Total absorption of light by a nanoparticle: an electromagnetic sink in the optical regime

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In this Letter, we give a general description of the illumination and object properties for obtaining total absorption. We show theoretically and numerically that properly designed sub-100 nm metallic particles are able to absorb all the energy of an incident beam if the latter is adequately shaped. In addition to their interest as absorbers, these particles act as efficient near-field probes as they convert the incident propagating beam into a localized nonradiative field.

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The design of optical components allowing total absorption of the incident light is a challenging issue that has attracted much attention in the last 20 years. Most of the studies on this subject, triggered in particular by the photovoltaic applications, consider planar components, generally periodic or random structured surfaces [1], illuminated by collimated incident beams. In this framework, a significant increase of the absorption level has been observed as a consequence of resonances in metallic or metallodielectric gratings [2–5]. In parallel, many studies have been carried on the radiative properties of nanoparticles [6,7], or even molecules [8], essentially for forming efficient scattering or absorbing markers. A strong increase of the extinction coefficient has been observed in the vicinity of the particle resonances [9]. In this Letter, we combine both approaches and aim at finding a configuration in which the incident light is totally absorbed by a nanoparticle (i.e., on the smallest possible volume). This issue is strongly related to the total light absorption by an atom [10], achieved by time-reversing spontaneous emission [11], although it remains in a classical electromagnetism framework.

We first derive the very general condition under which total absorption of a monochromatic illumination [with omitted time-dependence $\exp(-i\omega t)$] can occur. This problem has already been addressed in a promising way for scalar waves in one- or two-dimensional geometries, using the S-matrix formalism [12]. Here, we present an alternative approach, adapted to vectorial three-dimensional configurations, that better suits our application.

We consider a reference inhomogeneous medium, included in a ball W, that is depicted by its, possibly complex, relative permittivity $\varepsilon_{\rm ref}(\mathbf{r})$; see Fig. 1. Outside W, the relative permittivity is equal to one. A lossy object Ω is introduced in W so that the relative permittivity of the system reads $\varepsilon_{\rm ref}(\mathbf{r})+\chi(\mathbf{r})$ where $\chi(\mathbf{r})$ is equal to zero outside Ω. Without loss of generality, $\varepsilon_{\rm ref}$ is taken lossless and constant for all $\mathbf{r}\in\Omega$. W is illuminated by a monochromatic electromagnetic field that is created by sources that are placed beyond W. From Maxwell equations, the total electric field that exists in W is solution of

$$\nabla\times\nabla\times\mathbf{E}(\mathbf{r})-\varepsilon_{\rm ref}(\mathbf{r})k_0^2\mathbf{E}(\mathbf{r})=k_0^2\chi(\mathbf{r})\mathbf{E}(\mathbf{r}), \tag{1}$$

where $k_0=\omega/c$. The total field $\mathbf{E}(\mathbf{r})$ can be written as the sum of a reference field $\mathbf{E}_{\rm ref}(\mathbf{r})$, which verifies the homogeneous equation

$$\nabla\times\nabla\times\mathbf{E}_{\rm ref}(\mathbf{r})-\varepsilon_{\rm ref}(\mathbf{r})k_0^2\mathbf{E}_{\rm ref}(\mathbf{r})=0, \tag{2}$$

and a diffracted field $\mathbf{E}_d(\mathbf{r})=\mathbf{E}(\mathbf{r})-\mathbf{E}_{\rm ref}(\mathbf{r})$, which satisfies the outgoing wave boundary conditions beyond W. Note that contrary to $\mathbf{E}_d(\mathbf{r})$, the reference field $\mathbf{E}_{\rm ref}(\mathbf{r})$, which represents the field that would exist in absence of the object, does not satisfy the outgoing nor ingoing wave boundary conditions. To calculate $\mathbf{E}(\mathbf{r})$, we introduce the Green tensor of the reference medium $\overleftrightarrow{\mathbf{G}}$, which is the solution of

$$\nabla\times\nabla\times\overleftrightarrow{\mathbf{G}}(\mathbf{r},\mathbf{r}')-\varepsilon_{\rm ref}(\mathbf{r})k_0^2\overleftrightarrow{\mathbf{G}}(\mathbf{r},\mathbf{r}')=k_0^2\overleftrightarrow{\mathbf{I}}\delta(\mathbf{r}-\mathbf{r}'), \tag{3}$$

where $\overleftrightarrow{\mathbf{I}}$ denotes the unit tensor that satisfies the outgoing wave boundary conditions. Subtracting Eq. (2) from Eq. (1) to get the equation verified by the diffracted

