Trapping light in open plasmonic nanostructures

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In open resonators the energy associated with a localized photonic excitation is lost in the form of a radiated wave, in the same manner that a classical charged particle in a curved orbit loses energy in the form of electromagnetic radiation. As a consequence, photonic modes in conventional spatially bounded open resonators have finite decay times. Here, we theoretically show that, surprisingly, in the limit of vanishing material loss, plasmons give the opportunity to have light localization in open spatially bounded systems with infinitely large lifetimes.

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I. INTRODUCTION

Light-wave oscillations in closed lossless cavities, e.g., a closed metallic box with perfectly conducting walls, can in theory take place forever because the photons are unable to escape the cavity. In contrast, in open systems the energy of an excitation continuously leaks away in the form of emitted light, and hence the lifetime is invariably finite. Likewise, charged particles moving in curved orbits emit electromagnetic radiation, and this is the origin of the Rutherford atom instability problem, solved by Bohr with his famous quantum postulates, as any mechanical foundation seemed hopeless [1]. Yet Larmor observed at the time that "when steady orbital motions in a molecule are so constituted that the vector sum of the accelerations of all its ions or electrons is constantly small, there will be no radiation, or very little, from it, and therefore this steady motion will be permanent" [1]. The quest for mechanisms that prevent radiation loss continues to this day, and some of the known solutions are based on Anderson localization [2], mirrors based on metals or photonic band gaps [3], total internal reflection (e.g., whispering gallery modes) [4], and structures that support bound states within the continuum [5,6]. This last solution relies on a proposal by von Neumann and Wigner who showed that in quantum mechanics certain potentials may support spatially localized states with energies above the potential barriers [5–7]. Recently these ideas were extended to the case of light waves [8–12], and exceedingly high Q factors have been experimentally measured. However, an oscillation with *exactly* infinite lifetime requires an *unbounded* structure (e.g., photonic crystals).

Here, we describe a strategy to trap light in a *bounded open* resonator surrounded by a vacuum such that the natural oscillations have infinite lifetime. This is evidently fundamentally and profoundly different from having embedded eigenvalues in an unbounded structure [5–12]. It is theoretically demonstrated that volume plasmons—i.e., charge-density waves in metals—are the key to having light localization with infinitely long lifetimes in open bounded systems.

It is emphasized that while infinite-lifetime states are relatively easy to achieve with unbounded structures, (e.g., in the form guided waves), up to now there is no known solution that permits—even in the ideal case wherein material loss vanishes—the trapping of light in an open optically transparent bounded resonator. Indeed, because of the coupling to the radiation continuum, light tends to escape from the resonator. For example, whispering gallery resonators can have remarkably high radiation quality factors that increase exponentially with the radius of the cavity [13]. However, the lifetime of the free oscillations, even though very long, is always finite. Another good example is the case of metallic spheres. As shown in Ref. [14], the resonance linewidth associated with the radiation loss of small metallic spheres decreases extremely sharply with an increase of the angular quantum number; however, it is never zero. Quite differently, here we prove that in the limit of no material loss volume plasmons offer the opportunity to have free oscillations of trapped light with exactly infinite lifetimes. We envision that the proposed open resonators can be potentially useful for light-emitting devices (e.g., the plasmonic nanolaser [15,16]), chemical or biological sensing, optical switching, and enhanced nonlinear effects [17].

II. OPEN PLASMONIC RESONATOR

An archetype of our open resonator is depicted in Fig. 1. It consists of a core-shell nanostructure in a vacuum, such that the inner region has radius R_1 and permittivity ε_1 and the cover shell has radius R_2 and permittivity ε_2. All the materials are assumed nonmagnetic ($\mu = \mu_0$). We look for a localized oscillation associated with a (real-valued) frequency ω such that the electromagnetic field is concentrated in the vicinity of the nanoparticle, and there is no energy leakage. Let $\mathbf{j}_p = \partial_t \mathbf{P} = -i\omega(\varepsilon - \varepsilon_0)\mathbf{E}$ be the polarization current density associated with the excitation. To have an oscillation with infinite lifetime the current $\mathbf{j}_p$ cannot radiate energy. This is possible only if the corresponding far field vanishes, which is tantamount to saying that the multipole moments of $\mathbf{j}_p$ of arbitrarily higher order must vanish. There is a general theorem [18] that establishes that if a source far field is zero then the corresponding radiated electromagnetic fields must vanish identically in the vacuum region outside the source. In our case this tells us that a hypothetical mode of oscillation with infinite lifetime has $(\mathbf{E},\mathbf{H}) = 0$ in the region $r > R_2$. In Appendix A, we further refine this result and prove that the lifetime of any localized excitations of a wide class of bounded

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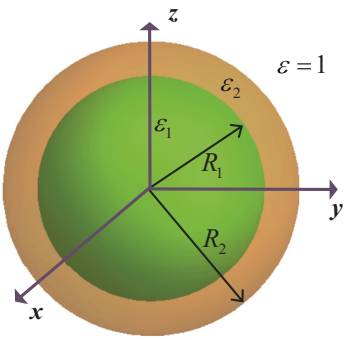

FIG. 1. (Color online) Geometry of the open bounded resonator formed by a core-shell nanoparticle. The cover shell is made of a low-permittivity (ENZ) material.

layered structures (e.g., a two-shell nanoparticle or truncated photonic crystals surrounded by a vacuum) made of regular materials with $\varepsilon \neq 0$ and $\mu \neq 0$ are necessarily *finite*.

One possibility to have electromagnetic fields identically zero when $r > R_2$ and an infinite lifetime is to have $\varepsilon_2 = -\infty$ so that the cover shell is perfectly conducting. This is not interesting to us because in such a case the resonator is opaque and inaccessible from the outside world. However, as described next, there is a startling, yet remarkably simple, opportunity to have open resonators with $Q = \infty$.

Suppose that the outer shell is made of a material with permittivity (epsilon) near zero (ENZ), such that $\varepsilon_2 = 0$ at some frequency $\omega = \omega_p$. ENZ materials are presently of great interest due to their remarkable potentials in supercoupling, tailoring wave fronts, and boosting the radiation from an emitter [19–21]. Moreover, core-shell nanoparticles with a plasmonic response have been considered previously in different contexts [22–25]. Let us assume first that all the materials are lossless because this is an obvious requirement to have undamped oscillations.

The electromagnetic field in spherical nanostructures can be conveniently written in terms of TE^r and TM^r vector spherical harmonics of the form [26,27], $\mathbf{M}_n^{\mathrm{TE}}(\mathbf{r}) = \nabla \times \{\mathbf{r} f_n^{\mathrm{TE}}(r) Y_n(\hat{\mathbf{r}})\}$ and $\mathbf{M}_n^{\mathrm{TM}} = \nabla \times \nabla \times \{\mathbf{r} f_n^{\mathrm{TM}}(r) Y_n(\hat{\mathbf{r}})/(ik)\}$, where $\mathbf{r} = x\hat{\mathbf{x}} + y\hat{\mathbf{y}} + z\hat{\mathbf{z}}$, Y_n is a generic spherical harmonic of order n, $k = \omega\sqrt{\varepsilon\mu}$ is the wave number, and f_n^p ($p =$ TE,TM) is a solution of the spherical Bessel equation $r^{-1}\partial_r^2[rf] + [k^2 - n(n+1)r^{-2}]f = 0$, i.e., a linear combination of spherical Bessel functions (j_n, y_n). Solutions such that $\mathbf{E} \sim \mathbf{M}_n^{\mathrm{TE}}$ are transverse electric (TE) with respect to the radial direction $\mathbf{E} \cdot \hat{\mathbf{r}} = 0$, and solutions with $\mathbf{E} \sim \mathbf{M}_n^{\mathrm{TM}}$ are transverse magnetic (TM), $\mathbf{H} \cdot \hat{\mathbf{r}} = 0$. Straightforward calculations show that the transverse wave impedance for TE^r and TM^r harmonics, defined such that $\hat{\mathbf{r}} \times \mathbf{E} = Z^p(\hat{\mathbf{r}} \times \mathbf{H}) \times \hat{\mathbf{r}}$ ($p =$ TE,TM), is given by

$$Z_n^{\mathrm{TE}} = i\omega\mu\, r_n^{\mathrm{TE}} \big/ \partial_r\left(r f_n^{\mathrm{TE}}\right) \quad \text{and}$$
$$Z_n^{\mathrm{TM}} = \partial_r\left(r f_n^{\mathrm{TM}}\right) \big/ \left[i\omega\varepsilon\, r f_n^{\mathrm{TM}}\right]. \tag{1}$$

This suggests that in the $\varepsilon \to 0$ limit the impedance of TM^r waves diverges, $Z_n^{\mathrm{TM}} \to \infty$, for any vector spherical harmonic ($n \geqslant 1$), whereas Z_n^{TE} remains finite. For example, if $f_n^p \sim j_n(kr)$, it can be easily checked that $\partial_r(rf_n^p)/(rf_n^p) \approx (n+1)/r$ for small r, which is independent of the material parameters. Thus, in the $\varepsilon \to 0$ limit the ENZ shell is generally seen as a perfect magnetic conductor (PMC) by the TM^r waves, whereas it is penetrable by the TE^r waves. Hence, despite the fact the cavity is electromagnetically open, it may support a wave oscillation with $Q = \infty$.

