Optimizing Nanoparticle Designs to Reach Ideal Light Absorption

Victor Grigoriev, Nicolas Bonod, Jerome Wenger, Brian Stout
Aix-Marseille Université, Institut Fresnel, Faculté des Sciences de Saint Jérôme, 13397 Marseille, France
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Ideal absorption describes a particular means of optimizing light-matter interactions with a host of potential applications. This work presents new analytic formulas and describes semi-analytical methods for the design of electric or magnetic ideal absorption in nanoparticles. These formulas indicate that ideal absorption is attainable in homogeneous spheres with known materials at specific sizes and frequencies. They also provide a means of designing core-shell particles to produce ideal absorption at virtually any frequency in the visible and near infrared range.

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I. INTRODUCTION

Subwavelength sized particles that feature electromagnetic resonances are ideal tools to concentrate light at the nanometer scale. They play an important role in a wide range applications including receiving elements in optical antennas, solar panel transducers and sensing. In this context, the design of nanoparticles that feature the ability to optimally convert light energy in at least one incident field mode into another form like photoluminescence or heat is of crucial importance. Such particles will be defined in this study as Ideal Absorption (IA) particles. Ideal absorption has also been called coherent perfect absorption since it results in a mode of the total field taking the form of a purely incoming being completely absorbed by the particle. In this work, we prefer to avoid this terminology since absorption is only ‘perfect’ for an ideal 4π illumination specifically adapted to the corresponding IA mode.

In this work, ideal absorption is first introduced by demonstrating that it corresponds to a fundamental upper bound in an absorption cross section channel, \( \sigma_a = 3\lambda^2/8\pi \) for a dipole resonator. A ‘point-like’ model is then shown to provide an analytical formula for predicting an electric dipole IA, but since this model is inaccurate for all but the smallest particles, significantly improved approximate formulas are derived. We proceed to show that the ensemble of IA solutions can be determined using the so called Weierstrass factorization of the analytical scattering response of spherical particles. Other considerations, like multi-mode absorption and IA bandwidth are discussed using illustrative calculations. We demonstrate that our formulas predict IA in homogeneous spheres with realizable materials at specific frequencies, and sizes. This work finishes with a derivation of a simple procedure for designing core-shell particles to exhibit IA at essentially any frequency in the visible and near visible frequency range.

II. SCATTERING THEORY FOR IDEAL ABSORPTION

Scattering in three dimensional electromagnetic problems can conveniently be expressed in terms of incoming and outgoing spherical Vector Partial Waves (VPWs). The VPWs are solutions of homogeneous media Maxwell equations of either electric source type \( e : \mathbf{N}^{(\pm)}_{n,m}(kr) \) or magnetic source type \( h : \mathbf{M}^{(\pm)}_{n,m}(kr) \), with the \((+/-)\) superscripts indicating that the functions satisfy outgoing (incoming) boundary conditions respectively. The subscript, \( n \), denotes the total angular momentum number, and \( m = -n, ..., n \), the angular momentum projection number.

The spatial dependence of the VPWs is scaled by the in-medium wavenumber, \( k = \sqrt{\varepsilon_\text{r} \mu_\text{r} \omega/c} = N_\text{b} \omega/c = 2\pi/\lambda \), where \( N_\text{b} \) is the refraction index of the homogeneous background material, and \( \lambda \), the in-medium wavelength.

Theoretical treatments of electromagnetic scattering from a homogeneous spherically symmetric particle go under the names of Lorenz-Mie-Debye theory, but the physical content is best viewed in the formalism of S or T matrices, originally developed for quantum mechanical scattering theory. The S-matrix seems best adapted to the discussion of IA, and is privileged from here on.