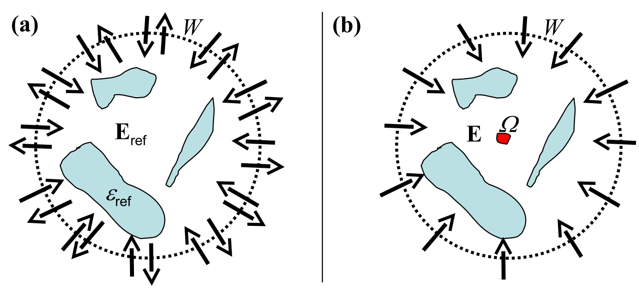

Fig. 1. (Color online) Illustration of the total absorption condition. (a) A reference medium is placed in a ball W is illuminated by sources placed outside W. The reference field $\mathbf{E}_{\rm ref}$ that is created in W satisfies neither the outgoing nor ingoing wave boundary conditions. (b) Lossy object Ω is introduced in the reference lossless medium. Under the total absorption condition, the field $\mathbf{E}$ that is created in W satisfies the ingoing wave boundary conditions.

field and using Eq. (3), one obtains the classical expression for the diffracted field [13]:

$$\mathbf{E}_d(\mathbf{r}) = \int_\Omega \overleftrightarrow{\mathbf{G}}(\mathbf{r},\mathbf{r}')\chi(\mathbf{r}')\mathbf{E}(\mathbf{r}')\mathrm{d}\mathbf{r}'. \quad (4)$$

If the lossy object Ω is designed so that total absorption occurs, then the total field $\mathbf{E}(\mathbf{r})$ must satisfy the ingoing wave boundary conditions. This condition implies that the outgoing part of $\mathbf{E}_{\mathrm{ref}}(\mathbf{r})$ cancels the diffracted field. The field solution of Eq. (1) that satisfies the ingoing wave boundary conditions is

$$\mathbf{E}(\mathbf{r}) = \int_\Omega \overleftrightarrow{\mathbf{G}}_*^*(\mathbf{r},\mathbf{r}')\chi(\mathbf{r}')\mathbf{E}(\mathbf{r}')\mathrm{d}\mathbf{r}', \quad (5)$$

where A^* stands for the complex conjugate of A and $\overleftrightarrow{\mathbf{G}}_*$ denotes the Green tensor that is the solution that satisfies the outgoing wave boundary conditions of Eq. (3) in which $\varepsilon_{\mathrm{ref}}$ has been replaced by $\varepsilon^*_{\mathrm{ref}}$. Thus $\overleftrightarrow{\mathbf{G}}_*^*$ is the solution of Eq. (3) that satisfies the ingoing boundary conditions. Generally, Eq. (5) does not have any nonnull solution. It is only with specific object potential and wavelengths that one can obtain a nonnull field satisfying Eq. (5). It is worth noting, at this point, that Eq. (5) needs to be satisfied only for $\mathbf{r} \in \Omega$ and that taking the phase conjugate of Eq. (5) gives the classical laser mode equation for the same geometry W in which ε_{ref} and χ are replaced by ε^*_{ref} and χ^*, respectively. Ω is replaced by a gain object Ω' with potential $\chi'(\mathbf{r}) = \chi^*(\mathbf{r})$, as pointed out in [12,14]. Once $\mathbf{E}(\mathbf{r})$ is known, the illumination configuration of W is perfectly defined. Indeed, the reference field can be deduced from Eqs. (4) and (5) as

$$\mathbf{E}_{\mathrm{ref}}(\mathbf{r}) = \int_\Omega \left[\overleftrightarrow{\mathbf{G}}_*^*(\mathbf{r},\mathbf{r}') - \overleftrightarrow{\mathbf{G}}(\mathbf{r},\mathbf{r}')\right]\chi(\mathbf{r}')\mathbf{E}(\mathbf{r}')\mathrm{d}\mathbf{r}'. \quad (6)$$

One verifies easily that $\mathbf{E}_{\mathrm{ref}}(\mathbf{r})$ is a solution of the homogeneous Eq. (2). When $\varepsilon_{\mathrm{ref}}$ is lossless, the reference field corresponds to the field existing in W, in absence of Ω, when the latter is illuminated by the time-reversed (or phase-conjugated) field radiated by the induced polarization density $\chi(\mathbf{r}')\mathbf{E}(\mathbf{r}')$ inside the object [15].

