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OPTICAL PHYSICS

Intuitive understanding of extinction of small particles in absorbing and active host media within the MLWA

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In an absorbing or an active host medium characterized by a complex refractive index $n_2 = n_2' + \mathrm{i}n_2''$, our previously developed modified dipole long-wave approximation (MLWA) is shown to essentially overlie with the exact Mie theory results for localized surface plasmon resonance of spherical nanoparticles with radius $a \lesssim 25$ nm ($a \lesssim 20$ nm) in the case of Ag and Au (Al and Mg) nanoparticles. The agreement for Au and Ag (Al and Mg) nanoparticles, slightly better in the case of Au than Ag, continues to be acceptable up to $a \sim 50$ nm ($a \sim 40$ nm), and can be used, at least qualitatively, up to $a \sim 70$ nm ($a \sim 50$ nm) correspondingly. A first order analytic perturbation theory (PT) in a normalized extinction coefficient, $\bar{\kappa} = n_2''/n_2'$, around a nonabsorbing host is developed within the dipole MLWA and its properties are investigated. It is shown that, in a suitable parameter range, the PT can reliably isolate and capture the effect of host absorption or host gain on the overall extinction efficiency of various plasmonic nanoparticles.

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1. INTRODUCTION

Electromagnetic scattering in an absorbing host characterized by a complex refractive index $n_2 = n_2' + \mathrm{i}n_2''$ (where n_2' and n_2'' are real) has more than 50 years of history. The traditional scattering theory neglects the host dissipation and gain [1,2], because those cases imply either a vanishing or infinite scattering wave at the spatial infinity. Once wave number $k = k' + \mathrm{i}k'' = 2\pi n_2/\lambda_0$, where λ_0 is the *vacuum* wavelength, is a complex number, i.e., $k'' \neq 0$, conventional expressions for cross sections cannot be straightforwardly extended for $k'' \neq 0$, because the expressions yield cross sections as *complex* quantities. Not surprisingly, the history of scattering in an absorbing host is filled in with a number of controversies [3–19]. This is probably why in classical textbooks it is only fleetingly mentioned in Section 12.1.3 of Ref. [2]. Already the definition of an incident intensity is not straightforward, as the field incident on a particle is different at different points on the particle as a result of an absorbing medium [4,5]. A great deal of effort was required to arrive at suitable definitions of cross sections for $k'' \neq 0$.

In contrast to the conventional case of $k'' = 0$, two sets of cross sections are commonly used: *inherent* and *apparent*. The former is obtained by performing surface integrals of corresponding Poynting vectors over the particle surface. The approach was developed a long time ago [4,5], but currently accepted expressions for the *inherent* cross sections were not presented until 1999 [7–10]. The focus of the present work will be on the apparent extinction cross section, C_{ext}, which is operationally defined in the far field (see Section 2.A below). The history of *apparent* cross sections began with the optical theorem of Bohren and Gilra [6]. Here, too, it took a long time, until 2007, to arrive at definitive expressions for the apparent extinction cross section, C_{ext} [11–14]. A curious feature of the apparent extinction cross section in an absorbing host is that it can be *negative* [20], which is neither an artifact of numerical simulations nor it violates any physical law. The apparent extinction cross section quantifies the difference in the readings of a forward-scattering detector taken with and without the particle. If the surrounding medium is absorbing, the presence of the particle can in fact make the detector signal stronger, thereby implying a negative extinction cross section. There is no violation of the energy conservation law, since in this case the extinction cross section is not used to quantify the energy budget

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of a finite volume encompassing the particle [20]. Another curiosity of apparent cross sections in an absorbing host is that an intrinsic definition of an apparent absorption cross section, C_{abs}, is still missing. The difficulty lies in that the very presence of a particle necessarily modifies the near field around the particle. The latter may be the cause of an additional absorption in the host medium *outside* the particle compared to what is happening in the absence of the particle [8,9]. The critical point is that because the field is disturbed by the particle, additional absorption may be realized in the medium external to the particle [21]. Consequently, unlike C_{abs} and C_{sca}, only apparent extinction cross section C_{ext} can be consistently defined (see Section 2.A below).

The focus of the present work is on the apparent extinction cross section C_{ext} in the so-called *modified long-wave approximation* (MLWA) [17,19,22–34]. First, it will be shown that the apparent extinction cross section conforms better to physical intuition than the intrinsic extinction cross section. Second, the MLWA [17,19,22–34], which can be viewed as a next-order approximation beyond the Rayleigh limit, is known to overcome a number of severe deficiencies of the Rayleigh limit [see Eq. (2) below] and, at least for nonabsorbing hosts, be surprisingly precise [17,19,27,31,33,34].

