Relating localized nanoparticle resonances to an associated antenna problem

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On an empirical basis, we indicate the possibility of conceptually unifying the description of resonances existing in some of the analytically studied metallic nanoparticles and optical nanowire antennas. To this end the nanoantenna is treated as a Fabry-Pérot-like resonator with arbitrary seminanoparticles forming the terminations. We show that the frequencies of the quasistatic dipolar resonances of the considered nanoparticles coincide with those where the round-trip phase of the complex reflection coefficient of the fundamental propagating plasmon polariton mode at the wire terminations amounts to 2π. The lowest order Fabry-Pérot resonance of the optical wire antenna occurs therefore even for a negligible wire length.

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I. INTRODUCTION

Small particles are among the earliest cases tackled by light scattering theory. The quasianalytical rigorous solution for spheres dates back to the pioneering work of Mie in 1908.[1] This approach can be considerably simplified if the size of the spheres is small compared to the illuminating wavelength resulting in the quasistatic approximation.[2,3] The resulting analytic formulas for the polarizability of the sphere exhibit resonant denominators, such as the well-known expression $\varepsilon_m(\nu) + 2\varepsilon_d(\nu) = 0$ for the dipole resonance of a metallic sphere in a dielectric host medium. In this case and generally, the permittivities of the spheres and their surroundings need to exhibit opposite signs at resonance. The scattered field exhibits strongly enhanced stationary evanescent components at the interface—a phenomenon that is termed localized surface plasmon polariton (LSPP).[4] Intriguingly, it was shown that in this approximation the quality factor of the resonance solely depends on the material properties rather than the particle shape,[5] which, however, affects the resonance frequency. Only for a few other particle shapes [e.g., ellipsoids and spherical shells or particles with a lower dimensionality (i.e., a cylinder)] the resonance condition can be put in a similar form known from the sphere. The exploitation of these LSPPs at nanoparticles of different shapes has led to various applications and is forming one branch of the prospering field of plasmonics.

If the metal is a perfect conductor, another resonance is supported by metallic wires if their length corresponds to a multiple of half the illumination wavelength. Such metallic wires constitute the basic building blocks of radio-frequency (RF) antennas. Recently, their downscaling into the visible attracted considerable interest and the field of optical antennas is now similarly established. Conceptually, optical antennas differ from RF antennas in that the field propagating along the wire is no more purely photonic but forms another polaritonic excitation.[6] This type of quasiparticle is referred to as a propagating surface plasmon polariton (PSPP) due to its sole energy transport in the propagation direction. As for any guided mode phenomenon, the PSPP dispersion relation may strongly depend on the wire's cross section (see, e.g., Ref. 7).

The origin of resonances in these finite-length nanowires is well understood in terms of Fabry-Pérot resonances of the PSPP mode confined between the partially reflecting wire terminations.[8–12] Unlike in antennas at microwave frequencies, here the reflection coefficients are complex valued, providing an additional phase term that mimics an increase of the wire length and depends on the actual shape of the termination. This resembles the situation in a planar Fabry-Pérot resonator with Bragg mirrors, where the number of layers also affects the actual phase shift and thus the resonance condition. This is also the reason why multiples of half the resonance wavelength differ from the wire length.[8,10–13] This peculiarity evoked research interest and both analytical and numerical results on the spectral and geometrical dependence of reflection coefficients were reported for abrupt or flat nanowire terminations.[14,15] Moreover, associated geometries (e.g., trenches, grooves, or slits on or in flat metal surfaces or metallic thin films) were analyzed with respect to their reflection and transmission properties of PSPP launched along the metal surfaces.[16] It allows for obtaining more insight into the underlying physics of phenomena observable in such systems. Examples thereof would be the enhanced transmission in subwavelength apertures.[17]

To date, aforementioned metallic nanoparticles are largely studied in terms of LSPP resonances whereas wire nanoantennas are commonly analyzed in terms of PSPP standing wave phenomena and these seemingly disparate approaches have not been systematically integrated. A few reports on variable length nanoantennas[13,18] consider spherical particles as the limiting case of cylindrical wires. Nevertheless, it is challenging to disclose the mechanism behind this convergence of resonances in analytically understood geometries at a physical level. Here we attempt to provide a unifying view on some of the analytically understood nanoparticles in research. It turns out that the LSPP resonances of all of them follow straightforwardly from solving the reflection problem at the nanowire termination.

