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CHARACTER OF SURFACE PLASMONS IN LAYERED SPHERICAL STRUCTURES

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Abstract—This article discusses the effect of geometry on the surface plasmon resonances. The static dielectric polarizability of a sphere suffers a singularity when its permittivity relative to the surroundings is -2. This well-known resonance condition is changed when the shape of the particle is no longer a sphere. In this article the character of the resonances is studied, with a particular emphasis on a two-layer sphere.

1. INTRODUCTION

As is well known, the dielectric response of a small sphere may have a very strange behavior if the permittivity is allowed to be negative. The normalized polarizability of a homogeneous dielectric sphere with relative permittivity ϵ is

$$\alpha_n = \frac{\alpha}{\epsilon_0 V} = 3\frac{\epsilon - 1}{\epsilon + 2} \tag{1}$$

Here the absolute polarizability (the ratio between the dipole moment and the field creating it) is normalized by the volume of the sphere V and the free-space permittivity ϵ_0.

Obviously the polarizability grows without limit if the permittivity approaches the value -2. Especially peculiar is the fact that this result arises from purely electrostatic considerations where the solution of Laplace equation suffices in calculating the polarizability.

It is certainly counterintuitive to attach arbitrarily large polarizability values to small scatterers. Small particles are Rayleigh (in other words, very inefficient) scatterers. But a large polarizability means a large dipole moment, and consequently this would also entail a scattering cross section much larger than the geometric cross section. However, the condition that is necessary for such resonating behavior is also quite strange: negative permittivity. But again, today when very much interest is focused into the study of metamaterials which often not only possess negative permittivity but also negative permeability, we may perhaps be psychologically more open than before to broaden the allowed ranges for the various quantities and parameters in the materials characterization [1].

The condition $\epsilon = -2$ creates such a drastic behavior in the dielectric response of the particle that it has been much studied from many different viewpoints and branches in physics, optics, and engineering. Therefore it is understandable that the condition is known by different names and labels. A mild expression is "polarization enhancement" which it certainly can be said to be, a stronger one is "Mossotti catastrophe," where the name reminds us of the famous scientist Octavio Mossotti and his studies on materials modeling from the mid-19th century [2]. Very often the infinity condition (or conditions, because there may be several of them for more complex shapes than a homogeneous sphere, as shall be seen later in this study) is termed "normal mode" or "surface mode". It is also important to remember that negative values of permittivity are inherently band-limited phenomena and only at a certain frequency such a condition can hold (also accompanied with an imaginary part of the permittivity). The infinity condition goes hence often under the name "Fröhlich frequency" referring to the frequency for which the (real part of the) permittivity hits the value -2. The text by Bohren and Huffman [3] is very enlightening in discussing the interpretation of the Fröhlich conditions.

It is also understandably natural to call this strong phenomenon as "resonance" due to the infinity, possibly softened by losses. However, it is important to remember that the resonating character does not arise from a particular match of a wave between length scale and temporal variation as in an "ordinary" resonance but it is a phenomenon which can emerge from arbitrarily small structures.

The geometry of the negative-permittivity particle has a strong effect on its surface-plasmonic properties. In the literature, studies can be found where the spectra of normal modes have been analyzed for cubes and other rectangular and polyhedral particles [4–8]. These studies have shown that the simple behavior of a spherical case is suddenly lost and the resulting resonance spectrum becomes very complicated.

In the present article, the aim is to take a closer look at the spherically symmetric case and its surface plasmonic responses. Instead of deforming the "perfect" symmetry of sphere with sharp corners and edges, let us allow a radial variation in the structure of the sphere. The question of interest: how does the layeredness of a sphere affect the way its electric response becomes resonant in the domain of negative permittivities?

To start this study, first we shall look at the field behavior for a homogeneous sphere when the Fröhlich resonance is near, then the way how an ellipsoidal structure will affect this behavior, and finally move on to the radially layered sphere.