The outline of our contribution is as follows. Section 2 first recalls in its Subsection 2.A the expression for distance-independent apparent extinction cross section C_{ext} in the framework of the Lorenz–Mie theory [13]. In Subsection 2.B our earlier developed MLWA [33,34] is summarized. In Subsection 2.C, still within the framework of MLWA, an analytic first order perturbation theory (PT) in a normalized extinction coefficient, $\bar{\kappa} = n_2''/n_2'$, representing the magnitude of the host dissipation, is developed. A motivation for developing the above PT is to isolate and capture the effect of the host absorption or host gain on the overall extinction, which is not possible within the MLWA (the latter captures only overall extinction). In Section 3, performance of the above approximations in describing localized surface plasmon resonances of spherical plasmonic nanoparticles in different absorbing and active hosts is investigated. Discussion of some of the observed features is provided in Section 4. We then conclude with Section 5.

2. THEORY

A. Apparent Extinction Cross Section in the Framework of the Lorenz–Mie Theory

The specific expression for distance-independent apparent extinction cross section C_{ext} in the framework of the Lorenz–Mie theory [13] is given as an infinite sum over different multipole orders $\ell \geq 1$ (see Fig. 1):

$$C_{\mathrm{ext}} = -\frac{2\pi}{k'} \mathrm{Re} \left\{ \sum_{\ell=1}^{\infty} \frac{1}{k} (2\ell+1)(T_{E\ell} + T_{M\ell}) \right\}. \quad \mathbf{(1)}$$

In the optical convention, $T_{p\ell}$ correspond to the familiar Mie's expansion coefficients a_ℓ and b_ℓ {Eq. (4.53) of Ref. [2]}, i.e., $T_{E\ell} = -a_\ell$ for electric, or transverse magnetic (TM), polarization, and $T_{M\ell} = -b_\ell$ for magnetic, or transverse electric (TE), polarization. The physical, or operational, meaning of the distance-independent apparent C_{ext} is that it determines the reading of a polarization-sensitive well-collimated radiometer (WCR) at a sufficiently large distance r from the particle [13]:

$$\text{WCR signal} \propto \exp(-2k''r)(\Omega - C_{\mathrm{ext}}) I_{\mathrm{inc}},$$

where Ω is the area of the objective lens of the WCR, and I_{inc} is the intensity of the incident homogeneous (uniform) plane wave at the center of the particle. Importantly, the distance-independent extinction cross section *cannot* be introduced in the context of evaluating the energy budget of an arbitrarily shaped volume containing the scattering particle [13].

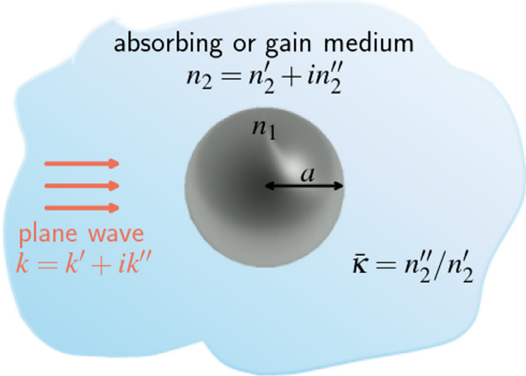

Fig. 1. Schematic representation of the problem under consideration. A spherical particle with radius a and refractive index n_1 is embedded in an absorbing or gain medium with $n_2 = n_2' + in_2''$, where n_2' and n_2'' are real. Imaginary refractive index of the host $n_2'' > 0$ for an absorbing medium and $n_2'' < 0$ for a gain medium, and $n_2'' = 0$ for a transparent medium. Normalized extinction coefficient $\bar{\kappa} = n_2''/n_2'$ is introduced to quantify the magnitude of the host dissipation or gain.

B. Dipole MLWA

The MLWA is a rational approximation to the Mie coefficients in terms of a fraction of simple polynomials in size parameter x that in a concise way combines three different elementary terms, involving a *size-independent* quasi-static Fröhlich term,

$$F_{p\ell} := \upsilon + \frac{\ell+1}{\ell},$$

the *dynamic depolarization* ($D_{p\ell} \sim x^2$), and the *radiative reaction* ($R_{p\ell} \sim x^{2\ell+1}$ for $p = E$ and $R_{p\ell} \sim x^{2\ell+3}$ for $p = M$) in the functional form [17,19,22–34]

$$T_{p\ell} = \frac{i R_{p\ell}(x)}{F_{p\ell} + D_{p\ell}(x) - i R_{p\ell}(x)}. \quad \mathbf{(2)}$$

Assuming nonmagnetic media, one has $\upsilon = \mu_1/\mu_2 = 1$ for magnetic (TE) polarization ($p = M$), and $\upsilon = \varepsilon := \varepsilon_1/\varepsilon_2$ for electric (TM) polarization ($p = E$), where the subscript 1 in Eq. (2) identifies the relevant quantities of a sphere (host).