The total field in a homogeneous region surrounding a particle can always be developed on the basis set of the incoming and outgoing VPWs, with \( a_{n,m}^{(e,\pm)} \) and \( a_{n,m}^{(h,\pm)} \) respectively denoting the electric and magnetic mode VPWs field coefficients. Since S-matrix of a spherically symmetric system is automatically diagonal in the VPW basis, its elements express the linear relationship between the outgoing and incoming field coefficients of the total field for both electric and magnetic source fields respectively, \( S_{n}^{(e)} = a_{n,m}^{(e,+)} / a_{n,m}^{(e,-)} \), and \( S_{n}^{(h)} = a_{n,m}^{(h,+)} / a_{n,m}^{(h,-)} \).

Algebraic manipulations involving the T-matrix, and the definition of the S-matrix, \( \mathbf{S} = \mathbf{I} + 2\mathbf{F} \),
provide convenient expressions for the S-matrix coefficients:

\[
S^{(c)}_n = \frac{h^{(-)}_n(kR)}{h^{(+)}_n(kR)} \sum_{j} \varphi^{(-)}_n(k_j R) - \varphi^{(+)}_n(k_j R)
\]

\[
S^{(h)}_n = \frac{h^{(-)}_n(kR)}{h^{(+)}_n(kR)} \sum_{j} \pi_s \varphi^{(-)}_n(k_j R) - \varphi^{(+)}_n(k_j R)
\]

where, \(\pi_s \equiv \varepsilon_s/\mu_h\), and \(\pi_a \equiv \mu_s/\mu_h\) are respectively the permittivity and permeability contrasts between the sphere and the host medium, and \(k_s\) is the wavenumber inside the sphere. The \(h^{(+)}_n(x)\) and \(h^{(-)}_n(x)\) functions respectively denote the outgoing and incoming spherical Hankel functions. The expressions in Eq.(1) also employ the functions:

\[
\varphi^{(\pm)}_n(x) \equiv \left[ \frac{x h^{(\pm)}_n(x)}{(\pm)^{1/2} h^{(\pm)}_n(x)} \right]', \quad \varphi_n(x) \equiv \left[ \frac{x j_n(x)}{j_n(x)} \right]',
\]

where \(j_n(x)\) are the \(n^{th}\) order spherical Bessel functions.

Flux conservation imposes an upper bound on the amplitude of the S matrix elements, \(|S^{(c,h)}_n| \leq 1\), where the upper limit, \(|S^{(c,h)}_n| = 1\) is satisfied by any lossless scatterer since this condition results directly in \(|a_n^{(h)}| = |a_n^{(-)}|\). The S-matrix of a lossless scatterer is characterized by zeros (absorbing modes) in the upper-half complex frequency plane, and poles (emitting modes) in the lower-half plane frequency plane.[11] Adding absorption to the particle causes the absorbing modes to descend towards (and finally into) the lower complex plane with IA occurring at those values of \(\varepsilon_s\) for which a zero of the S-matrix lies on the real frequency axis, i.e. there exists an \(n\) such that:

\[
S^{(e)}_n = 0 \Longleftrightarrow \varphi_n(k_s R) / \varepsilon_s = \varphi^{(-)}_n(kR) , \quad \text{or},
\]

\[
S^{(h)}_n = 0 \Longleftrightarrow \varphi_n(k_s R) / \pi_s = \varphi^{(-)}_n(kR),
\]

when \(\Im\{\omega\} = 0\) (these conditions being found by inspection of Eq.(1a) and Eq.(1b).

Scattering and absorption cross sections of a spherically symmetric scatterer are the sum of the contributions from all multi-pole orders, \(n\):

\[
\sigma_s = \sum_{n=1}^{\infty} \left( \sigma^{(e)}_{n,s} + \sigma^{(h)}_{n,s} \right) , \quad \sigma_a = \sum_{n=1}^{\infty} \left( \sigma^{(e)}_{n,a} + \sigma^{(h)}_{n,a} \right).
\]

The multi-pole contributions to the extinction, scattering, and absorption cross sections[12][15][16] can be respectively expressed in terms of the S-matrix as:

\[
\sigma^{(e,h)}_{n,s} = \pi \frac{k^2}{2} (2n + 1) \left( 1 - \left| S^{(e,h)}_n \right|^2 \right)
\]

\[
\sigma^{(e,h)}_{n,a} = \pi \frac{k^2}{2} (2n + 1) \left( S^{(e,h)}_n - 1 \right)^2
\]

\[
\sigma^{(e)}_{n,a} = \pi \frac{k^2}{2} (2n + 1) \Re \left\{ 1 - S^{(e)}_n \right\}
\]

where the \(2n + 1\) factors arise from the azimuthal mode degeneracy of the orbital modes.