The electric field associated with a TM$_n^r$ wave is such that $\mathbf{E} \sim \mathbf{M}_n^{\mathrm{TM}}$. For convenience let us denote $\tilde{f}_n^{\mathrm{TM}} = f_n^{\mathrm{TM}} k_0/k$ with $k_0 = \omega/c$. The continuity of the tangential electromagnetic field at an interface requires that $\varepsilon \tilde{f}_n^{\mathrm{TM}}$ and $\partial_r[r\tilde{f}_n^{\mathrm{TM}}]$ are continuous functions of r. In the limit $\varepsilon \to 0$ ($\omega = \omega_p$) the fields in the open resonator must satisfy

$$\tilde{f}_n^{\mathrm{TM}} = E_0 \times \begin{cases} j_n(k_1 r), & r < R_1, \\ A r^n + B r^{-n-1}, & R_1 < r < R_2, \\ 0, & r > R_2, \end{cases} \tag{2}$$

where E_0 is a normalization constant, $k_1 = \omega\sqrt{\mu_0\varepsilon_1}$, and A and B are constants chosen to ensure that $\partial_r[r\tilde{f}_n^{\mathrm{TM}}]$ is continuous:

$$B = A R_2^{2n+1}(n+1)/n \quad \text{with}$$
$$A = \frac{1}{n+1}\left[r j_n(k_1 r)\right]'_{r=R_1}\left(R_1^n - R_2^{2n+1}/R_1^{n+1}\right)^{-1}. \tag{3}$$

On the other hand, the continuity of $\varepsilon\tilde{f}_n^{\mathrm{TM}}$ requires that at $\omega = \omega_p$

$$j_n(k_1 R_1) = 0. \tag{4}$$

Remarkably, Eq. (4) is coincident with the characteristic equation for the TM$_n^r$ modes of a fictitious resonator obtained by replacing the ENZ region by a PMC wall. Thus, an electromagnetic wave can become trapped inside the nanoparticle core provided the frequency $\omega = \omega_p$ is *exactly coincident* with a resonant frequency of the equivalent PMC cavity. This strict condition is satisfied only for very specific values of the nanoparticle inner radius (R_1). For example, for waves associated with $n = 1$ the first zero of $j_1(u)$ occurs at $u = 4.49$, and thus one possibility to have a natural mode at $\omega = \omega_p$ is that $R_1 = R_{1,0} \equiv 4.49/(\omega_p\sqrt{\varepsilon_1\mu_0})$. The modes of oscillation are degenerate because there are a total of $2n+1$ independent spherical harmonics of order n.

The previous analysis confirms that the ENZ shield can effectively prevent the radiation from the nanoparticle core in the $\varepsilon \to 0$ limit. Interestingly, the magnetic field in the ENZ cover vanishes, $\mathbf{H} = \mathbf{0}$, whereas the electric field is inherently quasistatic such that $\nabla \times \mathbf{E} = \mathbf{0}$, but it is a transverse field ($\nabla \cdot \mathbf{E} = 0$). Figure 2 depicts the electromagnetic fields in the open resonator for the $n = 1$ case with $Y_1 = \cos\varphi\sin\theta$ [(r,θ,φ) is a system of spherical coordinates attached to the center of the nanoparticle], $R_1 = R_{1,0}$, and $R_2 = 1.1R_{1,0}$, supposing that the inner core is a vacuum ($\varepsilon_1 = \varepsilon_0$). The electric and magnetic fields are in quadrature, ensuring that the net power flow is zero, and E_x is the only nonzero field component at the origin. Figure 2 shows that the radial electric field is greatly enhanced in the ENZ shell, and confirms that the fields in the exterior region vanish. The strong radial electric field is due to the excitation of volume-plasmon-type oscillations. Usually the notion of a perfect conductor implies an infinitely large

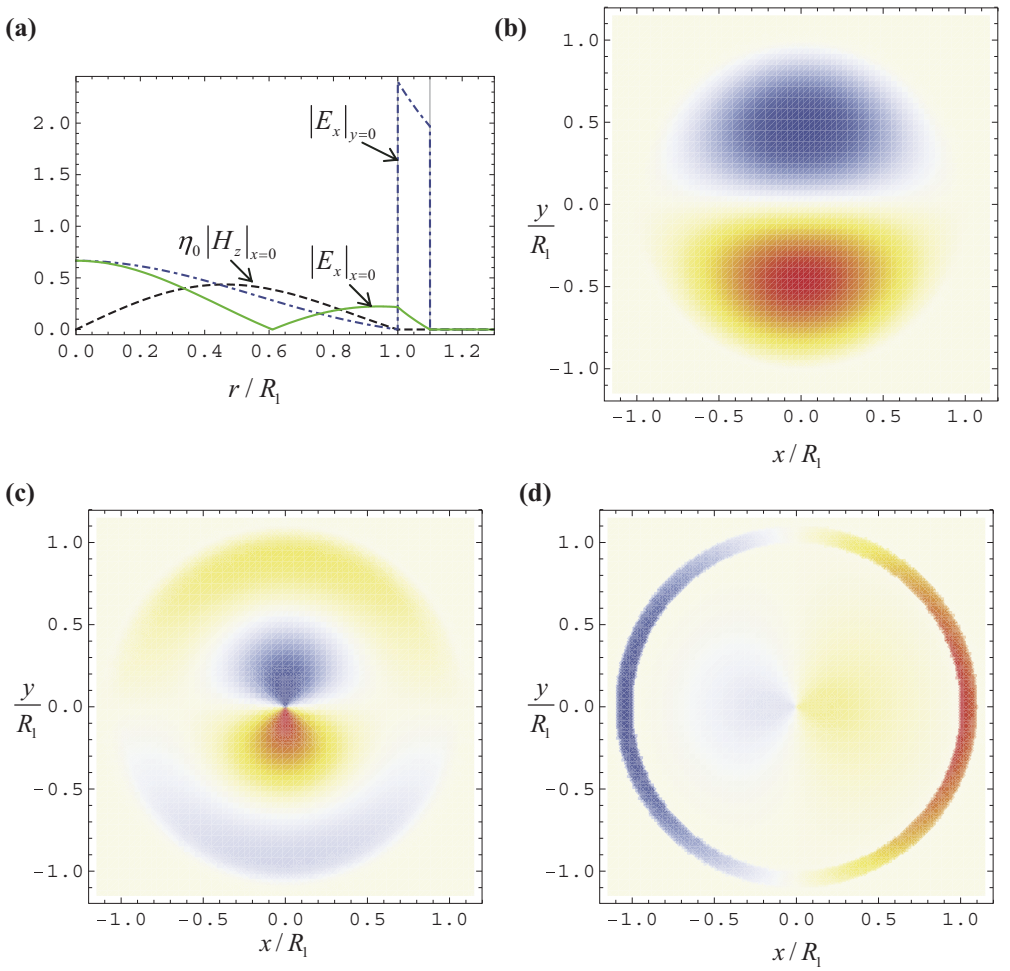

FIG. 2. (Color online) Optical field distributions associated with the natural mode of oscillation with infinite lifetime for $R_1 = R_{1,0}$, $R_2 = 1.1R_{1,0}$, $\varepsilon_1 = 1$, $\varepsilon_2 = 0$, and $n = 1$. (a) Amplitudes of the electromagnetic fields in the resonator (normalized to arbitrary units) as a function of the radial distance in the *xoy* plane. (b),(c),(d) Time snapshot of $H_z(t = T/4)$, $E_\varphi(t = 0)$, and $E_r(t = 0)$ in the *xoy* plane, with $T = 2\pi/\omega$ the period of oscillation. Bluish (reddish) [Darker (lighter) gray] colors represent positive (negative) excursions of the fields.

complex refractive index, and thus an opaque material. Here, the ENZ shell behaves as a perfect magnetic conductor, but intriguingly it is penetrable by the electric field. The reason is that unlike the "usual" perfect conductors the ENZ material has a vanishing refractive index. We note that the fields inside the core region are quite distinct from whispering-gallery modes [4], which typically are localized near the interface and are associated with spherical harmonics with an angular number $n \gg 1$. Moreover, whispering-gallery modes require resonators with diameters significantly larger than the light wavelength to reduce the radiation loss and achieve high (but—even in the absence of material loss—finite) Q factors. Quite differently, the diameter of our resonator is of the order of the wavelength.