2. SURFACE PLASMON ON A SPHERE

The basic problem of a homogeneous dielectric sphere in a homogeneous z-directed electric field $E_0\mathbf{u}_z$ (which can be derived from the potential $-E_0 z = -E_0\, r\cos\theta$) has the well-known solution for the scalar potential inside (ϕ_1) and outside (ϕ_o) the sphere [9]:

$$\phi_1 = -E_0\frac{3}{\epsilon+2}r\cos\theta \tag{2}$$

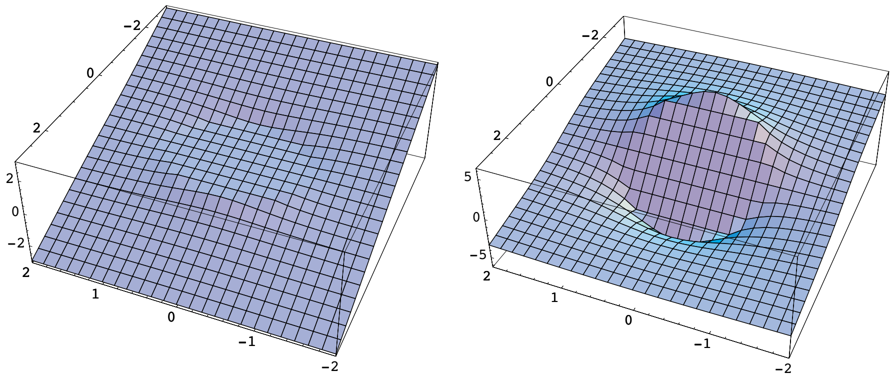
$$\phi_o = -E_0 r\cos\theta + E_0\frac{\epsilon-1}{\epsilon+2}\frac{a_1^3}{r^2}\cos\theta \tag{3}$$

and here a_1 denotes the radius of the sphere and ϵ its relative permittivity (it is assumed to be floating in the free space). From these relations it is seen that the internal field is decreased (and the potential flattened) by the factor $3/(\epsilon+2)$ compared with the incoming field amplitude and the field in the close neighborhood of the sphere is perturbed by the dipole field which is decaying as r^{-3} (the potential as r^{-2}).

Figure 1 shows the behavior of the potential inside and in the neighborhood of the sphere for two situations: far away and close to the surface plasmonic case.

3. ELLIPSOID

The case of a dielectric ellipsoid can fortunately be also treated analytically. The internal electric field and dipole moment of the ellipsoid, when posed into a uniform electric field is dependent on the axis ratios of the ellipsoid and its permittivity [11]. If the field points along one of the ellipsoid axes, the polarizability (ratio of the dipole



(a) $\epsilon = 5$ (b) $\epsilon = -1.5$

Figure 1. Potentials in the vicinity of the sphere with two different permittivities. Note the beginning of the development of a surface plasmon for the case of sphere with negative permittivity. As the permittivity decreases and passes the value -2, the internal field direction suddenly swaps and becomes opposite.

moment to the external constant field) is

$$\alpha_n = \frac{\epsilon - 1}{1 + N(\epsilon - 1)} \tag{4}$$

where N is the depolarization factor along this axis. Its value depends on the shape of the ellipsoid [12, 13]. The three depolarization factors for any ellipsoid satisfy

$$N_x + N_y + N_z = 1 \tag{5}$$

A sphere has three equal depolarization factors of 1/3. The other two special cases are a disc (depolarization factors $1, 0, 0$) and a needle $(0, 1/2, 1/2)$. For ellipsoids of revolution, prolate and oblate ellipsoids, various closed-form expressions for the factors can be found in [11].

The important observation to be gleaned from (4) is that the resonance condition depends on the depolarization factor in the following manner:

$$\epsilon = -\frac{1 - N}{N} \tag{6}$$

Therefore a spheroid (ellipsoid of revolution) has two Fröhlich resonances (for two orthogonal incident field directions) of which one happens for $\epsilon < -2$ and the other for $-2 < \epsilon < 0$. If the ellipsoid has three unequal axes, there are three resonances of which at least one is smaller than -2 and at least one larger than -2 (but of course negative).

4. MULTILAYER SPHERE

If the geometry of the scatterer is spherically symmetric the potential problem can be solved more easily than in the more general case. For example, consider a particle that has a homogenous spherical core (radius a_2, relative permittivity ϵ_2) surrounded by a spherical shell (radius a_1, relative permittivity ϵ_1) according to Figure 2. Again a constant static incident field $E_0\mathbf{u}_z$ illuminates the particle.