One finds that an IA mode contributes equally to the absorption and scattering cross sections by inserting the IA criteria, \(S_n = 0\) into Eq.(5a) and Eq.(5b) (in either the (e) or (h) channels), with values of:

\[
\sigma_{n,s} = \sigma_{n,a} = \frac{2n + 1}{8\pi} \lambda^2 ,
\]

which is an upper bound for the absorption cross section of a mode, but a factor four times less than the \(S_n = -1\), unitary limit[14] of the scattering or extinction cross sections, obtainable from Eq.(5b) and Eq.(5c). The equality of scattering and absorption cross sections in an IA channel follows from the fact that IA requires the field scattered by the particle to have perfectly destructive interference with the outgoing wave components of the local excitation field.
(leaving the $n$th order modes of the total field to be purely incoming). We also remark the many properties of IA in particles, like Eq.(6), find analogues in the ideal absorption of 1D systems, where for example the maximum absorption coefficient, $A$, of a plane wave illuminating a single side of a symmetric thin film occurs when $A = R + T = 0.5$. $^{15, 19}$

Since the $\varphi_n^{(-)}(kR)$ is a complex function of a real variable, and $\varphi_n(k_sR)$ is real for real valued $k_s$, the IA solutions of Eq.(3a) or Eq.(3b) can occur only for complex values of $\varepsilon_s$. Furthermore, Eqs.(3) are transcendental equations, and each has an infinite number of solutions. For the sake of clarity, further discussion in this work will be limited to the $n = 1$ (i.e. dipole) mode solutions of Eq.(3).

The real and imaginary parts of the solutions of Eq. (3a) with the lowest values of $\Re\{\varepsilon_s\}$ are plotted as the blue (solid) line in Fig.(1) for particle diameters ranging from zero up to a little more than a wavelength. These results were computed by taking advantage of the newly developed Weierstrass factorization for resonant photonic structures.$^{11}$ One readily remarks that this solution tends to the well-known quasi-static dipolar plasmon resonance at $\varepsilon_s = -2$ when $kR \to 0$. When this is the only IA mode of interest, one can try to describe IA with a ‘point-like’ model aimed at providing approximate descriptions of the lowest $\Re\{\varepsilon_s\}$ electric dipole resonance$^{20, 21}$.

FIG. 1: (Color on-line) The real and imaginary parts of $\varepsilon_s$ yielding the lowest IA solution are plotted as a function of $kR$ (blue (solid) curves in (a) and (b)). The point-like model prediction of Eq.(9), is plotted in green (long dashed), while the improved approximation of Eq.(13) is plotted in red (short dashed).

III. POINT-LIKE MODELS AND ANALYTIC EXTENSIONS

Point-like models have generally been derived from first principles using Green’s function theory, and the most rigorous derivations predict a point-like electric dipole polarizability, $\alpha$, that can be written:

$$\left[\alpha^{(e)}\right]^{-1} = \alpha_0^{-1} - \frac{k^2}{6\pi R} - i\frac{k^3}{6\pi}, \quad \alpha_0 = 4\pi R^3 \frac{\varepsilon_s - 1}{\varepsilon_s + 2},$$

where $\alpha_0$ is the quasi-static polarizability of a spherical particle. The $-ik^3/6\pi$ term in Eq.(7) has been extensively studied due to its importance in energy conservation, while the $-k^2/6\pi R$ term has received considerably less attention even though it is responsible for the well-known ‘red-shift’ of the localized plasmon resonance with increasing particle size.

