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Keywords
metamaterials, nanoantennas, optical frequencies, super-directivity

Comments
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Enhanced Directivity From Subwavelength Infrared/Optical Nano-Antennas Loaded With Plasmonic Materials or Metamaterials

Andrea Alù, *Member, IEEE*, and Nader Engheta, *Fellow, IEEE*

***Abstract*—Here, we explore theoretically the concept of enhanced directivity from electrically small subwavelength radiators containing negative-parameter materials, such as plasmonic materials with negative permittivity at THz, infrared and optical frequencies. In particular, we study higher order plasmonic resonances of a subwavelength core-shell spherical nano-antenna, and we analyze the near-zone field distributions and far-field radiation patterns of such a structure when it is excited by a small dipole source, demonstrating analytically and numerically the possibility of having highly directive patterns from a nano-structure with electrically small dimensions. Radiation characteristics and intrinsic limitations on performance are analyzed in detail, and a potential application of this novel technique for super-resolution detection of the displacement of a nano-object is also pointed out.**

***Index Terms*—Metamaterials, nano-antennas, optical frequencies, super-directivity.**

I. Introduction

REDUCING the dimensions of radiators represents a challenge of great interest in the engineering community, owing to the several practical advantages of this opportunity in many fields. In this sense, the interest in THz, infrared and optical antenna technology has significantly increased in the recent years, since raising the frequency of operation may have significant impact in reducing the physical size of the antenna and in increasing its operational bandwidth. Increasing the frequency of operation, however, does not necessarily allow squeezing the *electrical* size of the radiator for a desired level of performance. The intrinsic limitations in how much the size of a radiator may be reduced at the expense of other antenna parameters (e.g., efficiency and bandwidth) has been widely discussed in the technical literature over the past several decades (see e.g., [1]–[12] and references therein), and these limits are, in general, intrinsic and independent of the actual frequency of operation.

It is a common knowledge how electrically small apertures and antennas, if not matched properly, do not generally radiate efficiently, since they are far from their self-resonances, and their radiation patterns have low directivity, since the radiated energy is widely distributed over most directions. To increase the radiation directivity towards specific directions, therefore, the use of electrically large antenna arrays is a common practice. This enlarges the total radiating area, allowing the designer to squeeze the beam in certain desired directions. Theoretically, there is no upper limit to the level of directivity of a given electrical aperture with a fixed size [1]–[12], even though the efficiency, bandwidth and robustness of the antenna radiation decrease exponentially when trying to increase the directivity beyond the limit represented by a uniformly illuminated aperture of the same effective size. Superdirective arrays operate based on the destructive interference of radiation from the elementary radiators composing the array, which should necessarily be fed out of phase with respect to their neighbors placed in close proximity of one another [7]. This induces huge reactive (stored) fields in the array vicinity, making practically impossible for a feeding network to fulfill the required feed amplitudes and phases for elements in a realistic setup, due to matching and coupling issues. Moreover, the sensitivity of the super-directive effect on the relative amplitude and phase of each element becomes extremely high when the prescribed super-directivity is increased. This is why levels of directivity even slightly superior to those of a uniformly illuminated aperture are challenging to obtain in practice.

The limitation in the maximum directivity achievable with a given aperture size may be interestingly connected to the inherent limitation of lensing and focusing of subwavelength details into a far-field image [6]: if making a small aperture radiating directively towards a given direction were possible, then one would speculate on the possibility of resolving different tiny details of an object into the image plane placed in the far field. In this sense, it is interesting to remark that the resonance Q factor of antennas, a measure of the bandwidth and effective realizability of such super-directive radiators, is connected to the evanescent stored energy present near the radiator. This somehow connects this problem to the interesting debate on the possibility of achieving subwavelength resolution of an image using special materials. Indeed it has been shown recently how the use of metamaterials with negative constitutive parameters may overcome, at least in the near-field, the usual diffraction limitations governed by the evanescent fields in imaging tools [13].