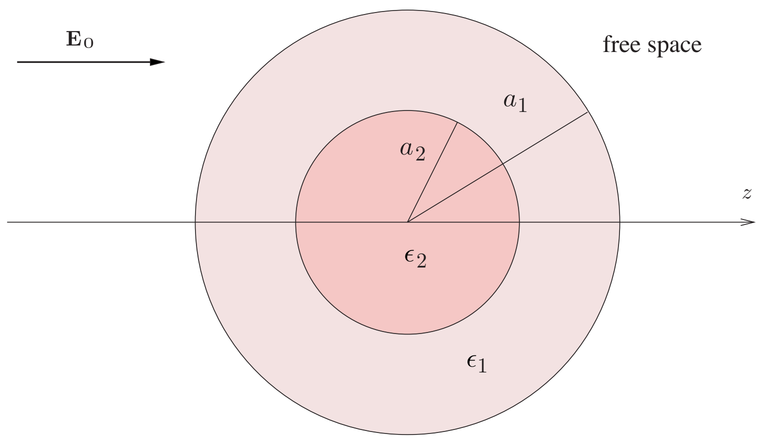


Figure 2. The core–and–shell sphere in a uniform electric field.

Laplace equation allows two type of solutions: a constant field and the dipole field in each of the three domains. The potential which satisfies the boundary conditions can be written [10] as

$$\phi_2 = -E_0 \frac{9\epsilon_1}{(\epsilon_1+2)(\epsilon_2+2\epsilon_1)+2\beta(\epsilon_1-1)(\epsilon_2-\epsilon_1)} r\cos\theta \quad (7)$$

$$\phi_1 = -E_0 \frac{3(\epsilon_2+2\epsilon_1)-3(\epsilon_2-\epsilon_1)(a_2/r)^3}{(\epsilon_1+2)(\epsilon_2+2\epsilon_1)+2\beta(\epsilon_1-1)(\epsilon_2-\epsilon_1)} r\cos\theta \quad (8)$$

$$\phi_o = -E_0 r\cos\theta + E_0 \frac{(\epsilon_1-1)(\epsilon_2+2\epsilon_1)+\beta(1+2\epsilon_1)(\epsilon_2-\epsilon_1)}{(\epsilon_1+2)(\epsilon_2+2\epsilon_1)+2\beta(\epsilon_1-1)(\epsilon_2-\epsilon_1)} \frac{a_1^3}{r^2}\cos\theta \quad (9)$$

where $\beta = a_2^3/a_1^3$ is the volume fraction that the core occupies from the whole particle. (Note that no dipole term is present in the core region, in the function ϕ_2)

From the amplitude of the dipole component in the external domain, the equivalent dipole moment of the particle can be identified, and the proportionality factor is the polarizability, which reads [11]

$$\alpha_n = 3\frac{(\epsilon_1 - 1)(\epsilon_2 + 2\epsilon_1) + \beta(2\epsilon_1 + 1)(\epsilon_2 - \epsilon_1)}{(\epsilon_1 + 2)(\epsilon_2 + 2\epsilon_1) + 2\beta(\epsilon_1 - 1)(\epsilon_2 - \epsilon_1)} \tag{10}$$

This expression reveals us the resonance conditions, in other words the values for the permittivity and structure parameters for which the polarizability grows large. Let us take a closer look at the two different cases, where either the core or the shell is plasmonic.

In the following, all the numerical examples are calculated for the case $\beta = 0.03$ which means that the radius ratio is $a_2/a_1 = \sqrt[3]{0.03} \approx 0.31$.

Plasmonic core

How does an "ordinary" dielectric coating affect the plasmonic behavior of a sphere? In other words, the case of infinite value for the polarizability α_n requires a certain negative value for the permittivity of the core ϵ_2 which is dependent on the (positive) value of the shell ϵ_1. Figure 3 shows the behavior of the polarizability as a function of the core permittivity ϵ_2 with the shell permittivity values $\epsilon_1 = 1$ (no shell) and $\epsilon_1 = 2$.

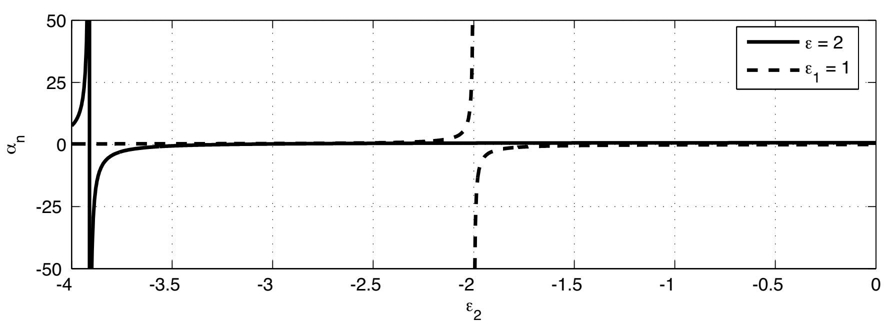


Figure 3. The polarizability of a plasmonic core (as a function of its permittivity ϵ_2) with a cover outside that has permittivity $\epsilon_1 = 2$. The core occupies a volume of $\beta = 0.03$. For comparison the polarizability curve of an uncovered sphere is shown with dashed line.