III. EFFECT OF MATERIAL LOSS AND OF A NONLOCAL RESPONSE

From the Kramers-Kronig relations any linear and causal material is necessarily lossy [27]. Even though the Kramers-Kronig relations do not forbid that ε can be precisely zero at some isolated fixed frequency of operation, for any realistic plasmonic material the imaginary part of the permittivity is greater than zero.

In the presence of material loss, the decay time becomes finite and the corresponding oscillation frequency is complex valued, $\omega = \omega' + i\lambda$, with $\lambda < 0$. The quality factor of the resonator is $Q = \omega' \times$ (stored energy)/(dissipated power) and can be written as $Q = \omega'/(-2\lambda)$. Thus, it is proportional to the ratio between the lifetime $\tau = 1/(-2\lambda)$ and the period of oscillation $T = 2\pi/\omega$, such that $Q/(2\pi) = \tau/T$.

To find $\omega = \omega' + i\lambda$, we note that for natural modes the fields in the resonator are generally determined by

$$\tilde{f}_n^{\mathrm{TM}} = \begin{cases} a^{\mathrm{TM}} j_n(k_1 r), & r < R_1, \\ b_1^{\mathrm{TM}} j_n(k_2 r) + b_2^{\mathrm{TM}} y_n(k_2 r), & R_1 < r < R_2, \\ c^{\mathrm{TM}} h_n^{(1)}(k_0 r), & r > R_2. \end{cases} \quad (5)$$

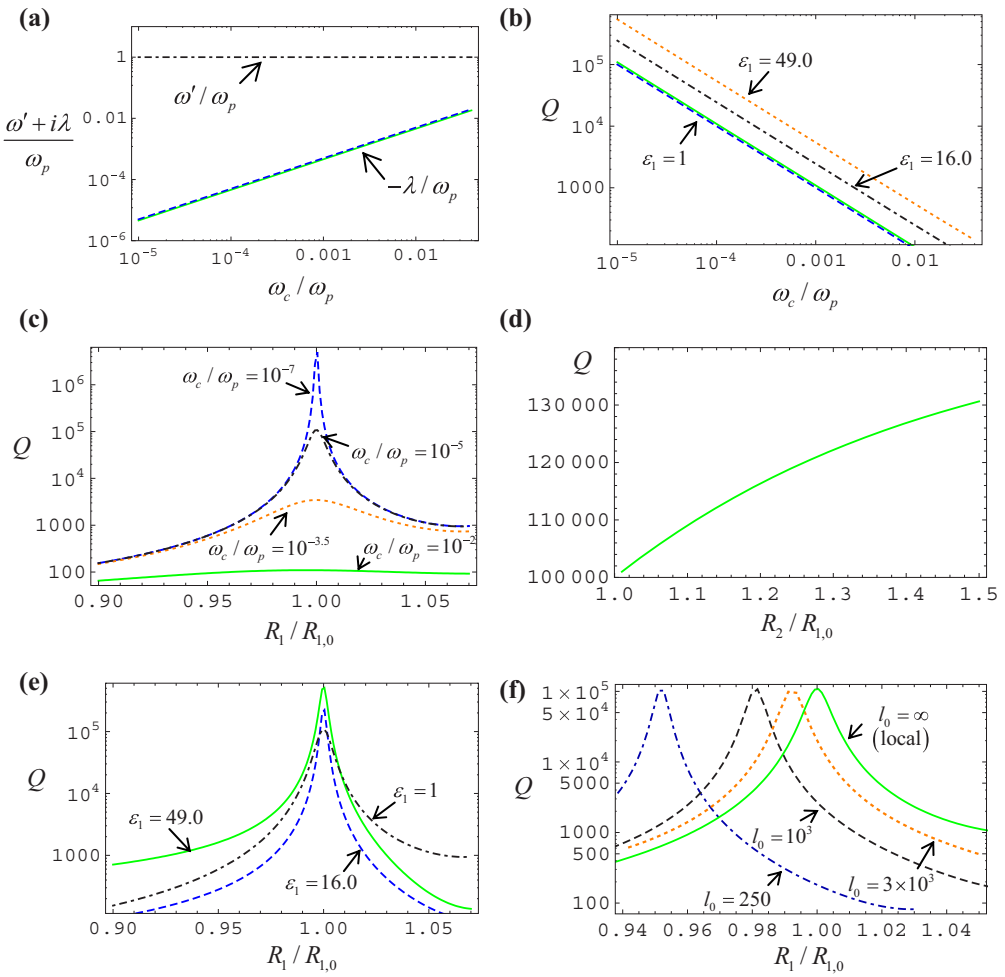

FIG. 3. (Color online) Natural oscillation frequency and quality factor of the open resonator. (a) Characteristic (complex) oscillation frequency for the case $\varepsilon_1 = 1$ and (b) quality factor, as a function of the normalized collision frequency of the ENZ cover material. The blue dashed line in (a) and (b) represents the curves $\lambda = -\omega_c/2$ and $Q = \omega_p/\omega_c$, respectively. The radii of the core and cover shells are $R_1 = R_{1,0}$ and $R_2 = 1.1R_{1,0}$. (c) Quality factor as a function of the air cavity radius for several ω_c/ω_p and $R_2 = 1.1R_{1,0}$. (d) Quality factor as a function of the ENZ cover radius for $\omega_c/\omega_p = 10^{-5}$ and $R_1 = R_{1,0}$. (e) Quality factor as a function of the core radius for different values of the core permittivity and $\omega_c/\omega_p = 10^{-5}$ and $R_2 = 1.1R_{1,0}$. (f) Effect of spatial dispersion in the ENZ cover for $\omega_c/\omega_p = 10^{-5}$, $R_2 = 1.1R_{1,0}$, and $\varepsilon_1 = 1.0$. Lower values of l_0 are associated with stronger nonlocal effects.

The coefficients a^{TM}, b_1^{TM}, b_2^{TM}, and c^{TM} must be such that $\varepsilon \tilde{f}_n^{\mathrm{TM}}$ and $\partial_r[r\tilde{f}_n^{\mathrm{TM}}]$ are continuous functions of r. This yields a homogeneous linear system,

$$\begin{pmatrix} -[j_n(k_1 r)\, r]'_{r=R_1} & [j_n(k_2 r)\, r]'_{r=R_1} & [y_n(k_2 r)\, r]'_{r=R_1} & 0 \\ -\varepsilon_1 j_n(k_1 R_1) & \varepsilon_2 j_n(k_2 R_1) & \varepsilon_2 y_n(k_2 R_1) & 0 \\ 0 & [j_n(k_2 r)\, r]'_{r=R_2} & [y_n(k_2 r)\, r]'_{r=R_2} & -\left[h_n^{(1)}(k_0 r)\, r\right]'_{r=R_2} \\ 0 & \varepsilon_2 j_n(k_2 R_2) & \varepsilon_2 y_n(k_2 R_2) & -\varepsilon_0 h_n^{(1)}(k_0 R_2) \end{pmatrix} \begin{pmatrix} a^{\mathrm{TM}} \\ b_1^{\mathrm{TM}} \\ b_2^{\mathrm{TM}} \\ c^{\mathrm{TM}} \end{pmatrix} = 0, \tag{6}$$