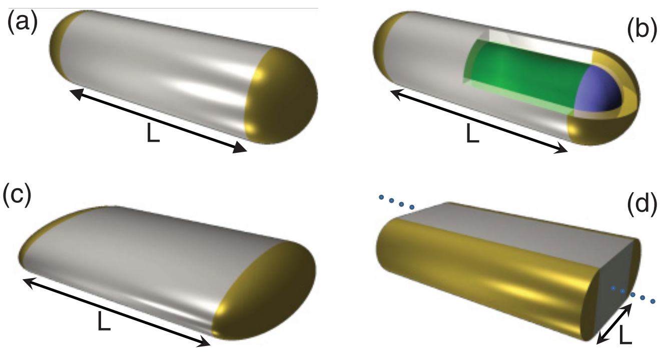

FIG. 1. (Color online) Sketch of considered metallic nanoantenna geometries. (a) Circular cylinder and (b) cylindrical shell with a hemispherical termination; (c) elliptical cylinder with a semi-elliptical termination; (d) one-dimensional (1D) nanoantenna with a semicylindrical termination. All nanoantennas are composed of a nanowire (metallic color) and a termination (golden) while L is the nanowire's length.

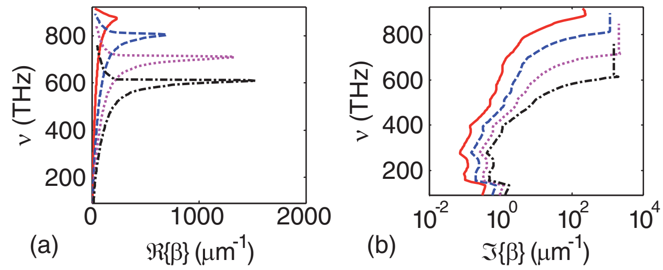

FIG. 2. (Color online) (a) Real and (b) imaginary part of the propagation constant β of the lowest order PSPP mode as a function of frequency ν for selected values of ε_d; $\varepsilon_d = 1$ (solid red), $\varepsilon_d = 2.8$ (dashed blue), $\varepsilon_d = 5.4$ (dotted magenta), $\varepsilon_d = 9$ (dotted-dashed black). The inset shows the H_ϕ-field norm for a core radius of 10 nm and $\varepsilon_d = 1$.

II. MODELLING SPHERE AS AN ANTENNA

Beginning with the simplest case, we take nanowires of circular cross section with hemispherical terminations at both ends [Fig. 1(a)]. This nanoantenna becomes a sphere for vanishing wire length L. We hypothesize that the dipolar resonance of a sphere that is observable in the far field (e.g., as a peak in the scattering cross section is caused by the constructive interference of the forward and backward propagating fundamental ($m = 0$) PSPP mode of the nanowire).[19–21] Then, the necessary condition for a Fabry-Pérot resonance—that is, the round trip reproduction of the phase factor—is a total phase accumulation of an integer multiple of 2π. This condition can already be met for a wire with negligible length (i.e., the sphere). It implies a phase shift of π upon reflection at each termination. In general, higher order PSPP modes do not need to be considered since all modes with $|m| \geqslant 2$ cut-off below a threshold wire radius.[22] Moreover, the PSPP mode with $|m| = 1$ diverges for a vanishing radius, thereby suffering from increasing radiation losses. Therefore, it cannot be excited anymore[23] and the system becomes simple enough to be understood in terms of a fundamental ($m = 0$) PSPP mode. In the limit when the radius becomes comparable to the incident wavelength, our model requires further substantiation owing to additional dynamics brought in by the higher order modes.

We use the COMSOL MULTIPHYSICS simulation platform to numerically solve the reflection problem at the wire termination. The metallic nanowire is assumed to be semi-infinite and it is surrounded by a dielectric medium with a permittivity ε_d. The computational cell is enclosed by perfectly matched layers to mimic an open space. Silver (Ag) is used as the metal, with its dispersive permittivity fully considered.[24] The cylindrical wire has a radius of 10 nm. The exact value of the radius is not important provided it is much less than the wavelength.