The respective terms $F_{p\ell}$, $D_{p\ell}$, $R_{p\ell}$ in the functional form of Eq. (2) have well-defined physical origin and meaning (see Fig. 2). This allows for an intuitive understanding of scattering from small particles, which, as it will be shown, allows for rather reliable substitution of infinite series expansion in terms of Bessel functions of the conventional Mie solution by

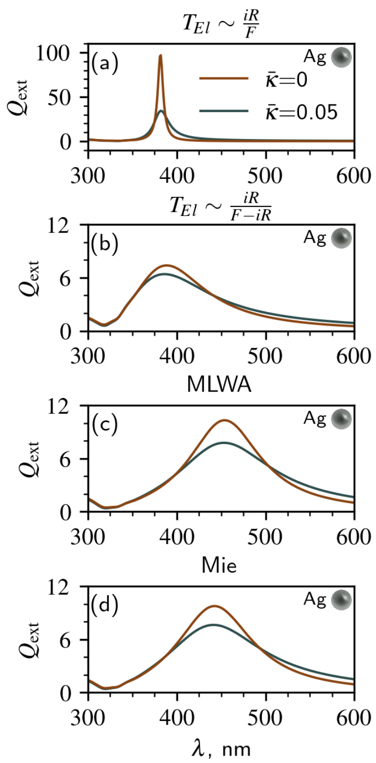

Fig. 2. Demonstration of different physical mechanisms described within MLWA on the extinction spectra of Ag sphere with $a = 40$ nm in the host with $n_2' = 1.33$. Here $\bar{\kappa} = 0$ for a transparent host and $\bar{\kappa} = 0.05$ in the case of a dissipative host. The refractive indices of Ag were taken from Ref. [35]. (a) Quasi-static Rayleigh approximation; (b) radiative correction taking the effect of retardation with respect to the incident field; (c) the MLWA including additionally the dynamic depolarization term. Obviously all the correction terms of the dipole MLWA given by Eq. (3) are necessary to achieve a fairly accurate approximation to the exact Mie theory shown in (d) for a comparison. All the above approximations, but the exact Mie theory, assume a constant field inside the sphere.

a single dipole term. The functional form of Eq. (2) makes it transparent that the usual Rayleigh limit, which amounts to setting $D_{p\ell}(x) = R_{p\ell}(x) \equiv 0$ in the denominator for $\ell = 1$ and $p = E$, is recovered for x, $x_s \ll 1$. Here x is in general complex size parameter, $x = 2\pi a/\lambda$, with λ being the wavelength in the host medium, whereas $x_s = 2\pi n_2 a/n_1\lambda = x/\sqrt{\varepsilon}$. The vanishing of the size-independent F in the denominator yields the usual quasi-static Fröhlich condition, which determines the quasi-static frequencies $\omega_{0\ell}$ for the occurrence of a localized surface plasmon resonance (LSPR). In what follows, we shall focus on the dipole MLWA, which yields [33,34]

$$T_{E1} \approx \frac{2i(\varepsilon-1)x^3/3}{\varepsilon+2-3(\varepsilon-2)x^2/5-2i(\varepsilon-1)x^3/3}, \tag{3}$$

where $\varepsilon = \varepsilon_1/\varepsilon_2$ is the relative dielectric function. An exceptional feature of the MLWA dipole contribution is that one can determine analytically an exact position of the complex pole of T_{E1} (the Mie coefficient $-a_1$), and hence the dipolar LSPR position, at

$$\varepsilon_{E1} = -2 \times \frac{1+3x^2/5+ix^3/3}{1-3x^2/5-2ix^3/3}, \tag{4}$$

which corrects formula Eq. (6) of Ref. [19]. The proof of that ε_{E1} yields complex zero of the denominator D of a_1 is relegated to Section S3 in Supplement 1. In the limit of small x, one can expand the denominator of ε_{E1} as $\sim 1+3x^2/5+2ix^3/3$, whereby Eq. (4) reduces to the familiar classical Bohren and Huffman result {cf. Eq. (12.13) of Ref. [2]}:

$$\varepsilon_{E1} \sim \varepsilon_{\rm BH} \approx -2 - \frac{12x^2}{5} \quad (|x| \ll 1). \tag{5}$$

C. Perturbation Theory in a Normalized Extinction Coefficient within the MLWA

The motivation for developing the perturbation theory is to isolate and capture the effect of the host absorption or host gain on the overall extinction, which is not possible within the MLWA (the latter captures only overall extinction). To this end, a useful parametrization of the relative dielectric function ε between the sphere and the host, suitable for studying the departure from a nonabsorbing host, is {cf. Eq. (9) of Ref. [19]}

$$\varepsilon = \frac{\varepsilon_1}{\varepsilon_2} = \frac{(n_1'+in_1'')^2}{(n_2'+in_2'')^2} = \frac{(n_1'/n_2'+in_1''/n_2')^2}{(1+in_2''/n_2')^2} = \frac{\varepsilon_t}{(1+i\bar{\kappa})^2}, \tag{6}$$

where $\varepsilon_t = (n_1'/n_2' + in_1''/n_2')^2 = \varepsilon_1/(n_2')^2$ is the relative (in general complex number if ε_1 is complex) dielectric function between the sphere and a *nonabsorbing* host with real refractive index n_2', and $\bar{\kappa} = n_2''/n_2'$ is a normalized extinction coefficient representing the magnitude of the host dissipation. On substituting the Taylor expansion of the electric dipole term a_1,