For a spherically symmetric scatterer, one can directly connect the isotropic polarizability in terms of the electric dipole S-matrix element, $S_1^{(e)}$, via the relation:

$$\alpha^{(e)} = \frac{3\pi i}{k^3} \left(1 - S_1^{(e)}\right),$$

leading to an IA polarizability of $\alpha_{1A} = 3\pi i/k^3$. An advantage of the point-like model is that one can algebraically solve Eq.(7) for the value of $\varepsilon_s^{(e)}$ producing an IA polarizability as a function of the size parameter, $\rho \equiv kR$:

$$\varepsilon_s^{(e)}(\rho) = -\frac{2 + \frac{2}{3}\rho^2 (1 - i\rho)}{1 - \frac{2}{3}\rho^2 (1 - i\rho)}.$$
The real and imaginary parts of this expression for $\varepsilon_s$ are plotted in Fig.(1a) and Fig.(1b) (green curves), and although $\varepsilon_s$ tends towards the numerically obtained result (blue curve) in the $kR \to 0$ limit, it differs significantly from the exact result for larger particle sizes.

One can determine much more reliable predictions of the IA conditions by analyzing the pole structure of the special functions. One first remarks that the $\varphi_{n}^{(\pm)}$ functions appearing in Eq.(1) have finite meromorphic expressions, which for the dipole example take the form:

$$h_1^{(\pm)}(z) = -e^{\pm iz} \frac{z \pm i}{z^2}; \quad \varphi_{1}^{(\pm)}(z) = \pm iz - \frac{1}{1 \mp iz}. \quad (10)$$

The $\varphi_n$ functions on the other hand have an infinite number of poles located along the real axis, and can be expressed in meromorphic form as:

$$\varphi_n(z) = n + 1 + \sum_{\alpha=1}^\infty \frac{2\pi z^2}{z^2 - a_{n,\alpha}^2}, \quad (11)$$

where the constants, $a_{n,\alpha}$, are the zeros of the spherical Bessel function, $j_n(x)$, and are tabulated.[25]

One can obtain analytic dipole approximations to Fig.(1) by replacing $\varphi_1$ of Eq.(11) with an approximate meromorphic function having the same two lowest poles, $a$ and zeros, $b$:

$$\varphi_1(z) \approx 2 \frac{1 - (z/b)^2}{1 - (z/a)^2}, \quad (12)$$

where $a = 1.4303\pi$ and $b = 0.87335\pi$. Adopting this substitution transforms $S_1^{(e,h)} = 0$ into quadratic equations in terms of $\varepsilon_s^{(e,h)}$ whose relevant solutions are:

$$\varepsilon_s^{(e)}(\rho) = \frac{1}{2} \frac{b^2}{\rho^2} \left[ 1 + \frac{\rho^2}{a^2} \frac{2(\rho + 1)}{\rho^2 - \rho - 1} \right] \approx 0 \quad \text{and} \quad \varepsilon_s^{(h)}(\rho) = \frac{3 + 3i\rho - \rho^2}{\rho^2 \left( \frac{2+2i\rho}{a^2} + \frac{1+i\rho}{\alpha^2} \right)} \quad (13a)$$

The electric dipole solution of Eq.(13a) is plotted in red in Fig.(1), while the plot of Eq.(13b) is indistinguishable on the scale of the numerically exact solution plotted in blue in Fig.(2a) and Fig.(2b).

**IV. EXACT SOLUTIONS**

Although the analytic expressions of Eq.(13) provide accurate approximations for the lowest IA modes, one may nevertheless desire higher precision or the predictions of additional IA solutions, either of which will require numerical solutions of Eq.(3a) or Eq.(3b). Finding these solutions is quite difficult when using commonly employed techniques involving Cauchy integrals in the complex plane or conjugate gradient methods; but they can be readily solved using techniques based on Weierstrass factorization[11] and which exploit the meromorphic expansion of Eq.(11).