The effect of a dielectric coating is that the Fröhlich condition $\epsilon_2 = -2$ will be shifted to more negative values. This is also obvious from the requirement that the denominator of the polarizability (10) vanish:

$$\epsilon_2 = -2\epsilon_1 \frac{\epsilon_1 + 2 - \beta(\epsilon_1 - 1)}{\epsilon_1 + 2 + 2\beta(\epsilon_1 - 1)} \quad (11)$$

Plasmonic coating

The case when the shell is plasmonic and the core "ordinary" dielectric is more interesting. As an example, Figure 4 shows the effect of a hollow core (void or bubble, $\epsilon_2 = 1$) inside a plasmonic covering ϵ_1 on the polarizability. It can be observed that two plasmonic resonances are

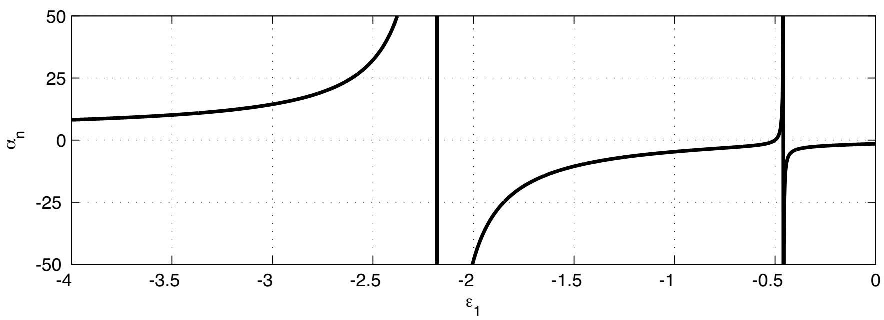


Figure 4. The polarizability of a plasmonic shell (as a function of its permittivity ϵ_1) which has a hollow core that occupies a volume of $\beta = 0.03$. Note the two resonances and the fact that the one closer to zero-permittivity is much narrower.

created which are located on the ϵ_1 axis to both ways from -2. One of these behaves like the dielectrically-covered case of Figure 3 ($\epsilon_1 < -2$) but the other resonance is moved to larger values of permittivity. This provides a way to create plasmonic resonances closer to zero-permittivity with a spherical symmetric structure.

The positions of the two resonances can again be calculated from the divergence of expression (10):

$$\epsilon_1 = -\frac{\epsilon_2 + 4 + 2\beta(\epsilon_2 + 1) \pm \sqrt{[\epsilon_2 + 4 + 2\beta(\epsilon_2 + 1)]^2 - 16\epsilon_2(1-\beta)^2}}{4(1-\beta)} \quad (12)$$

Potential distributions

How do the potentials and fields look like when a plasmon creeps into the scatterer? Visualization can be done using the expressions for the potentials that have been given in the earlier sections.

For the case of a plasmonic core with a dielectric cover, Figure 5 shows the scalar electric potential over the z-axis through the center of the sphere (here the shell permittivity is assumed $\epsilon_1 = 2$). For values $\epsilon_2 > 1$, the potential slope is small (field is small inside the sphere), but when the core permittivity goes negative, the electric field is strongly amplified, undergoes a singularity where it swaps direction (at $\epsilon_2 \approx -3.91$, cf. Equation (11)) and then stabilizes back to the low-internal-field behavior. Indeed, both cases $\epsilon_2 \to \pm\infty$ seem to be identical.

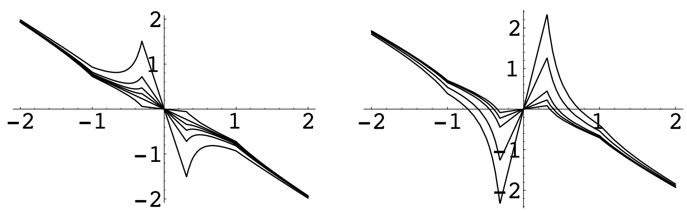


Figure 5. The potential of a core-plus-shell sphere. $\epsilon_1 = 2$ and the permittivity of the core varies $\epsilon_2 = 20, 2, .1, -1, -2, -3$ (left) and $\epsilon_2 = -4.5, -5, -7, -10, -20$ (right). The core occupies a volume of $\beta = 0.03$ corresponding to radius ratio of around 0.31. Note that the field direction in the core changes as the resonance condition $\epsilon_2 \approx -3.91$.