First we calculate the dispersion relation of the fundamental wire eigenmode and subsequently use this mode as illumination of the termination of the semi-infinite wire. Then the total (incident and scattered) field is calculated in a plane normal to the wire axis and located at $z = 0$. The complex reflection coefficient of this mode at the termination is extracted by employing the orthogonality relations obtained through unconjugated reciprocity theorem,[25] which yields an expression for reflection coefficient as

$$r = -\exp^{-i2\beta L} \frac{\int_0^\infty E_{\rho,0}(\rho,0)[H_{\phi,\mathrm{tot}}(\rho,0) - H_{\phi,0}(\rho,0)]\rho\, d\rho}{\int_0^\infty E_{\rho,0}(\rho,0)H_{\phi,0}(\rho,0)\rho\, d\rho} \tag{1}$$

where $H_{\phi,0}(\rho,0)$ being the azimuthal magnetic field and $E_{\rho,0}(\rho,0)$ the radial electric field components of the incident PSPP mode ($m = 0$) at $z = 0$, β is the associated propagation constant and L is the distance between origin ($z = 0$) and the initial plane of the antenna termination. All quantities, except the wire geometry, depend on frequency. In passing we note that the distinction of what belongs to the wire and what belongs to the termination is arbitrary to a certain extent. The phase accumulated due to propagation and the phase accumulation due to reflection can easily be merged. However, since we wish to discuss solely the properties of the termination, the length L is understood as the length of the nanoantenna along which no change of the cross-sectional profile occurs.

In Fig. 2 the complex-valued propagation constant β of the lowest order PSPP mode is displayed as a function of the frequency and the permittivity of the surrounding medium. The real part exhibits the usual dispersion characteristic where the propagation constant increases with frequency until back bending sets in. This back bending is associated with a strongly increasing damping (imaginary part). In the succeeding spectral domain, any analysis of the reflection coefficient tends to be cumbersome since dissipation entirely dominates the system.

Figure 3 shows the complex reflection coefficient as a function of frequency for different ε_d extracted from the simulation of a semi-infinite wire for the respective lowest order PSPP mode. It can be seen that at low frequencies the modulus is constant and large with a phase shift around zero, suggesting a perfect metal-like behavior. The phase increases with frequency and undergoes an abrupt change at a critical frequency. This jump is associated with the decrease in the reflected amplitude and it appears in the frequency interval where back bending occurs. Now it is easy to extract the frequency where a phase jump of π occurs and to compare it to the resonance frequency predicted by the quasistatic theory

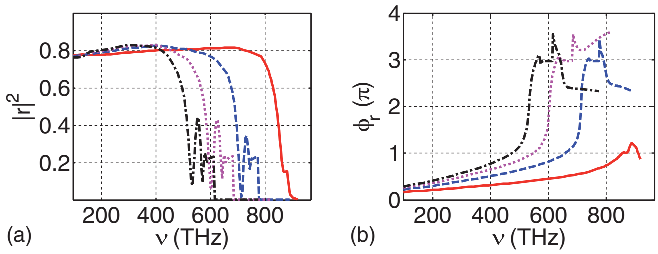

FIG. 3. (Color online) (a) Amplitude and (b) phase of the reflection coefficient of the nanowire for selected values of ε_d; $\varepsilon_d = 1$ (solid red), $\varepsilon_d = 2.8$ (dashed blue), $\varepsilon_d = 5.4$ (dotted magenta), $\varepsilon_d = 9$ (dotted-dashed black).

for a small sphere. Additionally, it can be seen that the phase surpasses even values corresponding to multiples of π. As the scope of this work is restricted only to resonances visible in the analytical formulation of quasistatic limit, we will not attempt to disclose any possible relation of these higher order Fabry-Pérot resonances to the ones accessible in rigorous Mie theory. But it suffices to say that for isolated particles such resonances are not observed because they are dipole forbidden and suffer from excessive damping.

Figure 4(a) shows a comparison of the resonance frequencies predicted by the quasistatic theory as well as the PSPP reflection calculation for different surrounding media. The excellent agreement between both approaches demonstrates that the resonances of a sphere in the quasistatic limit are directly related to the corresponding limit of the nanoantenna problem.