We now apply this analysis to the specific problem of the electromagnetic sink. We search the smallest possible lossy particle Ω with uniform relative permittivity ε that is able to absorb all the incident light (if the latter is adequately shaped) in a given reference medium. The particle, centered about $\mathbf{r}_0$, is assumed to be small enough compared to the wavelength for the electric field $\mathbf{E}$ to be constant and equal to $\mathbf{E}(\mathbf{r}_0)$ over the volume v of Ω. To calculate Eq. (5) at $\mathbf{r} = \mathbf{r}_0$, the Green tensor $\overleftrightarrow{\mathbf{G}}_*(\mathbf{r}_0,\mathbf{r})$ is split into a Dirac distribution, $-\overleftrightarrow{\mathbf{L}}\delta(\mathbf{r}_0 - \mathbf{r})$, and a principal value distribution [16,17], yielding

$$\int_\Omega \overleftrightarrow{\mathbf{G}}_*(\mathbf{r}_0,\mathbf{r}')\mathrm{d}\mathbf{r}' = -\overleftrightarrow{\mathbf{L}} + v\overleftrightarrow{\mathbf{G}}_*^{\mathrm{reg}}(\mathbf{r}_0,\mathbf{r}_0) \quad (7)$$

with $v\overleftrightarrow{\mathbf{G}}_*^{\mathrm{reg}}(\mathbf{r}_0,\mathbf{r}_0) = \mathrm{PV}\left[\int_\Omega \overleftrightarrow{\mathbf{G}}_*(\mathbf{r}_0,\mathbf{r}')\mathrm{d}\mathbf{r}'\right]$ [18]. Noting $\overleftrightarrow{\alpha}_0$ the quasi-static polarizability of Ω [19], $\overleftrightarrow{\alpha}_0 = v\chi(\mathbf{r}_0)\left\{\overleftrightarrow{\mathbf{I}} + \chi(\mathbf{r}_0)\overleftrightarrow{\mathbf{L}}\right\}^{-1}$, one can approximate Eq. (5) by

$$\overleftrightarrow{\alpha}_0^{-1}\mathbf{E}(\mathbf{r}_0) \approx \left[\overleftrightarrow{\mathbf{G}}_*^{\mathrm{reg}}\right]^*(\mathbf{r}_0,\mathbf{r}_0)\mathbf{E}(\mathbf{r}_0). \quad (8)$$

This expression gives the relationship between the particle polarizability and its environment for total absorption of light to occur. For a spherical nanoparticle with radius a in vacuum, it reads

$$\alpha_0^{-1} \approx \frac{2}{3v}[(1 - ik_0 a)e^{ik_0 a} - 1]^* = \frac{k_0^2}{4\pi a} - i\frac{k_0^3}{6\pi} + O(k_0 a), \quad (9)$$

where $\alpha_0 = 4\pi a^3(\varepsilon - 1)/(\varepsilon + 2)$. With permittivities taken from the Palik tables [20], this equality is almost reached for a silver nanosphere of radius $a = 35$ nm at $\lambda = 386$ nm and for a gold particle with radius $a = 63$ nm at $\lambda = 510$ nm. Both wavelengths are close to the spheres' plasmon resonances. To verify the validity of our approach we have simulated rigorously the field $\mathbf{E}_d$ scattered by a nanosphere centered about the origin in vacuum and illuminated by $\mathbf{E}_{\mathrm{ref}}(\mathbf{r}) = -2i\,\mathrm{Im}\left[\overleftrightarrow{\mathbf{G}}_0(\mathbf{r},0)\right]\mathbf{u}$, where $\mathbf{u}$ is a unit vector directed along the z axis and $\overleftrightarrow{\mathbf{G}}_0$ is the vacuum Green tensor. This incident field corresponds to the field that would be obtained in a 4-pi illumination configuration using two facing objectives or a planar or parabolic mirror [21–23]. We have calculated the Poynting vector flux of the total outgoing field $\mathbf{E}_d(\mathbf{r}) - \overleftrightarrow{\mathbf{G}}_0(\mathbf{r},0)\mathbf{u}$ through the sphere W surrounding the particle and compared it to the Poynting vector flux of the incoming field $\overleftrightarrow{\mathbf{G}}_0^*(\mathbf{r},\mathbf{0})\mathbf{u}$ through the same sphere. All the calculations were performed with the coupled dipole method [13,24]. The convergence of the results with respect to the sphere discretization was checked, using plane wave illumination, with a code based on the Mie theory. In Fig. 2, we plot in dB units the ratio between the outgoing energy flux and the incoming one for various wavelengths and radii of a silver [Fig. 2(a)] and a gold [Fig. 2(b)] nanosphere illuminated under 4-pi illumination. We observe that the radius and wavelength at which about 99% of the incident energy is absorbed by the particle are very close to that derived