$$a_1(\varepsilon) = a_1(\varepsilon_t) + \bar{\kappa}\left.\frac{da_1}{d\bar{\kappa}}\right|_{\varepsilon=\varepsilon_t} + \mathcal{O}(\bar{\kappa}^2),$$

in the expression Eq. (1) of the apparent cross section, $C_{\rm ext}$, it is in principle possible to provide in a systematic way the results for the extinction efficiency, $Q_{\rm ext} = C_{\rm ext}/\pi a^2$, in the first order of $\bar{\kappa}$ for both gain and absorbing media in the dipole approximation. The derivative $da_1/d\bar{\kappa}$ is determined by Eq. (S21) from Section S5 in Supplement 1:

$$\frac{da_1}{d\bar{\kappa}} = -12n_2'x'\bar{\kappa}\,{\rm Re}\left[\frac{2-\varepsilon_t(\varepsilon_t-1)+\frac{1}{5}(\varepsilon_t^2-\varepsilon_t+2)(x')^2}{n_2 D^2(\varepsilon_t,x')}\right], \tag{7}$$

where $x' = x_0 n_2'$, with $x_0 = 2\pi a/\lambda$ being the size parameter in the vacuum host, and

$$D(\varepsilon_t,x') = \varepsilon_t + 2 - 3(\varepsilon_t-2)(x')^2/5 - 2i(\varepsilon_t-1)(x')^3/3 \tag{8}$$

is the denominator D in Eq. (3) in the limit $\bar{\kappa} \to 0$. One finds eventually

$$Q_{\mathrm{ext}} = \frac{C_{\mathrm{ext}}}{\pi a^2} \approx \frac{2}{x'}\mathrm{Re}\left\{\frac{3}{x}\left[a_1(\varepsilon_t) + \bar{\kappa}\,\frac{da_1}{d\bar{\kappa}}\bigg|_{\varepsilon=\varepsilon_t}\right]\right\}$$
$$= \frac{2}{x'}\mathrm{Re}\left\{\frac{3}{x}a_1(\varepsilon_t)\right\}$$
$$- 12 n_2' x' \bar{\kappa}\,\mathrm{Re}\left[\frac{2-\varepsilon_t(\varepsilon_t-1)+\frac{1}{5}(\varepsilon_t^2-\varepsilon_t+2)(x')^2}{n_2 D^2(\varepsilon_t, x')}\right], \quad (9)$$

which defines the first order expansion of Q_{ext} in the parameter $\bar{\kappa}$ within the perturbation theory (PT). The first term on the rhs is the dipole MLWA in a nonabsorbing host characterized by ε_t. The second term on the rhs is the perturbation correction. In what follows, we will refer to Eq. (9) as the $\bar{\kappa}$-PT approximation.

3. RESULTS

A. Dipole MLWA versus Perturbation Theory

Figure 3 compares the performance of various approximations relative to the exact Mie theory involving

- the dipole MLWA [with the sole term T_{E1} in Eq. (1) given by Eq. (3)],
- the Mie theory approximation with a nonabsorbing host [determined by Eq. (1) with $n_2'' = 0$],
- the first order $\bar{\kappa}$-PT given by Eq. (9)

in a water-like host ($n_2' = 1.33$) for different host absorptions characterized by different values of $\bar{\kappa}$ on the example of Ag spheres with radii between 10 and 50 nm. Figure 4 compares results for spheres made from most common plasmonic materials Al, Ag, Au, Mg with radius $a = 25$ nm in a water-like host ($n_2' = 1.33$) with $\bar{\kappa} = \pm 0.01$ and ± 0.1 [similar results for a glass-like host ($n_2' = 1.5$) are presented in Fig. S2 in Supplement 1]. For different metals of plasmonic particles tabulated data of refractive indices have been used as follows: Al [35], Ag [35], Au [35], Mg [36]. As demonstrated in those figures, the dipole MLWA can be very precise. In the case of Ag and Au nanoparticles, the dipole MLWA essentially overlies with the exact Mie theory results for $a \lesssim 25$ nm. The agreement, slightly better in the case of Au than Ag, continues to be acceptable up to $a \sim 50$ nm (Fig. S1III in Supplement 1), and can be used, at least qualitatively, up to $a \sim 70$ nm (Fig. S1IV in Supplement 1). In the case of Al and Mg nanoparticles, a slight deviation from Mie theory results becomes visible by the naked eye already for $a \gtrsim 25$ nm (cf. Figs. S1I and S1II in Supplement 1). The agreement continues to be acceptable up to $a \sim 40$ nm and can be used at least qualitatively up to $a \sim 50$ nm (Fig. S1III in Supplement 1).