which has a nontrivial solution only when $\omega = \omega' + i\lambda$ is such that the determinant of the matrix vanishes. In Fig. 3(a) we plot the calculated oscillation frequency (mode with $n = 1$) as a function of the ENZ material loss for an air cavity. The cover material dispersion is described by a Drude model with plasma frequency ω_p and collision frequency ω_c. As seen, ω' is little affected by the loss and satisfies $\omega' \approx \omega_p$, whereas λ is roughly proportional to ω_c, such that when $\omega_c/\omega_p \to 0$ the lifetime is exactly infinite ($\lambda \to 0$). Our numerical calculations show that to an excellent approximation $\lambda \approx -\omega_c/2$ when $\varepsilon_1 = \varepsilon_0$. Thus, the quality factor of the open resonator is $Q \approx \omega_p/\omega_c$ [Fig. 3(b)]. Interestingly, this is the same as the quality factor of the volume plasmons supported by the ENZ material [solutions of $\varepsilon(\omega) = 0$]. Hence, the lifetime of the natural mode is determined by the volume plasmons, proving that the wave oscillation results from the hybridization of volume plasmons and the optical field in the air cavity. It is interesting to mention that the characteristic lifetimes of surface plasmon polaritons

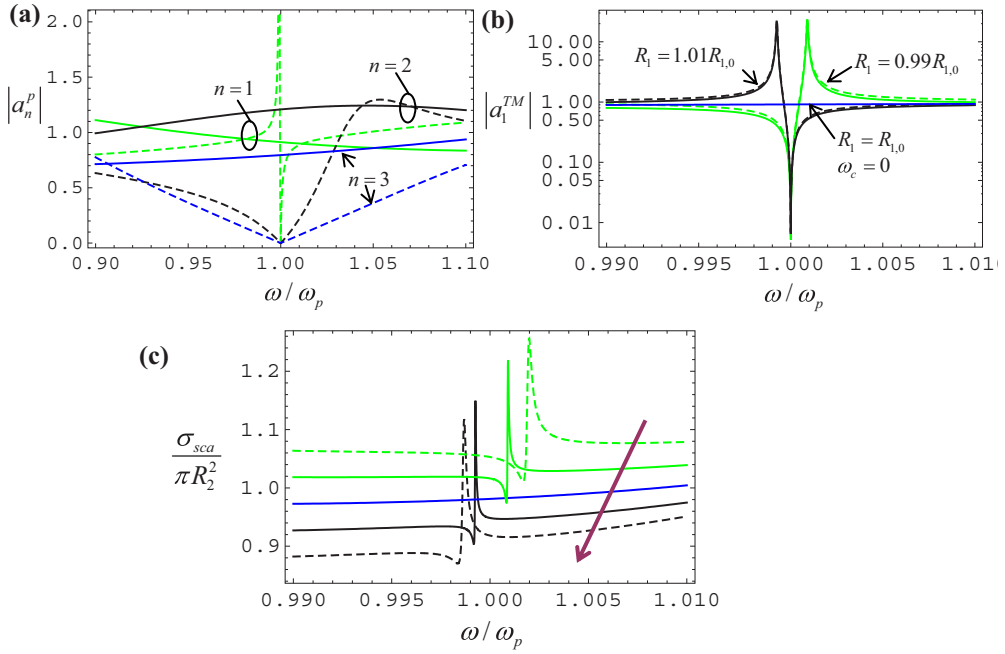

FIG. 4. (Color online) Mie coefficients in the core region and scattering cross section when the open bounded resonator is illuminated by a plane wave. (a) Mie coefficients in the core region for p = TE (solid lines) and p = TM (dashed lines) waves assuming a lossless Drude dispersion ($\omega_c/\omega_p = 0$) and $R_1 = 1.01R_{1,0}$. (b) Mie coefficient a_1^{TM} in the core region. The solid and dashed lines represent the exact and the approximate analytical result [Eq. (7)], respectively, and are almost coincident. The collision frequency is taken as $\omega_c = 10^{-5}\omega_p$, except in the case $R_1 = R_{1,0}$ where it vanishes. (c) Scattering cross section σ_{sca} as a function of frequency for $R_1/R_{1,0}$ =0.98, 0.99, 1.00, 1.01, and 1.02. The arrow indicates the direction of increasing R_1. In all the panels, the cover radius is $R_2 = 1.1R_{1,0}$ and the core region is a vacuum ($\varepsilon_1 = 1.0$).

in metal nanoparticles can also be fairly large due to their intrinsic quasistatic nature. However, as further discussed in Appendix B and in Ref. [14], even in the absence of material loss they have necessarily a finite decay time due to the radiation loss associated with high-order multipoles.

When the core is made of a dielectric [Fig. 3(b)], the quality factor can be enhanced because of the impedance mismatch between the vacuum region and the cavity. Figures 3(c) and 3(e) show the effect of detuning the cover radius for different values of the material loss and core permittivity. As R_1 is changed, so that ω_p is no longer a resonant frequency of the equivalent PMC cavity, the quality factor decreases and the excitation lifetime becomes finite. On the other hand, when the inner core is a vacuum the quality factor is nearly independent of the ENZ shell thickness [Fig. 3(d)]. In general, thinner ENZ shells have lower quality factors. The reason is that the electric field is enhanced in the ENZ material as the shell is made thinner, implying a higher sensitivity of thin ENZ covers to material loss. Interestingly, a spherical dielectric resonator with no ENZ cover, $\varepsilon_1 = 49\varepsilon_0$ and radius $R_1 = R_{1,0} \equiv 4.49/(\omega_p\sqrt{\varepsilon_1\mu_0})$ supports a $\mathrm{TM}^r_{n=1}$ natural mode of oscillation with $\omega' \approx \omega_p$ and quality factor $Q = 79.9$. The quality factor of the same resonator is increased to $Q = 5.4 \times 10^5$ when an ENZ shell with $\omega_c/\omega_p = 10^{-5}$ is used to shield the dielectric [Fig. 3(e)]. Thus, the ENZ cover effectively traps the radiation in the core region.

Since the operation of the open resonator is critically dependent on the ENZ response of the cover material, it is pertinent to study how spatial dispersion effects may affect the lifetime of the localized oscillation. The nonlocal response of a metal is a consequence of the quantum kinetics of the electron gas and can be conveniently modeled by adding a diffusion term to Ohm's law [28,29]. Within this approach, the frequency dispersion of the volume plasmons is $\omega^2 = \omega_p^2 + c^2k^2/l_0$ where k is the wave number and $c^2/l_0 = 3v_F^2/5$ with v_F the Fermi velocity. For example, for silver $v_F \approx c/200$ and hence we have $l_0 = 6.7 \times 10^4$ [30]. In Fig. 3(f) Q is plotted for different values of the l_0 parameter that characterizes the nonlocal effects in the ENZ cover. The details of the calculations are given in Appendix C. As seen, the nonlocal effects do not change the peak value of Q but may shift slightly the optimal value of R_1.