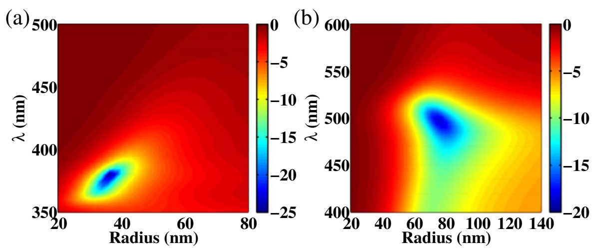

Fig. 2. (Color online) Metallic nanosphere in vacuum is illuminated under 4-pi configuration. Ratio in dB units of the outgoing energy flux over the incoming energy flux obtained as a function of the illumination wavelength and the sphere radius. Sphere made of (a) silver and (b) gold.

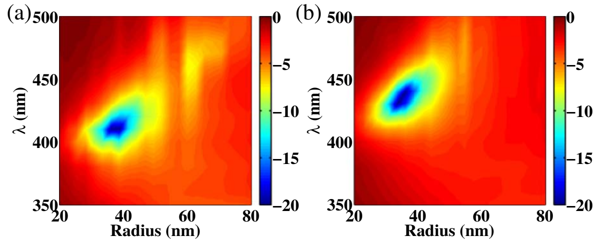

Fig. 3. (Color online) Same as Fig. 2 but the metallic sphere is coated with a thin dielectric layer of relative permittivity 2.25. The sphere core is made of silver. (a) The dielectric shell is 10 nm thick. (b) The dielectric shell is 20 nm thick.

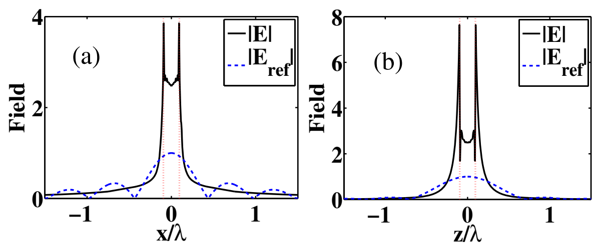

Fig. 4. (Color online) Modulus of the electric field in the vicinity of a silver nanosphere with radius $a = 36$ nm and $\lambda = 380$ nm under 4-pi illumination as a function of (a) x for $y = z = 0$, and (b) z for $x = y = 0$. Dashed curve, the field $\mathbf{E}_{\mathrm{ref}}$ existing without the sphere. Solid curve, the field $\mathbf{E}$ existing in presence of the sphere. The dotted curves indicate the position the nanosphere surface.

theoretically. Note that with the total absorption phenomenon being closely related to the plasmon resonance, it is possible to tune the absorption wavelength by coating the nanosphere with a thin layer of dielectric material as seen in Fig. 3. Apart from their interest as localized absorbers, these nanoparticles (combined with the appropriate illumination) could play an interesting role for imaging and sensing [25]. In Fig. 4 is plotted the magnitude of the reference field that exists in the absence of the particle (dashed curve) and the total field that exists in the presence of the particle (solid curve). We recall that the reference field is obtained by time reversing the field radiated by a dipole oriented along the z axis. It behaves roughly as $\sin(k_0 r)/r$ and generates a diffraction-limited light spot. On the contrary, once the particle is introduced, the total field is strongly localized, the field magnitude varying as $1/r^3$ in the near vicinity of the nanosphere. These simulations suggest that these particles could be used as efficient near-field probes.

In conclusion, we have derived a general condition on a lossy object and its illumination that ensures total absorption [Eq. (5)]. This condition imposes the values of the field inside the object and the values of the incident field. The latter is obtained by time reversing the field diffracted by the object while the former satisfies $\mathbf{E}(\mathbf{r} \in \Omega) = \int_\Omega \overleftrightarrow{\mathbf{G}}^*_*(\mathbf{r}, \mathbf{r}')\chi(\mathbf{r}')\mathbf{E}(\mathbf{r}')\mathrm{d}\mathbf{r}'$. For small metallic spheres in vacuum, the total absorption condition reduces to a simple requirement on the sphere polarizability coupled to an isotropic illumination [21,22]. We have shown that using this illumination scheme, a silver or gold nanosphere with radius smaller than 100 nm can absorb about 99% of the incident energy. We believe that this simple analysis could be used to design more complex absorbers (using particle arrays or aggregates).

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