On the other hand, the case of a plasmonic cover over a hollow bubble ($\epsilon_1 < 0$ and $\epsilon_2 = 1$) shows a more detail-rich behavior, as can be expected from the polarizability curve of Figure 4 that showed bifurcating resonances. Electric potential distributions in the case of a spherical void with $\beta = 0.03$ are shown in Figures 6–7.

These figures show that unlike in the case of a plasmonic core, now the surface plasmon can be generated both at the inner boundary and the outer boundary of the shell, and hence the character of the field distribution is also much more elaborate. For example, the narrow character of the inner-boundary plasmon and the breadth of the outer one are very conspicuous in the figures. Also the swapping of the internal field direction over the two "resonance" conditions from Equation (12), $\epsilon_1 \approx -0.459, -2.18$ can be seen from these figures.

Another look at the potential distributions can be taken if the potential cuts through the center are juxtaposed into a surface plot with varying negative permittivity parameter. First, in Figure 8, this potential is depicted for a homogeneous dielectric sphere, and the expected $\epsilon = -2$ resonance shows itself clearly.

A very similar three-dimensional plot would be the result

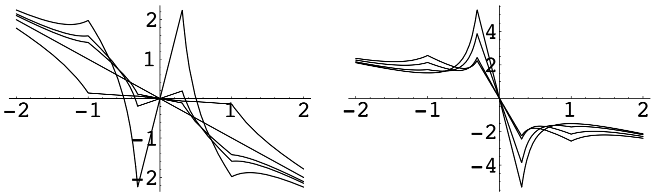


Figure 6. The potential of a core-plus-shell sphere. $\epsilon_2 = 1$ and the permittivity of the shell varies $\epsilon_1 = 20, 1, .1, -.1, -.34$ (left) and $\epsilon_1 = -.55, -.6, -.8, -1$ (right). The core occupies a volume of $\beta = 0.03$ corresponding to radius ratio of around 0.31. Note that the plasmon is located at the inner boundary (between core and shell), and after the first resonance $\epsilon_1 \approx -0.459$ is overtaken, the field direction switches.

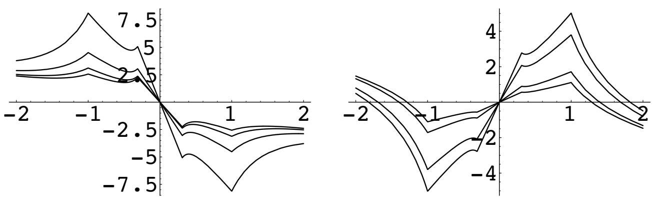
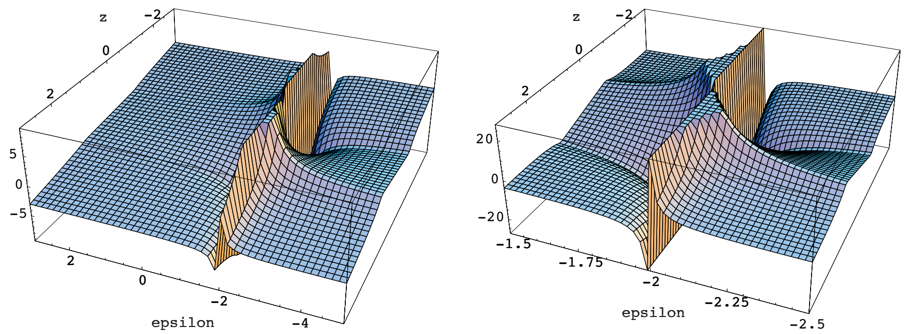


Figure 7. The potential of a core-plus-shell sphere. $\epsilon_2 = 1$ and the permittivity of the shell varies $\epsilon_1 = -1, -1.2, -1.5, -1.8$ (left) and $\epsilon_1 = -2.8, -3, -4, -5$ (right). The core occupies a volume of $\beta = 0.03$ corresponding to radius ratio of around 0.31. Note that the plasmon now has moved to the outer boundary, and after the second resonance $\epsilon_1 \approx -2.18$ again the field direction switches.