III. ADDITIONAL EXAMPLES

A. Nanoshell

To further investigate the applicability of this conclusion, we briefly analyze other well-known particle geometries in the following. Another special case of a spherical nanoparticle is one with a dielectric core and a metallic shell. It is of appreciable practical relevance since its resonance frequency can be tuned across the entire visible spectral region by varying the metallic shell thickness.[26] Such particles exhibit lower and higher energy LSPP resonances, which appear as a result of the hybridization of individual resonances of the constituting metallic sphere and dielectric void.[27] We approximate the structure similarly as before by a nanowire made of a dielectric core surrounded by a metallic shell terminated with a hemispherical shell of the same construction [Fig. 1(b)]. Guided by intuition, we regard the lower (upper) branch of the fundamental PSPP mode of the cylindrical metallic shell wire[28] as responsible for the lower (higher) resonance frequency of these particles. We repeat the aforementioned analysis for the lower branch PSPP mode and show a similar comparison between resonance frequencies predicted by quasistatic approximation and the reflected field where $\phi_r = \pi$ in Fig. 4(b) as a function of the ratio b/a between shell and core radii, respectively. An analysis of the highly dissipative upper branch was not attempted due to the conceptual difficulties associated with overdamped PSPPs (see also Fig. 2).

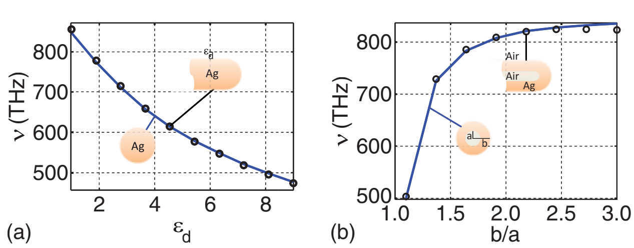

FIG. 4. (Color online) Resonance frequency of a sphere (a) with a radius of 10 nm as a function of permittivity of surrounding medium and of a spherical core shell particle (b) as a function of the ratio between shell and core radii b/a. The core radius $a = 10$ nm is fixed. The permittivity of both the core and the surrounding medium is $\varepsilon_d = 1$. Dots correspond to resonance frequencies as extracted from the phase of the reflection coefficients and the solid lines correspond to the predictions from quasistatic theory.

B. Ellipsoid

We extend the scope of our study toward ellipsoids which have a non-spherical geometry except in specific cases. First we consider an elliptical wire cross section [see Fig. 1(c)]. These particles can be envisioned straightforwardly to be composed of an elliptical nanowire (radii a and b) of negligible length and terminations consisting of rotational symmetric semi-ellipsoids [Fig. 1(c)]. Note that the resonance frequencies of ellipsoids depend upon the illuminating polarization.[29] We set the semi-axis a to be the symmetry axis and present the results for the electric field polarized both perpendicular and parallel to it. Figure 5(a) shows the resonance frequency as calculated from the phase of the reflected field at the wire termination compared to the predictions from quasistatic theory. In the antenna simulation the polarization is chosen by selecting the eigenmode corresponding to the nanowire under respective illumination. Overall we find good agreement in the predicted resonance frequencies by both methods.

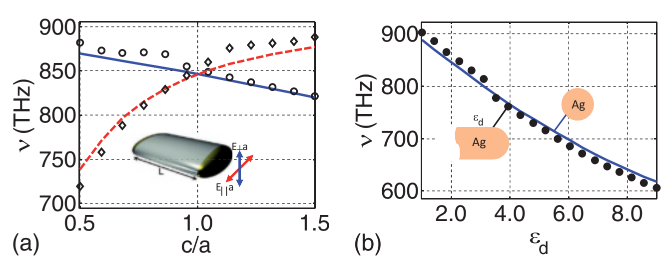

FIG. 5. (Color online) Resonance frequency in (a) of an ellipsoid with $b = c = 20$ nm surrounded by $\varepsilon_d = 1$ as a function of the ratio c/a between semi-axes and in (b) of a cylinder with a radius of 10 nm as a function of ε_d. In (a), solid blue (dashed red) curve corresponds to quasistatic resonance of the spheroid under illuminating polarization perpendicular (parallel) to semi-axis a. Circular (diamond) marks indicate frequencies at $\phi_r = \pi$ for polarization perpendicular (parallel) to semi-axis a. The polarization in the antenna simulation is selected by choosing respective wire mode as the illuminating field. In (b), the solid blue curve corresponds to quasistatic resonance while circular marks denote the frequencies at which $\phi_r = \pi$.