The real and imaginary parts of the numerically obtained dipole solutions of Eq.(3a) and Eq.(3b) for the $\varepsilon_s$ are plotted in Fig.(2) as a function of $kR$ for particles ranging in size from zero up to slightly larger than a wavelength ($kR = \pi$ corresponds to particle diameter=$\lambda$).

Even though the IA conditions can be found from the numerical solutions presented in Fig.(2), absorber design will generally require taking into account the other properties of the absorber, like IA bandwidth and absorption in other modes. We illustrate this point with some plots of the dimensionless absorption cross section, $8\pi \sigma_{abs}/(3\lambda^2)$, for particles designed to satisfy electric dipole IA at a few different sub-wavelength size parameters, notably $kR = 0.3$ and $kR = 1$ in Fig.(3a) and Fig.(3b). The total absorption cross sections are plotted in blue, the electric dipole absorption cross sections in red, and as a further piece of information, the magnetic dipole absorption cross section is plotted in green.

One remarks from Fig.(2) that IA can be satisfied for relatively modest values of $\Re{\varepsilon_s} > 1$ for size parameters roughly larger than unity (i.e. $D \gtrsim \lambda/3$). For IA designed to occur at $kR = 2$, absorption cross sections are
FIG. 2: (Color on-line) The real, (a), and imaginary parts, (b), of the electric dipole IA conditions on $\varepsilon_s$ are plotted as a function of $kR$. Magnetic dipole IA values for $\varepsilon_s$, are also plotted as functions of $kR$ in (c)-Re{$\bar{\varepsilon}_s$} and (d)-Im{$\bar{\varepsilon}_s$}. The lowest electric dipole solution with $\Re\{\varepsilon_s\} > 1$ is plotted as a dashed curve in (c) and (d) for the purpose of comparison.

...plotted for both the $\Re\{\varepsilon_s\} < 1$ solution value of $0.4809 + i1.148$ in Fig.(3c) and the lowest $\Re\{\varepsilon_s\} > 1$ solution of $\varepsilon_s = 4.968 + i1.361$ in Fig.(3d).

For scatterers that are quite small with respect to the IA wavelength, like $kR = 0.3$ in Fig.(3a), one clearly observes peaks in the total absorption cross section associated with higher electric multi-pole modes, although only the electric dipole mode (red curve), satisfies the IA condition per its design. We remark that the magnetic dipole contributions (green curve) are insignificant at such sizes. For somewhat larger sub-$\lambda$ particles, like the $kR = 1$, the electric dipole IA solution in Fig.(3b), one remarks that the electric dipole IA is accompanied by non-negligible absorption in higher order modes including the magnetic dipole contribution (in green). This behavior is accentuated for $kR = 2$ with magnetic dipole contributions coming close to the IA condition in both the $\Re\{\varepsilon_s\} < 1$ and $\Re\{\varepsilon_s\} > 1$ designs. We remark however that even when IA occurs for both electric and magnetic dipole modes in the same particle, they cannot both be satisfied at the same frequency (for a homogeneous particle at least). We also underline the fact that for $\Re\{\varepsilon_s\} > 1$ absorbers, it can be easier to obtain magnetic dipole IA solutions than those of electric dipoles. The ability for small particles to produce strong magnetic dipole absorption at optical frequencies has also been remarked recently by other authors[26].

V. IDEAL ABSORPTION WITH REALISTIC MATERIALS

Materials exhibiting plasmonic resonances, like silver and gold, are good candidates for achieving ideal absorption at small particle sizes since they provide $\Re\{\varepsilon_s\} < 1$ and modest absorption over the visible and near visible frequencies. Even for these materials however, ideal absorption will only occur at certain frequencies and sizes, and generally requires exploiting at least one tunable parameter like the permittivity, $N_b$, of the background medium.