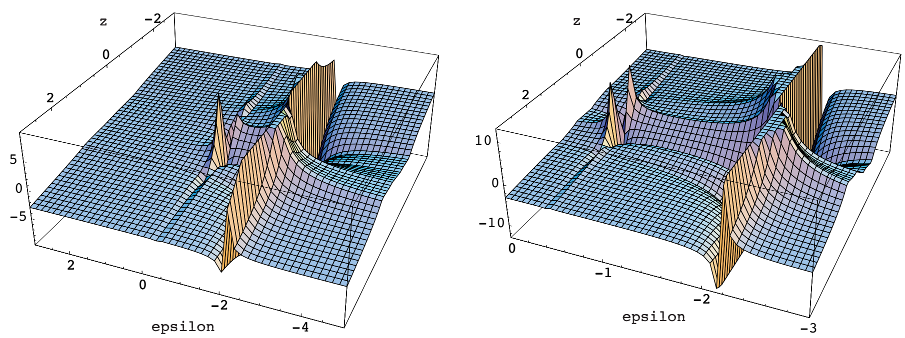
for a dielectrically-covered plasmonic sphere. However, when the characteristics of the electric potential of a hollow plasmonic shell is plotted, the result is shown in Figure 9. These provide another illuminating look into the narrowness and position of the two resonances that are created for the spherical void.

A third illustration of the plasmon can be taken by plotting the potential as height within a planar cut through the center of the sphere (either the xz or the yz-plane of Figure 2). This is done for the hollow plasmonic sphere in Figure 10 for certain values of the shell permittivity that are close to the two resonances. There figures are perhaps the most effective in transmitting an image of the two distinct plasmons that are localized either on the inner shell boundary (the narrow resonance) or the outer boundary (the broad resonance).



(a) $3 > \epsilon > -5$ (b) $-1.5 > \epsilon > -2.5$

Figure 8. Potential through the center of a homogeneous sphere as a function of the permittivity. The sphere surface is crossed at $z = \pm 1$.



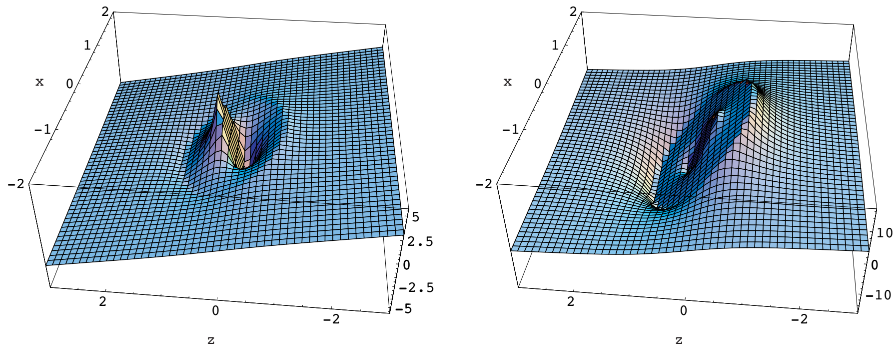
(a) $3 > \epsilon_1 > -5$ (b) $-1.5 > \epsilon_1 > -2.5$

Figure 9. Potential through the center of a hollow sphere as a function of the permittivity ϵ_1. The parameter $\beta = 0.03$, and the boundaries are at points $z = \pm 1$, $z = \pm 0.31$.

Zero-permittivity shell

In addition to the special attention of the present paper (surface-geometric effects on the normal mode structure of small scatterers), another connected and interesting case is the one which involves material with vanishing permittivity, $\epsilon = 0$. Let us take a closer look at the potential and field distributions of a core-plus-shell sphere where the covering layer has permittivity $\epsilon_1 = 0$.