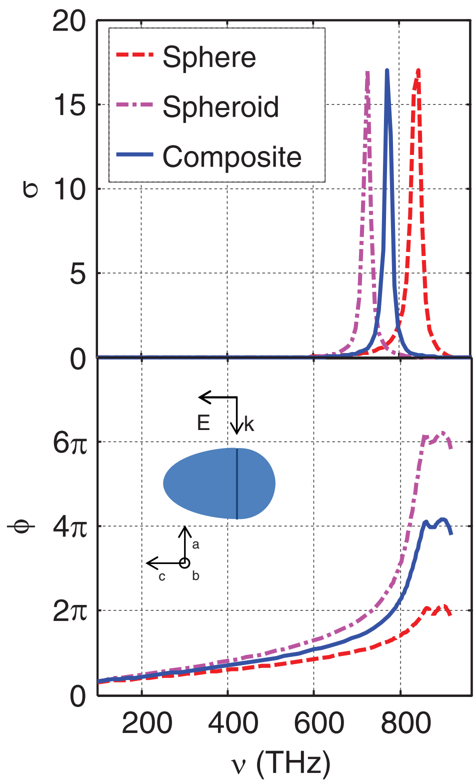

FIG. 6. (Color online) Normalized scattering cross section σ (top) and total phase jump ϕ (bottom) for an asymmetric composite particle (inset) made up of a hemisphere (radius = 10 nm) and a half prolate spheroid ($a = b = 10$ nm, $c = 20$ nm). Inset shows the 2D perspective of the proposed asymmetric composite particle.

C. 2D cylinder

The foregoing examples treated the case of 3D nanoparticles. Now we move on to a (quasi-) 2D object in the form of a cylinder extending to infinity along its axis. Such cylinders exhibit a dipolar resonance in the quasistatic limit under illumination with a plane wave having a magnetic field polarized along the cylinder's axis if $[\varepsilon_m(\nu) + \varepsilon_d(\nu) = 0]$. Technically, this is dealt with by considering a 2D insulator-metal-insulator (IMI) strip or nanowire having semicircular terminations [Fig. 1(d)]. The whole geometry then extends invariably out of the plane. The IMI strip waveguide is well known to support hybridized symmetric and antisymmetric PSPPs,[4] with the latter being strongly delocalized in the limit of a vanishing thickness (long-range surface plasmon polariton). The symmetric mode, on the contrary, localizes increasingly with decreasing thickness thereby standing out as the plausible source of LSPP resonance of cylinders in the quasistatic limit. Therefore, taking a 20-nm thick IMI strip terminated with semicircular caps of 20 nm diameter, we obtain the reflection coefficient for the symmetric plasmonic mode. Figure 5(b) shows the comparison of the theoretically predicted resonance frequencies with the standing wave resonance. Again, good agreement between the two approaches can be recognized.

IV. COMPOSITE PARTICLE

In the end, we present a practical application of the ideas established above by considering an asymmetric particle that cannot be divided into symmetric halves. Such a nanoparticle can be formed straightforwardly in Fig. 1(a) by replacing one of the two terminal caps by a half-spheroid whose semi-axes (a,b) perpendicular to the nanowire's axis are equal and the same as the nanowire's radius (inset of Fig. 6). The semi-axis (c) of the spheroid parallel to the axis of the nanowire is made different in length than the other two semi-axes to introduce asymmetry in the entire particle. In our simulation, we set the radius of the hemisphere as well as the semi-axes $a = b$ of the half-spheroid equal to 10 nm. The semi-axis c is finally set to 20 nm to form an egg-shaped particle. As there is no analytical formulation to predict a resonance in these particles, we rigorously compute the far-field scattering cross section by illuminating the particle with a plane wave having the **E** field polarized parallel to the c axis of the semispheroid (inset of Fig. 6). A first-order Fabry-Pérot resonance is obtained when the round-trip phase jump ϕ upon reflection from both hemispherical and semispheroidal ends becomes equal to 2π. As it is evident from Fig. 6, both methods are in excellent agreement.

V. CONCLUSION

In conclusion, we have proposed an alternate perspective for looking at the LSPP resonances of specific metallic nanoparticles and relate them to resonances of corresponding optical nanoantennas. We show that the LSPP resonances appear at frequencies where the total phase jump upon reflection of PSPPs from both terminals amounts to 2π. Numerical studies show that the resonance frequencies coincide with those obtained for LSPPs in the quasistatic approximation provided that this approximation is valid. Moreover, we foresee that further research devoted to the question of how to relate the reflection phase to measurable scattering quantities will be a fruitful direction.

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