We plot in Fig.(4a), the path traced out by $\varepsilon_s$ for gold and silver in a water background, $N_b = 1.33$, as the vacuum wavelength of light varies from the near ultra-violet to the mid-visible range (using interpolated Johnson & Christy data[27]). The values of $\varepsilon_s$ required for producing the lowest electric dipole IA are also plotted in this figure as the...
FIG. 3: (Color on-line) Dimensionless absorption cross sections, $8\pi \sigma_{\text{abs}}/(3\lambda^2)$, plotted as a function of $kR$ for IA designed to occur at: (a) $kR = 0.3, \varepsilon_s = -2.21 + i0.0611$; (b) $kR = 1, \varepsilon_s = -1.87 + i2.91$; (c) $kR = 2, \varepsilon_s = 0.4809 + i1.148$; and (d) $kR = 2, \varepsilon_s = 4.968 + i1.361$. Total absorption cross sections are given by blue curves, electric dipole absorption by red curves, and magnetic dipole absorption by green curves.

Although inspection of Fig.(4a) indicates 3 possible IA solutions in a water background (cf. Fig.(I)), we caution that only the lowest particle sizes correspond to resonances dominated by a dipolar response. Dipole IA solutions at larger particles sizes will include significant absorption in higher order modes similar to that seen in Figs.(3c-d).

| $D_{IA}$ (nm) | $\lambda_{IA}^{(nm)}$ | $kR_{IA}$ | $\varepsilon_{Ag}$ |
|---------------|---------------------|-----------|------------------|
| 34.04         | 393.76              | 0.361     | -4.094 + i0.199  |
| 115.53        | 295.80              | 1.63      | 0.704 + i3.01    |
| 291.4         | 321.0               | 3.13      | 0.700 + i0.856   |
| $D_{IA}$ (nm) | $\lambda_{IA}^{(nm)}$ | $kR_{IA}$ | $\varepsilon_{Au}$ |
| 91.74         | 540.69              | 0.709     | -5.30 + i2.20    |
| 121.50        | 451.81              | 1.12      | -1.76 + i5.27    |
| 75.94         | 273.38              | 1.16      | -1.37 + i5.17    |

TABLE I: Particle diameters, $D_{IA}$, and vacuum wavelengths, $\lambda_{IA}^{(nm)}$, required to produce optical sinks of silver and gold in water, $N_b = 1.33$.

Analogous curves are plotted in Fig.(I) for a higher background index material, $N_b = 1.5$, like that typical of polymers and glass. At this higher index background medium, only one IA solution exists for both silver and gold. The IA values for gold and silver deduced from an analysis of the intercepts between the IA conditions and the experimental permittivity functions of Fig.(I) are given in Table I and Table II for $N_b = 1.33$ and $N_b = 1.5$ respectively.

Although IA is predicted for both silver and gold in transparent material media, one can expect significant differences in their IA behavior in view of the considerable differences in their respective dispersions relations and predicted IA sizes. This is indeed the case, as illustrated in Fig.(5) where the scattering and absorption cross sections of the silver
FIG. 4: (Color on-line) Values of $\varepsilon_s$ obtained by varying vacuum wavelength, for silver (red) $\lambda_v \in \{188 - 500\}$nm and gold (magenta) $\lambda_v \in \{188 - 600\}$nm suspended in a (a) water background medium, $N_b = 1.33$, and (b) glass background medium, $N_b = 1.5$. The values of $\varepsilon_s$ required for IA in the lowest electric dipole mode are plotted in dashed blue for $kR$ varying from 0 to 4. Dipole model predictions of Eq.(9) are in plotted in dashed green.

| $D_{IA}$ (nm) | $\lambda_{IA}^{(v)}$ (nm) | $kR_{IA}$ | $\varepsilon_{Ag}$ |
|--------------|-----------------|-----------|-----------------|
| 30.62        | 412.77          | 0.35      | -5.16 + i0.227  |
| 76.23        | 559.68          | 0.64      | -6.57 + i1.95   |

TABLE II: Particle diameters, $D_{IA}$, and vacuum wavelengths, $\lambda_{IA}^{(v)}$, required to produce optical sinks of silver and gold in a $N_b=1.5$ background medium.