A first general observation is that with increasing sphere radius the performance of the dipole MLWA worsens. This is not surprising, as the MLWA is by definition a long wavelength

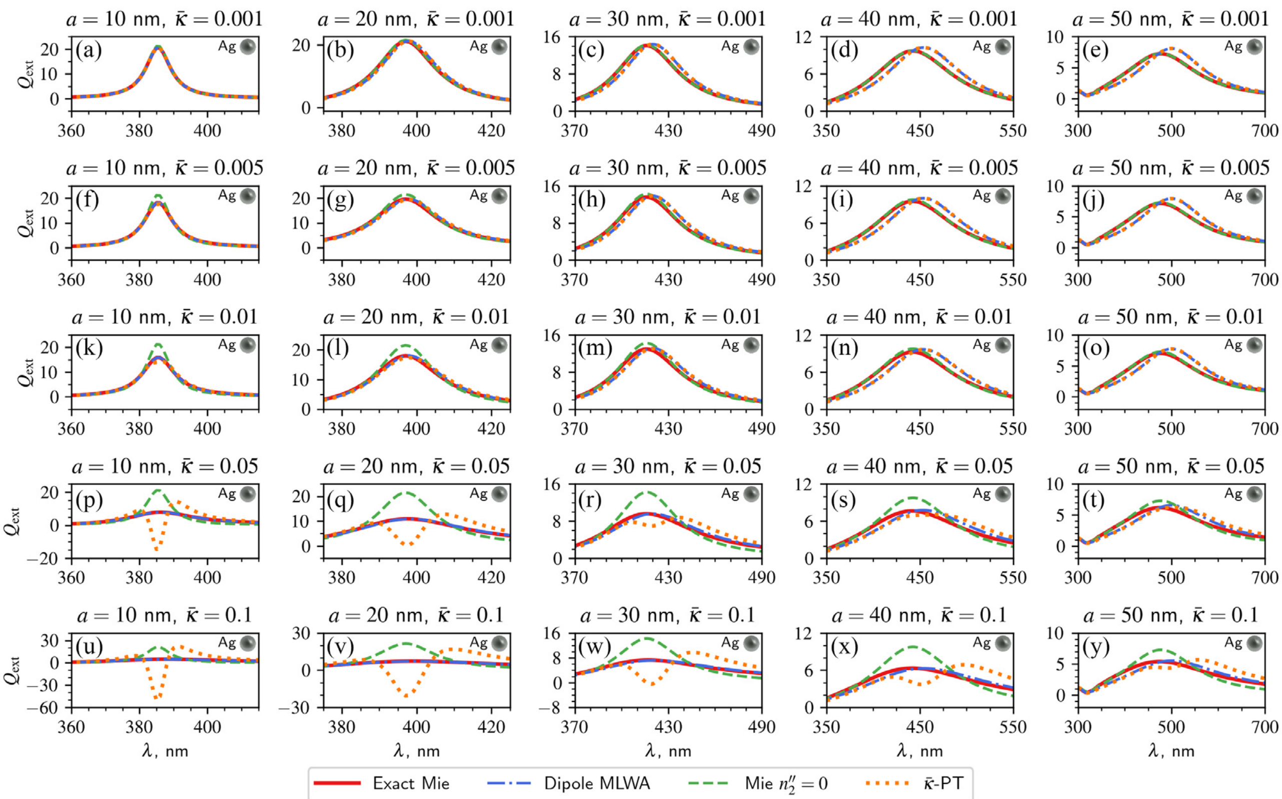

Fig. 3. Extinction spectra, Q_{ext}, for individual spherical Ag nanoparticles with radii $a = 10, 20, 30, 40, 50$ nm in water-like host ($n_2' = 1.33$) with $\bar{\kappa} = 0.001, 0.005, 0.01, 0.05, 0.1$, as shown in the legend for each plot. Spectra were calculated by Eq. (1) of the exact Mie theory (solid red line), the dipole MLWA by Eq. (3) (blue dotted-dashed line), the Mie theory in nonabsorbing host ($n_2'' = 0$; dashed green line), and the first order $\bar{\kappa}$-PT of our Eq. (9) (dotted orange line). Because the Mie theory in the nonabsorbing host is independent of $\bar{\kappa}$, its plots are identical at each given column. The refractive indices of Ag were taken from Ref. [35]. Note a broadening and suppression of plasmon resonances with increasing host absorption.