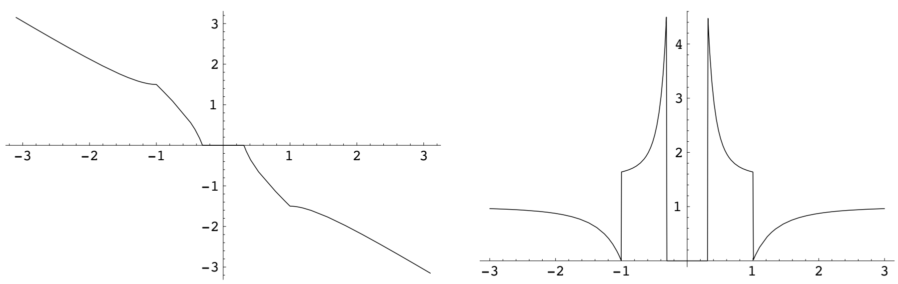
Figure 11 shows the amplitudes of the potential and the electric field over the z axis in the $\epsilon_1 = 0$ case. The field inside the core vanishes totally. In other words, the $\epsilon_1 = 0$ -covering shields its inside from external fields just like a metal or a very high-permittivity layer.



(a) $\epsilon_1 = -0.4$ (b) $\epsilon_1 = -2$

Figure 10. Potential in the vicinity of a hollow sphere with negative permittivity. Left: case close to the "narrow" resonance, located at the inner boundary of the shell. Right: "broad" outer boundary resonance.

However, what is different between these two cases is that for the metal cover, the field vanishes also in the cover but as Figure 11 shows, the electric field for the zero-permittivity shell does penetrate into the cover. Field exists through the sphere because there is a potential difference across it.

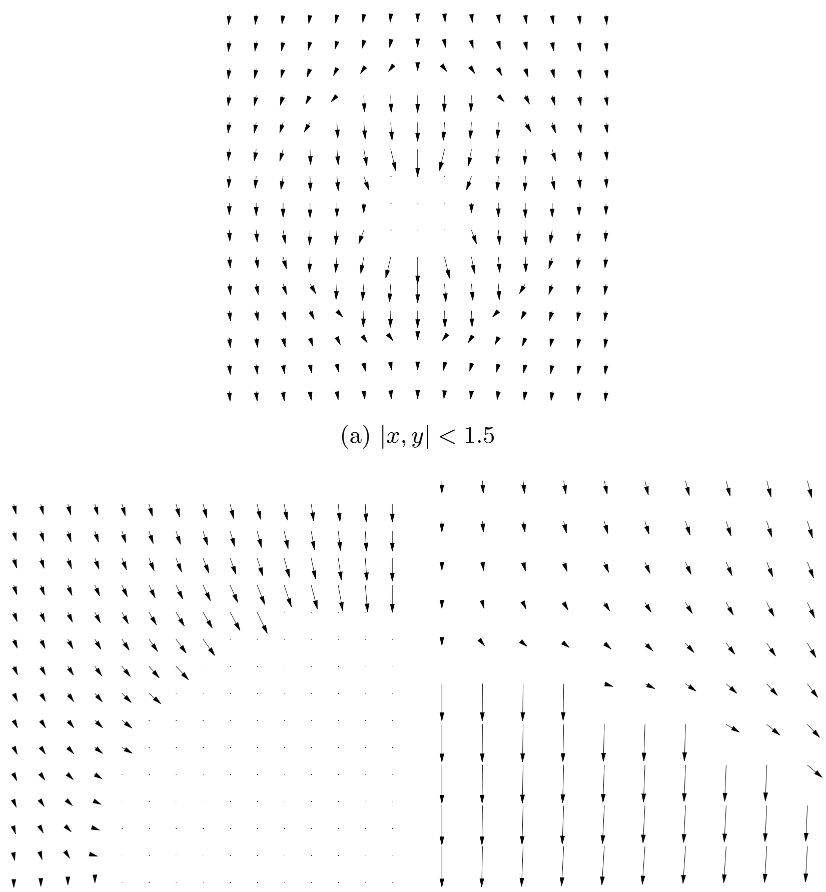


(a) potential (b) field

Figure 11. Potential and field for a hollow sphere with zero-permittivity shell.

How does the field avoid the core but enter the layer? Figure 12 shows the direction and the amplitude of the field distribution in the xz plane.

Because the layer has permittivity $\epsilon_1 = 0$, no flux exists there, and hence the external field (outside the particle) cannot have any radial component on the boundary. Therefore it has to "flow around" the sphere, avoiding it like a river perturbed by an island. The boundary conditions require that the electric field be tangential on the surface of a zero-permittivity material (since normal component is excluded by the zero flux requirement in the shell). Also on the inner boundary one must notice that the field on the side of the $\epsilon_1 = 0$ -domain is normal to the boundary (because there is no field in the core and the tangential electric field needs to be continuous). These observations can be visually appreciated in Figure 12.