(a)-(b) and gold particles (c)-(d) are plotted for frequencies in the visible range for particles whose IA diameters are taken from Table II ($D_{IA} = 30.6$nm ($\lambda_v = 413$nm) for silver and $D_{IA} = 76.2$nm $\lambda_v = 560$nm for gold).

The frequencies at which IA is predicted are indicated by vertical dashed lines, and the IA cross sections of $8\pi\sigma_{abs}/(3\lambda^2) = 1$ are indicated by horizontal dashed lines in these figures. The total absorption and scattering cross sections are drawn in blue, and electric dipole contributions are drawn in red, but these curves are nearly indistinguishable on the scale of these graphs except for short wavelengths in gold. Magnetic dipole contributions are plotted in green, but are negligible in all graphs excepting some mild magnetic dipole absorption in gold at short wavelengths.

In Fig. 5, one remarks significant differences between the respective behaviors of silver and gold IA particles near their IA resonances. Notably, the electric quadrupole, magnetic dipole and higher order corrections are essentially negligible near the silver IA resonance, while non-electric dipole orders contribute significantly to absorption (but not scattering) at wavelengths below the IA resonance in gold. One should also remark that although the dipole contributions to the absorption cross sections are limited by their theoretical upper bound of $\sigma_{abs} = 3\lambda^2/(8\pi)$, the electric dipole scattering cross section of gold rises to values above $3\lambda^2/(8\pi)$, at frequencies below the IA resonance, but this is allowed by the general theoretical considerations of Eq.(5) where one sees that the scattering cross section of a dipole mode is only required by unitarity to satisfy $\sigma_{scat} \leq 3\lambda^2/(2\pi)$. Nevertheless, one can see in both Fig.(5c) and Fig.(5d) that $\sigma_{scat} = 3\lambda^2/(8\pi)$ at the IA resonance as required by the general theoretical restraint of Eq.(6).
Absorption cross section  
Scattering cross section

FIG. 5: (Color on-line) Cross sections (blue) of IA particles suspended in a $N_b = 1.5$ background medium. Cross section for silver are plotted in: (a)-$\sigma_{\text{abs}}$ and (b)-$\sigma_{\text{scat}}$ and those for gold in: (c)-$\sigma_{\text{abs}}$ and (d)-$\sigma_{\text{scat}}$ over the visible frequencies, $\lambda_v \in \{350 - 800\}$nm. Particle diameters are $D_{\text{IA}} = 30.6$nm for silver and $D_{\text{IA}} = 76.2$nm for gold, with their respective IA frequencies specified by vertical dashed lines. Dipole contributions to the cross sections are plotted in red for the electric dipole and green for the magnetic dipole.

VI. IDEAL ABSORPTION FOR COATED SPHERES

Although we saw that changing the background material index allows some control over IA frequency with homogeneous inclusions, applications are likely to want to design IA to occur at particular frequencies with a set of available materials. This will require additional adjustable geometric parameters and concentric coatings is one of the simplest ways to achieve this.

If we consider inclusions consisting of a core and a single concentric coating, one has two adjustable parameters, the radius of the outer shell $R$, and its concentric spherical core, $R_c$ (cf. Fig.(6). It proves convenient to fix these two dimensioned parameters, $R$ and $R_c$, in terms of the dimensionless size parameter, $kR$, and core material filling factor $f \equiv (R_c/R)^3$. Unlike the homogeneous sphere studied in the previous section, the analytic properties of the $S$ matrix cannot be directly exploited to determine the parameters producing IA solutions, since no IA solutions are guaranteed to exist when varying the parameters $kR$ and $f$ of a coated sphere with fixed permittivities.

Given the complexity of the analytic coated sphere solutions, the possible non-existence of solutions at finite frequency, and finally the fact that there are multiple adjustable parameters, it is advantageous to develop compact approximate formulas that allow one to rapidly determine which parameters and materials can provide viable designs for coated sphere IA. We derive now an approximation which allows the design of core-shell electric dipole IA scatterers by taking advantage of the fact that one is generally interested in designing sub-wavelength IA particles.