IV. EXCITATION OF THE OPEN RESONATOR BY A PLANE WAVE

Consider now the scenario wherein the nanostructure is illuminated by a linearly polarized plane wave. In Fig. 4(a) we plot the Mie coefficients a_n^p (p = TE,TM) for $R_1 = 1.01R_{1,0}$. The coefficients a_n^p determine the electromagnetic fields in the core region, and their exact definition is given in Appendix D. It can be seen that a_n^{TM} vanishes at $\omega = \omega_p$, confirming that the ENZ shell behaves as a PMC wall for TM^r vector spherical harmonics, but that a_n^{TE} are nonzero, clearly demonstrating that our resonator is electromagnetically open. The coefficient a_1^{TM} is peaked at a frequency near ω_p, which corresponds to the resonance of the inner cavity. We verified that when $\varepsilon_1 \approx \varepsilon_0$, $\varepsilon_2 \approx 0$, and $R_1 \approx R_{1,0}$, the Mie coefficient a_1^{TM} satisfies to a good approximation

$$a_1^{\mathrm{TM}} \approx e^{i\phi_0}\frac{(\omega-\omega_L)(\omega+\omega_L^*)}{(\omega-\omega_r)(\omega+\omega_r^*)} \quad \text{when} \quad \omega \approx \omega_p, \qquad (7)$$

where ϕ_0 is some phase factor, $\omega_r = \omega' + i\lambda$ is the complex frequency of the natural mode of the open resonator [obtained by solving the characteristic equation discussed below Eq. (6)], and ω_L is the complex frequency associated with the volume-plasmon free oscillations, $\varepsilon(\omega_L) = 0$. The accuracy of this formula is demonstrated in Fig. 4(b). It is seen that the resonances have Fano-type line shapes [17]. Curiously, when $\omega_c \to 0$ and $R_1 \to R_{1,0}$ the zero associated with ω_L and the resonance associated with ω_r merge so that $|a_1^{\mathrm{TM}}| \to 1$. Thus, the emergence of the infinite-lifetime state corresponds to a pole-zero cancellation in the Mie coefficient a_1^{TM}. This implies that, surprisingly, in the limit $\omega_c \to 0$ the incoming wave can be coupled to the $\mathrm{TM}^r_{n=1}$ mode of the resonator. In Appendix D the time dynamics of this effect is studied in detail for a pulse of finite duration. It is found that the fields pushed into the core by the incident wave do not remain stored in the resonator after the source is switched off (i.e., once the incoming pulse overtakes the nanostructure, the fields associated with the $\mathrm{TM}^r_{n=1}$ mode in the nanostructure quickly fade away), and thus, as expected the resonator cannot be directly pumped from the outside. Consistent with this, it is seen in Fig. 4(c) that the scattering cross section of the open resonator is always finite under the plane-wave excitation. Moreover, when $R_1 = R_{1,0}$ (blue curve) the scattering cross section does not exhibit any resonant features due to the previously discussed pole-zero cancellation.

The property $|a_1^{\mathrm{TM}}| \to 1$ may look at first sight inconsistent with the reciprocity principle. However, when $\omega_c = 0$ the open resonator may support a natural mode with infinite lifetime, and hence if a time-harmonic source is placed inside the resonator the fields may grow without limit and a steady state is impossible to reach. In other words, the reciprocity theorem is based on the assumption that for a given source there is a unique steady-state solution of the Maxwell equations, which is not the case when $Q = \infty$. Thus, $|a_1^{\mathrm{TM}}| \to 1$ is not inconsistent with reciprocity, simply because the reciprocity theorem breaks down in this limit (see Appendix D).

V. CONCLUSION

In summary, volume plasmons in $\varepsilon = 0$ materials can effectively shield the electromagnetic field in an open bounded cavity. The circulating currents are blocked from radiating because the ENZ shield behaves as a PMC for the TM^r waves. The open resonator is electromagnetically transparent, but it cannot be directly pumped by an external source. However, it can be pumped either by an internal source or by using an electron beam to originate emission via processes such as the Smith-Purcell and Cherenkov effects. Moreover, a nonlinear response of the core material may permit the resonator to be pumped from the outside, and may help to alleviate the strict requirement $R_1 = R_{1,0}$ and enable self-sustained oscillations for specific values of the stored field energy. Even though the conditions required to have $Q = \infty$ are physically demanding (e.g., $\varepsilon = 0$ is required while $\varepsilon/\varepsilon_0 \sim 0.27i$ at $\omega = \omega_p$ for silver in the UV; the damping is smaller in alkali metals such as K, which has $\omega_c/\omega_p \sim 0.05$ [31]) our theory gives hints on how to have open resonators with higher quality factors, and crucially shows that even within a classical framework it is possible to have self-sustained circulating currents with no energy leakage. Active (gain) plasmonic materials may provide a route to achieve ε precisely zero and compensate for the effects of material loss.

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APPENDIX A: THEOREM ON THE LIFETIMES OF THE NATURAL MODES OF LAYERED DIELECTRIC STRUCTURES

The following theorem is demonstrated in Ref. [18, p.165]:

Theorem. Assume that the bounded domain D is the open complement of an unbounded domain, and let $(\mathbf{E},\mathbf{H})$ be a radiating smooth solution of the Maxwell equations in $\mathbb{R}^3/\overline{D}$ for which the electric far-field pattern vanishes identically. Then $(\mathbf{E},\mathbf{H}) = 0$ in $\mathbb{R}^3/\overline{D}$.

This result assumes that the region $\mathbb{R}^3/\overline{D}$ is uniform ($\varepsilon =$ const and $\mu =$ const) and that the wave number $k = \omega\sqrt{\mu\varepsilon}$ is real valued and nonzero (e.g., the region $\mathbb{R}^3/\overline{D}$ can be a vacuum).

In order to further generalize the theorem, we consider the generic open resonator depicted in Fig. 5. It consists of a bounded layered structure formed by conventional isotropic materials with $\varepsilon \neq 0$ and $\mu \neq 0$ (the material parameters may, however, be either negative or complex valued) and surrounded by a vacuum.

Suppose that the open resonator supports a natural mode of oscillation with infinite lifetime. As discussed in the main text, this implies that the far-field vanishes and thus from the theorem, it follows that $(\mathbf{E},\mathbf{H}) = 0$ in the vacuum region (leftmost panel in Fig. 5). Applying the Stratton-Chu formulas [18, p. 158] to the ε_1,μ_1 material domain (taking as the boundaries of the integral representation the boundary surface with the ε_2,μ_2 material, ∂D_2, and the boundary surface with the vacuum, ∂D_1), we find that

$$\mathbf{E}(\mathbf{r}) = -\nabla \times \int_{\partial D_1 \cup \partial D_2} \hat{\mathbf{n}}' \times \mathbf{E}(\mathbf{r}')\Phi_0(\mathbf{r}-\mathbf{r}')d^3\mathbf{r}' + \frac{1}{i\omega\varepsilon}\nabla \times \nabla \times \int_{\partial D_1 \cup \partial D_2} \hat{\mathbf{n}}' \times \mathbf{H}(\mathbf{r}')\Phi_0(\mathbf{r}-\mathbf{r}')d^3\mathbf{r}', \tag{A1}$$

where $\hat{\mathbf{n}}'$ is the unit normal vector directed into the exterior of the ε_1,μ_1 domain, and $\Phi_0 = e^{ikr}/(4\pi r)$. Because $(\mathbf{E},\mathbf{H}) = 0$

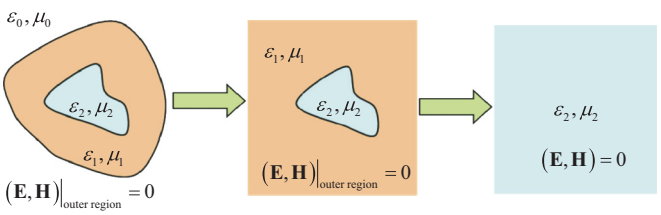

FIG. 5. (Color online) Sketch of the proof that a bounded layered structure formed by conventional isotropic materials with $\varepsilon \neq 0$ and $\mu \neq 0$ standing in a vacuum cannot support localized oscillations with infinite lifetimes.

in the vacuum region, the tangential components of $(\mathbf{E},\mathbf{H})$ vanish at the boundary with the vacuum (∂D_1). Hence, it follows that the electromagnetic fields in the ε_1,μ_1 region can be written as a surface integral of equivalent sources defined only over the ε_2,μ_2 material boundary (∂D_2). It is well known that the integral in the right-hand side of Eq. (A1) always evaluates to zero outside the ε_1,μ_1 domain [18, p. 158]. Thus, because the integration domain does not include ∂D_1, it follows that the right-hand side of Eq. (A1) defines a smooth (analytic) solution of Maxwell equations in the region outside ∂D_2 that vanishes identically in the region outside ∂D_1. But because of the analyticity of the electric field [18, p. 159], we conclude that $\mathbf{E}$ must identically vanish in the entire ε_1,μ_1 domain. The same arguments (or the Maxwell equations) can be used to prove that $\mathbf{H}$ also vanishes.

Now, it is possible to replace the vacuum region by the material with parameters ε_1,μ_1 without perturbing the original electromagnetic fields (middle panel in Fig. 5). The previous discussion shows that the electromagnetic field vanishes in the extended ε_1,μ_1 region. Thus, repeating the previous arguments we can prove that for layered structures $(\mathbf{E},\mathbf{H})$ vanishes in all space (rightmost panel in Fig. 5), and thus this class of open resonators cannot support oscillations with infinite lifetimes. This result applies to any open layered resonator formed by conventional isotropic materials with $\varepsilon \neq 0$ and $\mu \neq 0$, and in particular to any truncated photonic crystal whose basic unit cell is a layered structure analogous to the one considered here.