(a) $|x, y| < 1.5$

(b) $-0.4 < x < 0 < y < 0.4$

(c) $0 < x < 0.6;\ 0.7 < y < 1.3$

Figure 12. Field distribution in the xz-plane for a hollow sphere with zero-permittivity shell. The lower figures are close-ups of the fields across the inner (b) and outer (c) boundary of the shell.

It is interesting to note—and as a matter of fact quite obvious—that the polarizability of a $\epsilon_1 = 0$ -covered sphere is $-3/2$, totally independently of the core size β and the core permittivity ϵ_2.

5. DISCUSSION

The expressions, results, and illustrations of the surface plasmonic behavior of layered spheres that have been presented above give rise to several observations.

- The singular behavior of the polarizability is not a wave resonance but a electrostatic phenomenon in the sense that all its characteristics are known from the solution of Laplace equation and scalar potential. At the same time it is necessary to remember that any material response characterized with negative permittivity is not compatible with statics. Materials with strongly frequency-dependent material response, e.g. Lorentz-dispersive media, can display effectively negative permittivity behavior. But such a frequency region is of course band-limited. However, if the particle is very small compared with the wavelength, we can neglect retardation effects and can assume the field distribution be in-phase over the whole region of interest. Then the solution of the electrodynamic problem can be found from the electrostatic one. And the singularity is there no matter how small the particle is.
  A related problem is present: since the negative values of the permittivity are only allowed for dispersive materials, Kramers–Kronig-relations [9, 14] dictate that the medium is dissipative and contains losses. Therefore for practical calculations the imaginary part of the permittivity has to be included. However, since the relations connecting the spectra of the real and imaginary parts are global integrals, a negative value of the real part does not necessary imply a large imaginary part at the same frequency but the losses may be concentrated around another spectral region.
- Particle shape has effect on the value of the negative permittivity corresponding to the Fröhlich resonance. We saw that the dielectric covering shifts the resonance into more negative values, cf. Equation (11). On the other hand, with ellipsoids, oriented in a certain direction, it was possible to achieve Fröhlich resonances with permittivities closer to zero (using depolarization factor values N larger than $1/3$, see Equation (6)).
  However, in the case of a hollow sphere (an "ordinary" dielectric covered with plasmonic material, $\epsilon_1 < 0$) we could see that one

plasmonic resonance was indeed located in the interval $-2 < \epsilon_1 < 0$.

- For the plasmonic coating case, there are two resonances. But the character of these two is very different: the one "closer to zero" ($-2 < \epsilon_1 < 0$) is very narrow but the one further away ($\epsilon_1 < -2$) has a broad extent. One could say that the single resonance that takes place in a plasmonic core with dielectric coating falls between these two in this respect. Also the plasmon $-2 < \epsilon_1 < 0$ is localized at the inner boundary of the shell whereas the other one is extended more broadly but clearly in the neighborhood of the outer boundary.
- A very interesting observation is that even if the geometry of the particle becomes more complicated than a homogeneous sphere, the spectrum of the resonances remains simple. Only one or at most two resonances are generated. This is is stark contrast with the case of a cube that displays a very complex structure of polarizability singularities for negative permittivity values [6].
  Again this is a reason to remind that the "resonances" in the polarizability are different from the wavelength-related resonances in cavities. For a spherical metal cavity (or a spherical dielectric resonator for that matter) there exists an infinite number of resonant frequencies, just like for a cubic cavity. However, the normal modes for surface plasmons are a different story: for a homogeneous sphere there is only one singularity, for a cube several.
- Important physics can also be gleaned from the analysis of the field behavior of a zero-permittivity shell. Such a covering acts similarly with a metal shell ($\epsilon_1 \to \pm\infty$) in the Faraday-cage sense that the core is shielded from any outside fields. But the difference is that for a metal shell, the field is zero also in the covering, and in the other case, non-zero field (even if not flux density) exists in the $\epsilon_1 = 0$ -layer.
  Also the polarizability of a metal-covered spherical core ($\alpha_n = 3$) and that of the $\epsilon_1 = 0$ -covered core ($\alpha_n = -3/2$) are independent of the size or the material of the core.
- And finally, it is worth noting that analytical results for the plasmonic behavior can be calculated for multilayer spheres with arbitrary number of layers and the findings of this article can be greatly generalized. Closed-form expressions of the polarizabilities and potential and field functions can be calculated using the transmission-line approach presented in [10].

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