We saw that Eq.(13a) provides an accurate approximation for the required IA permittivity contrast, $\varepsilon_s$, over the entire range of the size parameter $\rho = kR$. The Maxwell-Garnett effective medium approach is derived as the effective permittivity of a concentric sub-wavelength core-shell particle whose volume fraction matches that of the bulk material.

The effective permittivity, $\varepsilon_{\text{eff}}$, of a core-shell system can thus be written as a function of filling fraction, $f$:  

$$\varepsilon_{\text{eff}} = \varepsilon_{\text{sh}} \left( 1 + 3f \frac{\varepsilon_c - \varepsilon_{\text{sh}}}{\varepsilon_c + 2\varepsilon_{\text{sh}} - f(\varepsilon_c - \varepsilon_{\text{sh}})} \right), \quad (14)$$

where $\varepsilon_{\text{sh}}$ and $\varepsilon_c$ are the shell and core relative permittivities respectively. This equation can then be algebraically
FIG. 6: (Color on-line) Schematic of a concentric coated sphere where the core, shell, and background permittivities are denoted respectively \( \varepsilon_c \), \( \varepsilon_{sh} \), and \( \varepsilon_b \). The filling fraction of the core material is defined as \( f \equiv (R_c/R)^3 \).

inverted to yields filling fraction as a function of \( \varepsilon_{eff} \) i.e.:

\[
f = \frac{\varepsilon_{eff} - \varepsilon_c \varepsilon_{sh} + 2 \varepsilon_c}{\varepsilon_{sh} - \varepsilon_c \varepsilon_{eff} + 2 \varepsilon_c}.
\]  

(15)

One can then solve for core-shell IA by replacing the “effective” index in Eq.(15) by the \( \varepsilon^{(e)} \) of Eq.(13a) that specifies the permittivity required for the IA condition. Since \( \varepsilon_{eff} \) and at least one of the materials is complex valued, the filling fraction, \( f \), found by Eq.(15) is generally complex, but one can vary the size parameter, \( \rho = kR \) to obtain a \( 3\{f\} = 0 \) solution graphically. Provided that the real part of the filling fraction is less than 1 when \( 3\{f\} = 0 \), then one has found a valid IA solution.

We apply the graphic solution method described in the previous paragraph to the core-shell IA design consisting of a silver or gold metallic layer around a lossless high index core of \( N_c = 2.8 \) in a background medium of index \( N_b = 1.5 \). The results are illustrated in Fig.(7). The filling fractions of the shell material and the radius of the particle are given as a function of the vacuum wavelength of the incident field. We remark that the chosen core-shell design with a high index center and a metallic shell allows one to design IA for any wavelength higher than the homogeneous sphere solutions given in Table (II). It was also possible to design IA for other geometries, like a dielectric shells around a metallic core, but the metallic shell design illustrated here tended to give the smallest IA solutions and seems more realizable from a practical standpoint.

VII. CONCLUSIONS

We saw in this work that the IA criteria takes the form of a limit behavior of an absorption cross section channel. Precise electromagnetic calculations in the complex frequency plane were shown to be able to predict this phenomenon, and new analytic expressions were showed to accurately predict the lowest order electric and magnetic modes. Precise calculations also indicated the intriguing possibility of observing IA in high index absorbing materials and not just in metals.

We showed that IA should be observable at visible frequencies for homogeneous spheres composed of gold or silver plasmonic particles at given frequencies and particle sizes. The signature of IA in terms of cross sections was also presented and should prove useful in experiments. The methods and formulas developed in this work led to a simple scheme for approximately predicting core-shell geometries supporting effective IA conditions throughout the visible spectrum. Experimental observations of IA for such sub-wavelength particles, could help to better understand IA and its experimental signature in quantum systems.

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FIG. 7: (Color on-line) Plots of the filling fraction (a) and the size parameter (b), required to achieve IA in a core shell structure immersed in a $N_b = 1.5$ background material. The core is considered to be a lossless high index material, $N_c = 2.8$, and the shell either silver (red-solid line) or gold (blue-dashed line).

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