The reason why our theorem does not hold in the presence of materials with $\varepsilon = 0$ can be understood by inspection of Eq. (A1). As seen, because of the term $1/(i\omega\varepsilon)$ the Stratton-Chu representation of the electric field becomes ill defined. The singularity can be lifted by expanding the $\nabla \times \nabla \times$ operator, which gives:

$$\begin{aligned}\mathbf{E}(\mathbf{r}) = &-\nabla \times \int_{\partial D_1 \cup \partial D_2} \hat{\mathbf{n}}' \times \mathbf{E}(\mathbf{r}')\Phi_0(\mathbf{r}-\mathbf{r}')d^3\mathbf{r}' \\ &- i\omega\mu \int_{\partial D_1 \cup \partial D_2} \hat{\mathbf{n}}' \times \mathbf{H}(\mathbf{r}')\,\Phi_0(\mathbf{r}-\mathbf{r}')d^3\mathbf{r}' \\ &+ \nabla \int_{\partial D_1 \cup \partial D_2} \frac{1}{i\omega\varepsilon}\mathrm{Div}[\hat{\mathbf{n}}' \times \mathbf{H}(\mathbf{r}')]\,\Phi_0(\mathbf{r}-\mathbf{r}')d^3\mathbf{r}'. \end{aligned} \tag{A2}$$

The third integral in the right-hand side depends on $\mathrm{Div}[\hat{\mathbf{n}} \times \mathbf{H}]/i\omega\varepsilon$. At an interface $\mathrm{Div}(\hat{\mathbf{n}} \times \mathbf{H}) = -\hat{\mathbf{n}} \cdot \nabla \times \mathbf{H} = i\omega\varepsilon\hat{\mathbf{n}} \cdot \mathbf{E}$, with $\hat{\mathbf{n}}$ the unit normal vector to the interface, and thus $\mathrm{Div}[\hat{\mathbf{n}} \times \mathbf{H}]/i\omega\varepsilon = \hat{\mathbf{n}} \cdot \mathbf{E}$. The key point is that having $\hat{\mathbf{n}} \times \mathbf{H} = \mathbf{0}$ at the interface is insufficient to conclude that $\hat{\mathbf{n}} \cdot \mathbf{E} = 0$ when $\varepsilon = 0$. Thus, when $\varepsilon = 0$ the third term in the right-hand side of Eq. (A2) does not vanish, and hence the integral over ∂D_1 cannot be dropped and the theorem does not apply.

APPENDIX B: LIFETIMES OF THE FREE OSCILLATIONS OF METALLIC NANOPARTICLES

It is interesting to have an idea of the characteristic lifetimes of the natural modes of resonators made of metallic nanostructures. To this end, we consider a metallic nanoparticle with radius R surrounded by a vacuum (see the inset of Fig. 6). For simplicity, the geometry is two dimensional so that the structure is invariant to translations along the z direction. It is assumed that the nanoparticle is described by a lossless Drude model $\varepsilon_m/\varepsilon_0 = 1 - 2\omega_{SPP}^2/\omega^2$, where ω_{SPP} corresponds to the surface plasmon resonance of a cylindrical structure $[\varepsilon_m(\omega_{SPP})/\varepsilon_0 = -1]$. The characteristic equation of the natural modes of oscillation (with magnetic field along the z direction) can be derived using methods similar to those reported in Sec. III and is

$$\frac{1}{\varepsilon_m}\frac{k_m J_n'(k_m R)}{J_n(k_m R)} = \frac{1}{\varepsilon_0}\frac{k_0 H_n^{(1)\prime}(k_0 R)}{H_n^{(1)}(k_0 R)}, \tag{B1}$$

where n determines the azimuthal variation of the fields $(e^{in\phi})$, $k_0 = \omega/c$, $k_m = \omega\sqrt{\varepsilon_m\mu_0}$, and J_n and $H_n^{(1)}$ denote the cylindrical Bessel and Hankel functions. Figure 6 shows the numerically calculated frequency of oscillation $\omega = \omega' + i\lambda$ for the surface plasmon supported by the cylindrical nanoparticle with $n = 1$ as a function of the normalized radius. As seen, even though the system is lossless, the imaginary part of ω is always nonzero and thus the lifetime is always finite. The lifetime is larger for smaller nanoparticles, but the physical size cannot be arbitrarily small and this imposes a bound on Q. For example, for silver we may estimate $\omega_{SPP} = 2\pi \times 914/\sqrt{2}$ THz, [32] and thus for $R = 24$ nm we have $\omega_{SPP}R/c = 0.32$ which limits Q to 13.4.

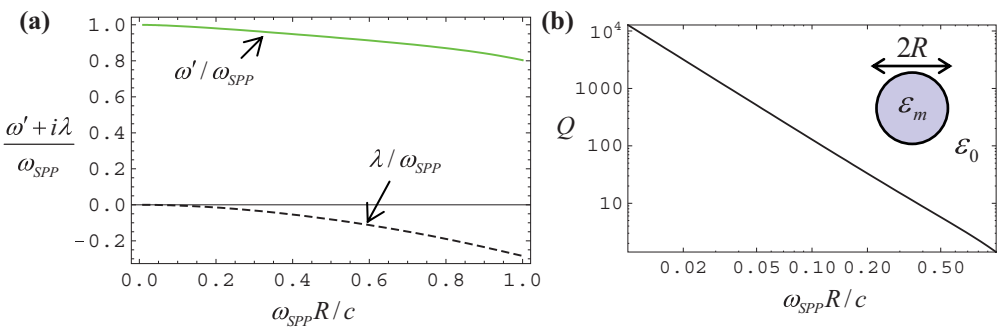

FIG. 6. (Color online) (a) Characteristic (complex) oscillation frequency of the surface plasmon supported by a cylindrical nanoparticle as a function of the normalized radius R. (b) Quality factor as a function of the normalized radius R.

APPENDIX C: NATURAL MODES OF OSCILLATION OF A CORE-SHELL NANOPARTICLE WITH A NONLOCAL PERMITTIVITY RESPONSE

Here, we study the effects of spatial dispersion in the ENZ cover on the lifetime of the natural mode supported by the open resonator. The methods described next follow closely Refs. [29,30,33]. Our model is based on the assumption that in a metal the electron gas can be modeled by an electric current density $\mathbf{J}_c$ that satisfies a drift-diffusion equation:

$$\left(1+\frac{D}{i\omega}\nabla\nabla\right)\cdot\mathbf{J}_c=\sigma\mathbf{E}. \tag{C1}$$

Here, σ is the electric conductivity and D is the diffusion coefficient. The Maxwell's equations are

$$\nabla\times\mathbf{E}=i\omega\mu\mathbf{H},\quad \nabla\times\mathbf{H}=\mathbf{J}_c-i\omega\varepsilon_b\mathbf{E}, \tag{C2}$$

where ε_b represents the dielectric response due to the bound charges of the material. It is convenient to introduce the auxiliary function $\mathbf{F}=i\omega\mu\mathbf{J}_c$. The coupled equations (C1) and (C2) support transverse and longitudinal solutions.

The transverse solutions are such that $\nabla\cdot\mathbf{E}=0$ and $\mathbf{F}=i\omega\mu\sigma\mathbf{E}$ and hence satisfy:

$$\nabla\times\nabla\times\mathbf{E}=k_t^2\mathbf{E},\quad k_t^2=k_b^2+i\omega\mu\sigma,\quad \nabla\cdot\mathbf{E}=0, \tag{C3a}$$

$$\mathbf{F}=i\omega\mu\sigma\mathbf{E},\quad \mathbf{H}=\frac{1}{i\omega\mu}\nabla\times\mathbf{E} \tag{C3b}$$

where $k_b^2=\omega^2\mu\varepsilon_b$. For future reference we define a transverse permittivity ε_t such that $\varepsilon_t=k_t^2/(\omega^2\mu)=\varepsilon_b+\sigma/(-i\omega)$.

