associated in Ref. [11] with the asymmetry in the scattered intensity distribution [cf. the last term in Eq. (4)] even though it was not explicitly related to \(g\). We next show that they are proportional. Notice that the moduli of the first two terms \((F_e)\) and \((F_m)\), corresponding to the forces on the induced pure electric and magnetic dipoles, can be written as

\[
\langle F_e \rangle + \langle F_m \rangle = \frac{\epsilon_0\epsilon}{2}|E_0|^2 \sigma^{(e)} = \frac{\epsilon_0\epsilon}{2}|E_0|^2 \sigma_s,
\]

where the last equality holds for nonabsorbing particles, while the interference term

\[
\langle F_{e,m} \rangle = -\frac{\epsilon_0\epsilon}{2}|E_0|^2 \sigma_s g.
\]

We then have a formal result for the total force

\[
(F) = (\langle F_e \rangle + \langle F_m \rangle)(1 - g),
\]

which is the force analog of Eq. (3). We can summarize the above discussion in a single expression

\[
1 - g = \frac{\sigma^*}{\sigma_s} = \frac{\langle F \rangle}{\langle F_e \rangle + \langle F_m \rangle} = \frac{\epsilon_s}{\epsilon^*}.
\]

Equation (12) is another main result of this work. When the particle is nonabsorbing, \(1 - g\) becomes just the ratio between the magnitudes of the total force and the sum of the pure electric dipole forces. This quantities in a specific way the nature of the interaction force component in terms of the forward-backward asymmetry of the angular distribution of scattered intensity by the particle. It also establishes the connection between these forces and the transport and scattering MFP’s.

If no restrictions are imposed on \(\alpha_e\) and \(\alpha_m\), and hence one may consider them in Eq. (5) as independent variables, it is straightforward to see from this equation that \(g\) takes on extreme values when either \(\alpha_e = \alpha_m\) (with \(g\) being a maximum \(g = 1/2\)) or \(\alpha_e = -\alpha_m\) (with \(g\) being a minimum \(g = -1/2\)). The first condition corresponds to the so-called first Kerker condition and has been discussed in the context of scattering from a special case of magnetodielectric particles [11,22]. These particles lead to a zero-backward differential scattering cross section and have \(g = 1/2\), i.e., \(l^* = 2l_s\) (notice that forward scattering would correspond to \(g = 1\) and \(l^* = \infty\)). The second condition \((\alpha_e = -\alpha_m)\), which minimizes the scattered intensity in the forward direction, can only be fulfilled approximately since the imaginary part of the polarizabilities

![Figure 2](http://doc.rero.ch)

FIG. 2. (Color online) (a) Forward and backward differential scattering cross sections and asymmetry factor versus the wavelength for a silicon spherical particle of radius \(a = 230\) nm and \(\epsilon = 12\). (b) Different contributions to the total radiation pressure versus the wavelength for the same particle. Normalization is done by \(F_0 = 4\pi a/k|E_0|^2/2\). The vertical lines mark, from right to left, the first and second Kerker conditions.
must always be positive, as required from causality [11]. In the quasistatic approximation, they produce zero-forward scattered power [22]) with an asymmetry factor \( g \approx -1/2 \), which means that \( \ell^* \approx (2/3)\ell_s \). [note that strong backscattering would correspond to \( g = -1 \) and \( \ell^* = (1/2)\ell_s \)]. Thus, for a dilute suspension of arbitrary nonabsorbing dipolar particles, Eq. (5) and the Kerker conditions impose the following limits to the MFP:

\[
0.66\ell_s \lesssim \ell^* \lesssim 2\ell_s. \tag{13}
\]

While the results above are of general application, it is interesting to discuss two specific examples that could be realized experimentally. As a direct consequence of the discussion above, perfectly conducting dipolar spheres can be realized experimentally. As a direct consequence of the discussion above, perfectly conducting dipolar spheres can be realized experimentally. As a direct consequence of the discussion above, perfectly conducting dipolar spheres can be realized experimentally. As a direct consequence of the discussion above, perfectly conducting dipolar spheres can be realized experimentally. As a direct consequence of the discussion above, perfectly conducting dipolar spheres can be realized experimentally. As a direct consequence of the discussion above, perfectly conducting dipolar spheres can be realized experimentally. As a direct consequence of the discussion above, perfectly conducting dipolar spheres can be realized experimentally. As a direct consequence of the discussion above, perfectly conducting dipolar spheres can be realized experimentally. As a direct consequence of the discussion above, perfectly conducting dipolar spheres can be realized experimentally. As a direct consequence of the discussion above, perfectly conducting dipolar spheres can be realized experimentally. As a direct consequence of the discussion above, perfectly conducting dipolar spheres can be realized experimentally. As a direct consequence of the discussion above, perfectly conducting dipolar spheres can be realized experimentally. As a direct consequence of the discussion above, perfectly conducting dipolar spheres can be realized experimentally. As a direct consequence of the discussion above, perfectly conducting dipolar spheres can be realized experimentally.

Silicon spheres with radius \( a = 230 \) nm have been proven to behave as dipolar magnetodielectric particles with a strong magnetic dipole response in the near infrared region. In Fig. 2 we show the forward and backward differential scattering cross sections and the asymmetry factor \( g \) as well as the variation of \( \langle F_{\text{e},m} \rangle \), \( \langle F_e \rangle \), and \( \langle F_m \rangle \) for a Si sphere of radius 230 nm. Notice when the first Kerker condition is fulfilled, \( g = 1/2 \) and \( \frac{d\sigma_{\text{in}}}{d\Omega_{\text{in}}} \left(180^\circ\right) = 0 \) [Fig. 2(a)] and \( F_e = F_m = -F_{e-m} = F \) [Fig. 2(b)]. Nevertheless, if one imposes restrictions on \( \alpha_e \) and \( \alpha_m \), then other situations appear. From Eq. (6) one can see that \( g \) is maximal and equal to 1/2 where \( \frac{d\sigma_{\text{in}}}{d\Omega_{\text{in}}} \left(180^\circ\right) \) is zero and it has a minimum value at a wavelength where \( \frac{d\sigma_{\text{in}}}{d\Omega_{\text{in}}} \left(0^\circ\right) \) is minimal. This minimum value of \( g \) is also negative if \( \frac{d\sigma_{\text{in}}}{d\Omega_{\text{in}}} \left(0^\circ\right) < \frac{d\sigma_{\text{in}}}{d\Omega_{\text{in}}} \left(180^\circ\right) \). This is illustrated in the case of the above-mentioned Si sphere of radius 230 nm. As seen in Fig. 2(a), at \( \lambda = 1530 \) nm \( g \) has a minimum equal to \(-0.15\), which corresponds to the minimum forward DSCS, whereas where the first Kerker condition holds, when the backscattering cross section is zero, the \( g \) factor has a maximum equal to 1/2.

In conclusion, we have demonstrated that, surprisingly and without further assumptions about collective interactions, dilute suspensions of dipolar semiconductor spheres, e.g., Si and Ge, have an optical frequency range in which their scattering asymmetry parameter is negative and hence they acquire a transport MFP smaller than their scattering MFP. This is made possible by the magnetodielectric nature of these particles and the consequent electric-magnetic dipole interference, which in addition leads to a simple relation between the electric-magnetic interaction photonic force and the asymmetry factor. This also applies to perfectly conducting spheres at longer wavelengths.

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