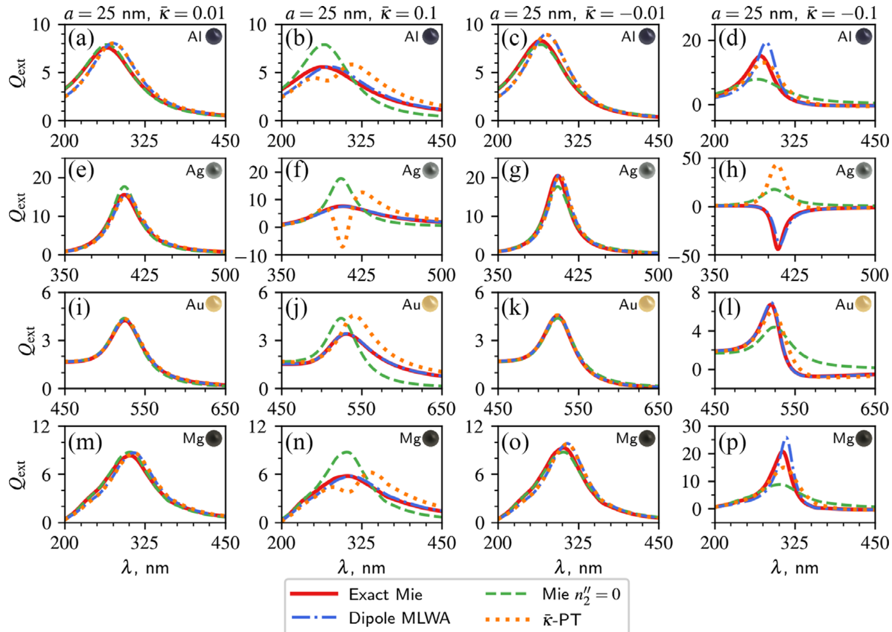

Fig. 4. Extinction spectra, Q_{ext}, for Al, Ag, Au, and Mg spherical nanoparticles with $a = 25$ nm embedded in a host absorption medium with $n_2' = 1.33$ (left two columns) and gain host medium (right two columns) for $\bar{\kappa} = 0.01$, 0.1 ($\bar{\kappa} = -0.01$, -0.1). Q_{ext} are shown as calculated by Eq. (1) of the Mie theory (solid red line), within dipole MLWA of Eq. (3) (blue dotted-dashed line), the Mie theory approximation with the nonabsorbing host (green dashed line), and by the $\bar{\kappa}$-PT given by Eq. (9) (dotted orange line). The refractive indices of Ag, Al, Au were taken from Ref. [35] and those of Mg from Ref. [36]. Whereas absorption decreases the height of a LSPR and broadens its width, the gain does just the opposite. Note enlarged scale on the vertical axis in the last column for $\bar{\kappa} = -0.1$.

approximation. The same applies to the $\bar{\kappa}$-PT. The $\bar{\kappa}$-PT overlies with the dipole MLWA for all $a \le 50$ nm, provided that $\bar{\kappa} \le 0.01$. The latter justifies *a posteriori* that our $\bar{\kappa}$-PT is correct. Another interesting tendency revealed by Fig. 3 (Figs. S1I and S1II in Supplement 1) is that the maximal radius a for which the dipole MLWA remains accurate increases with increasing $\bar{\kappa}$. This observation can be explained by the fact that for a given particle radius an increasing host absorption suppresses the contribution of higher-order multipoles that would normally arise at larger particle sizes. This allows the dipole MLWA to remain valid over a wider range of particle sizes, as the multipole effects are suppressed in absorbing media.

Importantly, the results of $\bar{\kappa}$-PT and the dipole MLWA overlie with the exact Mie theory results for $a \lesssim 25$ nm, which supports an earlier observation on the range of validity of the MLWA in an absorbing host by Khlebtsov [17]. Whereas the dipole MLWA continues to overlie with the exact Mie theory results for $a \lesssim 25$ nm irrespective of $\bar{\kappa}$, the results of the $\bar{\kappa}$-PT begin to deviate from those of the dipole MLWA for $\bar{\kappa} \gtrsim 0.05$. An extreme case of such a deviation is provided for the Ag sphere with $a = 25$ nm and $\bar{\kappa} = \pm 0.1$ shown in Figs. 4(f) and 4(h). Whereas the dipole MLWA nearly overlies the exact result showing a positive (negative) extinction for $\bar{\kappa} = 0.1$ ($\bar{\kappa} = -0.1$), the $\bar{\kappa}$-PT in the respective cases indicates a negative (positive) extinction. This is to be expected, because the $\bar{\kappa}$-PT is a first order perturbation theory of the dipole MLWA in $\bar{\kappa}$ and is expected to eventually break down above a certain threshold value of $|\bar{\kappa}|$. Negative extinction shown in Fig. 4(h) is an indication of that particle losses have been more than compensated for by the gain medium—a precursor of lasing action [37–39].

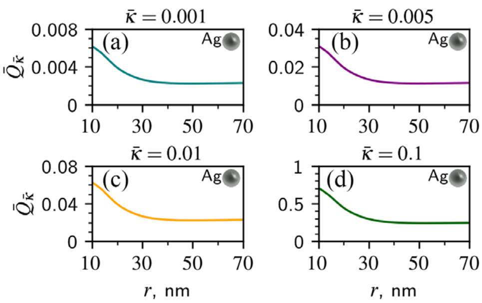

Fig. 5. Size dependence of $\bar{Q}_{\bar{\kappa}}$, the ratio of the PT correction to the leading term on the rhs of Eq. (9), for Ag sphere at the (size-dependent) LSPR wavelength for different $\bar{\kappa}$: (a) 0.001, (b) 0.005, (c) 0.01, and (d) 0.1.