On the other hand, the longitudinal solutions are such that $\mathbf{E}=\nabla\phi$ and $\mathbf{F}=-k_b^2\mathbf{E}$, for some scalar potential ϕ. Thus, from Eq. (C1) we have $(1+\frac{D}{i\omega}\nabla\nabla)\cdot\nabla\phi=\frac{1}{-k_b^2}i\omega\mu\sigma\nabla\phi$, which is equivalent to $\phi+\frac{D}{i\omega}\nabla^2\phi=\frac{1}{-k_b^2}i\omega\mu\sigma\phi$. Therefore:

$$\nabla^2\phi+k_L^2\phi=0,\quad k_L^2=-\frac{1}{D}\left(\frac{\sigma}{\varepsilon_b}-i\omega\right), \tag{C4a}$$

$$\mathbf{E}=\nabla\phi,\quad \mathbf{F}=-k_b^2\mathbf{E},\quad \mathbf{H}=0. \tag{C4b}$$

The wave number $k_D=i\,k_L$ is known as the Debye wave number. In this work, we assume that $\varepsilon_b=\varepsilon_0$ and that the diffusion and conductivity coefficients are such that ε_t follows a Drude dispersion model, $\varepsilon_t/\varepsilon_0=1-\omega_p^2/[\omega(\omega+i\omega_c)]$, and that k_L varies with frequency as

$$k_L^2=\frac{l_0}{c^2}\left[\omega\,(\omega+i\omega_c)-\omega_p^2\right], \tag{C5}$$

where l_0 is some dimensionless parameter. When $\omega_c=0$ the dispersion of the longitudinal plane-wave modes with propagation factor $e^{i\mathbf{k}\cdot\mathbf{r}}$ (volume plasmons) is $\omega^2=\omega_p^2+c^2k^2/l_0$, which is consistent with the so-called hydrodynamic model if $c^2/l_0=3v_F^2/5$ where v_F is the Fermi velocity [34]. For realistic metals l_0 is extremely large ($l_0>10^4$) and the volume plasmons are nearly dispersionless.

Next, we derive the characteristic equation for the TM^r polarized modes of oscillation supported by a core-shell nanoparticle. It is supposed that the core is made of a regular dielectric and that the cover shell is a metal with the electron gas described by the drift-diffusion model. For TM$_n^r$-polarized waves the electromagnetic field is a superposition of the transverse and longitudinal waves such that [30]

$$\mathbf{E}=\mathbf{E}_t+\mathbf{E}_l=\nabla\times\nabla\times\{\mathbf{r}\,\psi_T(r)Y_n(\hat{\mathbf{r}})\}+\nabla\{\psi_L(r)Y_n(\hat{\mathbf{r}})\}, \tag{C6a}$$

$$\mathbf{F}=\mathbf{F}_t+\mathbf{F}_l=\left(k_t^2-k_b^2\right)\nabla\times\nabla\times\{\mathbf{r}\,\psi_T(r)Y_n(\hat{\mathbf{r}})\}-k_b^2\nabla\{\psi_L(r)Y_n(\hat{\mathbf{r}})\}, \tag{C6b}$$

$$\mathbf{H}=+i\omega\varepsilon_t\psi_T(r)\,\hat{\mathbf{r}}\times\mathrm{Grad}Y_n(\hat{\mathbf{r}}), \tag{C6c}$$

where Y_n is a spherical harmonic of order n, Grad is the surface gradient operator, and ψ_T and ψ_L are solutions of the spherical Bessel equation, such that $r^{-1}\partial_r^2[r\psi_T]+[k_t^2-n(n+1)r^{-2}]\psi_T=0$ and $r^{-1}\partial_r^2[r\psi_L]+[k_L^2-n(n+1)r^{-2}]\psi_L=0$. For natural modes of oscillation of the core-shell nanoparticle, ψ_T and ψ_L are of the form

$$\psi_T=\begin{cases}Aj_n(k_1r), & r<R_1,\\ B_{1T}j_n(k_tr)+B_{2T}y_n(k_tr), & R_1<r<R_2,\\ Ch_n^{(1)}(k_0r), & r>R_2,\end{cases} \tag{C7a}$$

$$\psi_L=\begin{cases}B_{1L}j_n(k_Lr)+B_{2L}y_n(k_Lr), & R_1<r<R_2,\\ 0 & \text{otherwise},\end{cases} \tag{C7b}$$

with $k_0=\omega\sqrt{\varepsilon_0\mu_0}$ and $k_1=\omega\sqrt{\varepsilon_1\mu}$. The constants A, B_{1T}, B_{2T}, B_{1L}, B_{2L}, and C must be consistent with the continuity of the tangential components of the electromagnetic field and with the condition $\hat{\mathbf{r}}\cdot\mathbf{F}=0$ at the interfaces of the metallic region. The latter boundary condition ensures that the normal component of $\mathbf{J}_c$ vanishes at the interfaces [29,30,33]. Straightforward calculations show that

$$\hat{\mathbf{r}}\times\mathbf{E}(\mathbf{r})=\frac{1}{r}\{[r\psi_T(r)]'+\psi_L(r)\}\hat{\mathbf{r}}\times\mathrm{Grad}Y_n(\hat{\mathbf{r}}), \tag{C8}$$

where the prime denotes differentiation with respect to r. Thus, from the previous result and Eq. (C6c), it is seen that the continuity of the tangential electromagnetic fields is equivalent to the continuity of $[r\psi_T(r)]'+\psi_L(r)$ and $\varepsilon_t\psi_T(r)$. Hence, taking also into

account that

$$\hat{\mathbf{r}}\cdot\mathbf{F}=\left[\left(k_t^2-k_b^2\right)\frac{n\,(n+1)}{r}\psi_T\,(r)-k_b^2\psi_L'\,(r)\right]Y_n\,(\hat{\mathbf{r}})\,, \tag{C9}$$

we can readily obtain a homogeneous linear system of equations of the form $\mathbf{M}\cdot\mathbf{x}=0$ with $\mathbf{x}=(A,B_{1T},B_{2T},B_{1L},B_{2L},C)^T$ and the matrix $\mathbf{M}$ given by

$$\begin{pmatrix}
-\left[j_n\,(k_1r)\,r\right]'_{r=R_1} & \left[j_n\,(k_tr)\,r\right]'_{r=R_1} & \left[y_n\,(k_tr)\,r\right]'_{r=R_1} & j_n\,(k_LR_1) & y_n\,(k_LR_1) & 0\\
-\varepsilon_1 j_n\,(k_1R_1) & \varepsilon_t j_n\,(k_tR_1) & \varepsilon_t y_n\,(k_tR_1) & 0 & 0 & 0\\
0 & q\,j_n\,(k_tR_1) & q\,y_n\,(k_tR_1) & k_LR_1j_n'\,(k_LR_1) & k_LR_1y_n'\,(k_LR_1) & 0\\
0 & q\,j_n\,(k_tR_2) & qy_n\,(k_tR_2) & k_LR_2j_n'\,(k_LR_2) & k_LR_2y_n'\,(k_LR_2) & 0\\
0 & \left[j_n\,(k_tr)\,r\right]'_{r=R_2} & \left[y_n\,(k_tr)\,r\right]'_{r=R_2} & j_n\,(k_LR_2) & y_n\,(k_LR_2) & -\left[h_n^{(1)}\,(k_0r)\,r\right]'_{r=R_2}\\
0 & \varepsilon_t j_n\,(k_tR_2) & \varepsilon_t y_n\,(k_tR_2) & 0 & 0 & -\varepsilon_0 h_n^{(1)}\,(k_0R_2)
\end{pmatrix} \tag{C10}$$

with $q=(1-\varepsilon_t/\varepsilon_b)n(n+1)$. The frequencies of oscillation $\omega=\omega'+i\lambda$ (with $\lambda\leqslant 0$, i.e., in the lower-half-frequency plane) of the natural modes of oscillation of the open resonator are the solutions of the characteristic equation $\det(\mathbf{M})=0$.

APPENDIX D: SCATTERING BY A CORE-SHELL NANOPARTICLE

The scattering by a core-shell nanoparticle can be determined using standard Mie theory [27,35]. The electric field in all space can be written in terms of vector spherical harmonics as follows:

$$\mathbf{E}=E_0\sum_{n\geqslant 1}\frac{(2n+1)}{n\,(n+1)}i^n\left[\mathbf{M}_{n,o}^{\mathrm{TE}}+\mathbf{M}_{n,e}^{\mathrm{TM}}\right], \tag{D1a}$$

$$\mathbf{M}_{n,o}^{\mathrm{TE}}(\mathbf{r})=\boldsymbol{\nabla}\times\left\{\mathbf{r}\,f_n^{\mathrm{TE}}(r)\,Y_n^{\mathrm{TE}}(\hat{\mathbf{r}})\right\},$$
$$Y_n^{\mathrm{TE}}=-\sin\varphi\,P_n^1(\cos\theta), \tag{D1b}$$

$$\mathbf{M}_{n,e}^{\mathrm{TM}}(\mathbf{r})=\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\left\{\mathbf{r}\,\frac{1}{ik}f_n^{\mathrm{TM}}(r)\,Y_n^{\mathrm{TM}}(\hat{\mathbf{r}})\right\},$$
$$Y_n^{\mathrm{TM}}=-\cos\varphi\,P_n^1(\cos\theta), \tag{D1c}$$

where P_n^l are the generalized Legendre polynomials, and (r,θ,φ) is a system of spherical coordinates attached to the center of the spherical scatterer. The radial functions f_n^{TE} and f_n^{TM} satisfy the spherical Bessel equation, and can be decomposed into incident and scattered waves:

$$f_n^p=\begin{cases}f_n^{p,\mathrm{inc}}+f_n^{p,s}, & r>R_2,\\ f_n^{p,s}, & r<R_2,\end{cases}\qquad p=\mathrm{TE,\,TM}. \tag{D2}$$

When the incident electric field is a plane wave $\mathbf{E}^{\mathrm{inc}}=E_0\hat{\mathbf{x}}e^{ik_0z}$, the incident radial functions satisfy

$$f_n^{\mathrm{TE,inc}}=f_n^{\mathrm{TM,inc}}=j_n(k_0r). \tag{D3}$$

On the other hand, for a two-layer spherical particle the scattered radial functions are of the form

$$f_n^{p,s}(r)=\begin{cases}a_n^p\,j_n(k_1r), & r<R_1\\ b_{n,1}^p\,j_n\,(k_2r)+b_{n,2}^p\,y_n(k_2r), & R_1<r<R_2\,,\\ c_n^p\,h_n^{(1)}(k_0r), & r>R_2\end{cases}\qquad p=\mathrm{TE,\,TM}, \tag{D4}$$

where $k_i=\omega\sqrt{\mu_i\varepsilon_i}$. The unknown coefficients a_n^p, $b_{n,1}^p$, $b_{n,2}^p$, and c_n^p are determined by imposing the requirement that the total fields f_n^p satisfy the boundary conditions

$$f^{\mathrm{TE}}\quad\text{and}\quad\mu^{-1}\partial_r(rf^{\mathrm{TE}})\quad\text{are continuous,} \tag{D5a}$$
$$\mu^{-1}kf^{\mathrm{TM}}\quad\text{and}\quad k^{-1}\partial_r(rf^{\mathrm{TM}})\quad\text{are continuous.} \tag{D5b}$$

Let us suppose that the inner core is a vacuum ($\varepsilon_1=\varepsilon_0$) and that the permittivity of the cover shell is near zero and is modeled by $\varepsilon_2/\varepsilon_0=1-\omega_p^2/[\omega(\omega+i\omega_c)]$. When the radius of the inner core is tuned so that $R_1\approx R_{1,0}$ (the first resonance associated with the $n=1$ inner mode), it can be verified that for ω near the plasma frequency ω_p the Mie coefficient a_1^{TM} satisfies to a good approximation Eq. (7). From Eq. (D1) the contribution of the $n=1$ TM wave to the electric field inside the core is

$$\mathbf{E}_1^{\mathrm{TM}}=\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\left\{E_0a_1^{\mathrm{TM}}\frac{3}{2}\frac{j_1\,(k_0r)}{k_0}\mathbf{r}\cos\varphi\sin\theta\right\}, \tag{D6}$$

where we used $P_1^1\,(\cos\theta)=-\sin\theta$. In particular, at the origin we have

$$\mathbf{E}_1^{\mathrm{TM}}\big|_{\mathbf{r}=\mathbf{0}}=E_0a_1^{\mathrm{TM}}\hat{\mathbf{x}}. \tag{D7}$$

Thus, a_1^{TM} can be regarded as the transfer function that relates the incident-field amplitude (E_0) and the x- field component at the center of the inner core ($E_{\mathrm{in}}=E_0a_1^{\mathrm{TM}}$).

It is interesting to study the time dynamics of the field inside the open resonator. To this end we suppose that $E_0=E_0(t)$ has a finite (albeit very long) duration, and most of the spectral

content is concentrated at the frequency $\omega = \omega_p$. Then the corresponding field $E_{\text{in}}(t)$ at the center of the open resonator is given by the time convolution of $E_0(t)$ and $h(t)$, $E_{\text{in}}(t) = \int_{-\infty}^{+\infty} d\tau\, E_0(\tau) h(t-\tau)$, with $h(t)$ the inverse Fourier transform of a_1^{TM} [Eq. (7)]. Neglecting the phase ϕ_0 (which in general corresponds to a time delay $\phi_0 \approx \omega\, t_d$), we obtain

$$h(t) \approx \delta(t) + u(t) \frac{1}{\text{Re}\{\omega_r\}} \text{Im}[(\omega_r - \omega_L)(\omega_r + \omega_L^*) e^{-i\omega_r t}], \tag{D8}$$

where $u(t)$ represents the Heaviside step function. This formula shows several interesting things. First of all, it proves that the impulse response $[h(t)]$ has a characteristic duration determined by $e^{-i\omega_r t} \sim e^{\lambda t}$ where $\omega_r = \omega' + i\lambda$ and $\lambda < 0$. Thus, the lifetime is indeed of the order $1/(-2\lambda)$. Second, it shows that in the limit of no loss ($\omega_c \to 0$) and for $R_1 = R_{1,0}$ we have $h(t) \approx \delta(t)$, because in these conditions $\omega_r = \omega_L = \omega_p$. Thus, in this limit the incoming plane wave is unable to pump the open resonator because the amplitude of the corresponding self-sustained oscillation (the term $e^{-i\omega_r t}$) vanishes. However, surprisingly the fields inside the open resonator are nonzero in the considered limit,

$$E_{\text{in}}(t) \approx E_0(t) \quad (\text{when } \omega_c \to 0 \text{ and } R_1 = R_{1,0}), \tag{D9}$$

and follow closely the oscillations of the incoming field (as mentioned previously the time delay associated with the phase ϕ_0 is disregarded). At first sight, this surprising result might suggest that reciprocity is violated. The reciprocity theorem requires that $\int \mathbf{E}_1 \cdot \mathbf{j}_2 d^3\mathbf{r} = \int \mathbf{E}_2 \cdot \mathbf{j}_1 d^3\mathbf{r}$, where $(\mathbf{E}_i, \mathbf{H}_i)$ are solutions of the time-harmonic Maxwell equations associated with the source current density $\mathbf{j}_i$ ($i = 1,2$). Let $\mathbf{j}_1$ be associated with a source placed in the outer vacuum region (e.g., it radiates the plane wave), and $\mathbf{j}_2$ be a source placed inside the open resonator. When the metal collision frequency is nonzero, $\omega_c > 0$, the radiated fields obviously satisfy the reciprocity theorem. However, if $\mathbf{j}_1$ and $\mathbf{j}_2$ are kept fixed, the field $\mathbf{E}_2$ may diverge to infinity inside the resonator when $\omega_c \to 0^+$ (even though it stays finite in the exterior region to fulfill the reciprocity theorem), because no steady state can be reached for the inner excitation when $\omega_c = 0$ and $R_1 = R_{1,0}$. Thus, the reciprocity theorem does not hold when $\omega_c = 0$ and $R_1 = R_{1,0}$. Indeed, in this limit the system supports natural modes of oscillation with infinite lifetimes (the homogeneous problem can have nontrivial solutions), and thus the solution of a source problem is not unique, and, moreover, for some time-harmonic excitations a steady state may be impossible to reach.

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