Surprisingly, the smaller the particle, the greater the deviation of the $\bar{\kappa}$-PT relative to the dipole MLWA for $\bar{\kappa} \gtrsim 0.05$ (see Figs. S1 and S2 in Supplement 1). The origin of this behavior is that $D(\varepsilon_t)$ is typically small in a proximity of a LSPR [cf. the generalized Fröhlich condition Eq. (5)]. Whereas the first term in Eq. (9) is of the order $1/D(\varepsilon_t)$, the term proportional to $\bar{\kappa}$ is of the order $1/D^2(\varepsilon_t)$. However, when the size parameter x increases, $|D(\varepsilon_t)|$ at a LSPR decreases (due to a larger separation from the complex zero), and, as illustrated in Fig. 5, $\bar{Q}_{\bar{\kappa}}$, defined as the ratio of the PT correction to the leading term on the rhs of Eq. (9), gradually decreases, whereby the first order $\bar{\kappa}$-PT begins to approximate the MLWA. Note also how the ratio $\bar{Q}_{\bar{\kappa}}$ increases with $\bar{\kappa}$, which, as expected, explains why the $\bar{\kappa}$-PT begins to deviate from the MLWA with increasing $\bar{\kappa}$.

B. Mie Theory Approximation with Nonabsorbing Host

Similar behavior is observed for the Mie theory approximation with a nonabsorbing host ($n_2'' = 0$) that begins to deviate from the exact Mie theory results, but somewhat earlier, beginning with $\bar{\kappa} = 0.01$. Again the smaller the particle, the greater the deviation. Nevertheless, in a relatively weakly absorbing host the Mie theory approximation with a nonabsorbing host becomes the best approximation. For example, the Mie theory in a nonabsorbing host begins to overlie with the Mie theory in an absorbing host for $a \gtrsim 50$ nm and $\bar{\kappa} = 0.01$ (see Fig. S1 in Supplement 1). A threshold radius for which it happens increases with $\bar{\kappa}$.

Why the Mie theory approximation with a nonabsorbing host becomes the best approximation for sufficiently large a can be explained by the increasing relevance of higher-order multipole contributions with increasing particle size. This is understandable, because with increasing a higher-order multipoles, which are obviously absent in any dipole approximation, become more and more relevant. This is demonstrated also in Fig. 6 showing the effect of increasing the radius of spherical Ag nanoparticles in a poly(3-hexylthiophene) (P3HT) host medium, which was used in a number of recent studies [15,17,19,40]. Unlike inherent dipole approximations, the Mie theory approximation with a nonabsorbing host captures reasonably well both the dipole and quadrupole peaks. Obviously, the higher-order MLWA of Ref. [34] could have captured the quadrupole peak, but this goes beyond the scope of the present study.

C. Isolating the Host Dissipative Effects on the Extinction Cross Sections

The dissipative effects of the host on the extinction cross sections, Q_{ext}, can obviously be isolated by subtracting from the exact value $Q_{\mathrm{ext}} = Q_{\mathrm{ext};\varepsilon}$ the value of $Q_{\mathrm{ext};\varepsilon_t}$ obtained by the Mie theory for the nonabsorbing host characterized by $n_2'' = 0$

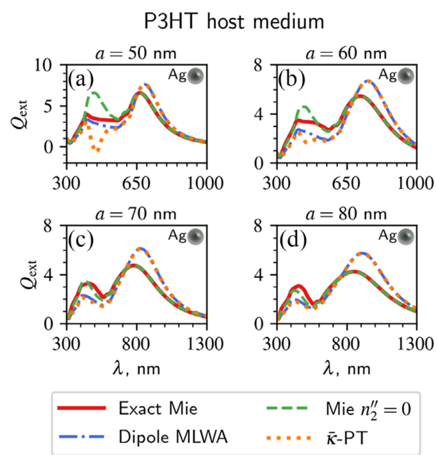

Fig. 6. Extinction spectra, Q_{ext}, for spherical Ag nanoparticles with (a) $a = 50$ nm, (b) $a = 60$ nm, (c) $a = 70$ nm, and (d) $a = 80$ nm embedded in poly(3-hexylthiophene) (P3HT) host medium. Spectra are shown as calculated with the exact Mie theory (solid red line), the Mie theory for nonabsorbing media ($n_2'' = 0$) (green dashed line), the dipole MLWA (blue dotted-dashed line), and the $\bar{\kappa}$-PT (dotted orange line).

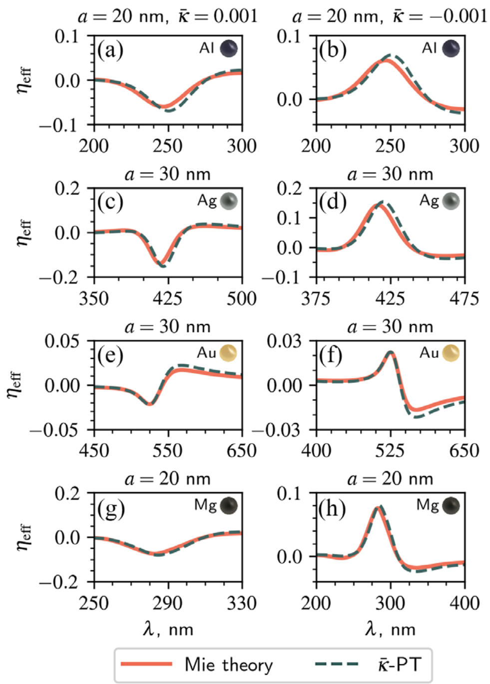

Fig. 7. The effect η_{eff} of the host absorption (left column) and the gain host medium (right column) on Q_{ext} in the Mie theory (solid red line) and the $\bar{\kappa}$-PT (dashed dark green line) for Al, Ag, Au, and Mg materials with different radii a embedded in the host medium with $n_2' = 1.33$ and $\bar{\kappa} = 0.001$ (left column) and $\bar{\kappa} = -0.001$ (right column).

(i.e., $\varepsilon \to \varepsilon_t$). In what follows, we denote the difference by $\eta_{\mathrm{eff}} := Q_{\mathrm{ext};\varepsilon} - Q_{\mathrm{ext};\varepsilon_t}$. Thus η_{eff} quantifies the contribution of the host absorption or gain in the resulting Q_{ext}. To our satisfaction, it turns out that, in a suitable parameter range, the analytic $\bar{\kappa}$-PT can reliably capture the effect of the host absorption on the extinction efficiency of a plasmonic nanosphere as demonstrated in Fig. 7. Not surprisingly, the agreement can also be reached in the case of gain media, which opens the door for analysis of promising applications involving active media, see Fig. 4 (Fig. S5 in Supplement 1), such as spasers [37–39]. Therefore, within the range of its validity, the first order $\bar{\kappa}$-PT allows one to both intuitively and analytically understand the mechanisms of host dissipation or gain on the extinction efficiency of a plasmonic nanosphere.

4. DISCUSSION

First-principles far-field computations based on the general Lorenz–Mie theory showed that increasing absorption in the host medium *broadens* and suppresses plasmon resonances in the apparent extinction [14]. Such a broadening and suppression of plasmon resonances is also present in all approximations shown in Figs. 3 and 4. The effect of absorption in the host thus goes in the same direction as increasing absorption within the particle. Noteworthy, increasing gain in the host medium *narrows* plasmon resonances and increases their amplitude in the apparent extinction as demonstrated in Fig. 4 (cf. also Fig. S3, Supplement 1). This goes along physical intuition. Contrary to the apparent extinction, in the case of the inherent cross sections a surrounding lossy medium was shown to *narrow* the plasmonic resonances, as well as *increase* their amplitude dramatically [15,17].

The dipole MLWA can, in principle, be used up to the laser threshold associated with the so-called spectral singularity of non-Hermitian models. Indeed, as it has been verified both experimentally and theoretically more than 15 years ago, a description of gain media consisting of changing the sign of extinction is a valid approximation up to the laser threshold [41]. This could be particularly important for applications in spasers and nano-lasers.

For the sake of completeness, a rather exhaustive MLWA analysis in the absorbing case has been recently provided by Khlebtsov [17] by employing slightly different MLWA. The study of Ref. [17] arrived at similar conclusions that MLWA is very precise for $a \lesssim 25$ nm. Our study complements Ref. [17] with (i) a first order analytic perturbation theory around a non-absorbing host in a normalized extinction coefficient $\bar{\kappa}$ ($\bar{\kappa}$-PT) and (ii) an investigation of gain media. In addition, we have examined the behavior of Al and Mg nanoparticles.

5. CONCLUSIONS

Our previously developed modified dipole long-wave approximation (MLWA) was shown to essentially overlie with the exact Mie theory results for $a \lesssim 25$ nm ($a \lesssim 20$ nm) in the case of Ag and Au (Al and Mg) nanoparticles. The agreement for Au and Ag (Al and Mg) nanoparticles, slightly better in the case of Au than Ag, continues to be acceptable up to $a \sim 50$ nm ($a \sim 40$ nm), and can be used, at least qualitatively, up to $a \sim 70$ nm ($a \sim 50$ nm). Expanding the discussion to the case of larger nanoparticles with $a > 70$ nm almost certainly requires consideration of MLWA for higher-order multipoles [31,34]. We developed within the dipole MLWA a first order analytic perturbation theory (PT) around a nonabsorbing host in a normalized extinction coefficient $\bar{\kappa}$ and investigated its properties. It was shown that, in a suitable parameter range, the $\bar{\kappa}$-PT can reliably isolate and capture the effect of host absorption or host gain on the overall extinction efficiency of spherical plasmonic nanoparticles. Considering growing interest in light-matter interactions, we expect that our results will help in designing optimal systems comprising plasmonic nanoparticles embedded in suitable dissipative or gain media for various applications, such as photothermal therapy [42,43], spasers, and other active medium devices [37–39].

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Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

Supplemental document. See Supplement 1 for supporting content.

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