In this context, the use of negative-index, or double-negative (DNG), metamaterials to overcome certain conventional limitations in different phenomena has recently attracted a great deal of attention in the physics and engineering communities. Their realization, made possible at microwave frequencies by inducing a conjoined resonance of electric and magnetic inclusions in a host material that may lead to negative values for real

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parts of effective permittivity and permeability in a given band of frequencies, has been first shown experimentally in 2000 by the group in the University of California, San Diego [14]. The speculations on the possible applications of such materials have been numerous, some of which are connected with the problem of subwavelength resolution [13]–[15]. This mechanism, which works in the near-field through the anomalous "growth" of evanescent waves in a DNG material, has raised a lot of attention, since, as already anticipated, it may allow in principle to overcome the inherent limitations of near-field imaging and microscopy. We have proposed in recent years other ways of utilizing negative-index metamaterials for overcoming the limitations that conventional positive-parameter materials exhibit in different setups and configurations. Sub-wavelength resonant cavities and waveguides [16]–[18] and subwavelength resonant scatterers [19] are examples of these possibilities. Since most of these applications work in the subwavelength scale, it is generally possible to employ ε-negative (ENG) or μ-negative (MNG) materials, instead of double-negative ones that can be more challenging to construct, for the specific polarization of interest. ENG materials, moreover, are already available in nature as polar dielectrics and noble metals at THz, infrared and optical frequencies [20].

As for the potential features of such double negative and single-negative (SNG) materials, it has been shown in the past how pairing materials with oppositely-signed constitutive parameters may induce a resonance at their interface, in which the field amplitude rapidly varies in a small scale [15]. Interestingly enough, in some sense this intuitively reminds us of the sharply varying amplitudes required in super-directive arrays, which produce a directive beam from a small radiator or aperture. Here, however, the sharp variation of the field is an inherent property of the interface between complementary materials with oppositely-signed material parameters, and as a result such a field distribution may, in principle, be formed without a need for sensitive feed lines. We may therefore speculate that perhaps this field distribution might be less sensitive to imperfections and material losses and it may lead to a relatively more robust way to achieve super-directive radiation from subwavelength sources when compared with the classic techniques for enhanced directivity proposed in the literature.

Motivated by this intuition about the peculiar field variation at the interface between oppositely-signed-parameter materials, in the following we investigate the possibility of employing metamaterials with negative constitutive parameters, i.e., DNG, ENG or MNG materials, to design nano-radiators with enhanced directivity. This is based in particular on the resonant excitation of higher order *material polaritons* [21] in subwavelength structures, that may dominate the near-zone electromagnetic field distributions with rapid spatial variation in a small scale concentrated near the interface between the negative-parameter and positive-parameter materials. This effect may be connected to a directive radiation, similar to what happens in superdirective arrays. The advantage of this configuration is that the resonant higher order mode around the radiator may establish automatically in a subwavelength scale without a need for complicated feeding networks as in tightly packed arrays, and it may therefore present a new possibility for synthesizing efficient subwavelength super-directive radiators. Although completely general, this anomalous effect may be particularly appealing in those frequency regions, namely THz, infrared and visible frequencies, for which materials are naturally characterized by negative real part of permittivity and reasonably low losses (for instance, silver (Ag) has a loss tangent of about 10^{-2} at a wavelength of 750 nm and silicon carbide (SiC) has a loss tangent of about 0.05 at 11 μm). Polar dielectrics, noble metals and some semi-conductors are examples of naturally available ENG materials and in the following we envision nano-antennas made of such materials supporting proper resonant modes that meet the previous requirements.

As an aside, it should be mentioned how the general use of materials with special values of their effective constitutive parameters is not novel in the technical literature as an effective way to enhance the performance of antenna setups. The use of low-index superstrates to enhance the radiation directivity has been proven effective in several applications [22]–[27]. However, such a configuration relies on a totally different physical phenomenon: the directivity is enhanced by enlarging the effective radiating aperture of the antenna by several wavelengths. This phenomenon is not a superdirective effect, but rather it exploits the possibility that low-index materials offer for enlarging the constant-phase radiating aperture of an antenna. Plasmonic materials have also been recently proposed as covers on simple radiating structures to maximize the power extracted from subwavelength radiators [28]–[30], but although these configurations possibly improve the antenna gain, efficiency and matching, they do not modify (and they are not intended to improve) the radiation pattern and directivity noticeably. In this sense, nano-antenna technology based on plasmonic resonant nano-particles has also been subject of some recent numerical and experimental investigations [31]–[37].

In the following sections we present our theoretical and numerical results on the possibility of employing plasmonic materials or metamaterials to synthesize high-directivity nano-antennas supporting plasmonic resonances in subwavelength devices. Application of these concepts at THz, infrared and optical frequencies is envisioned. An $e^{j\omega t}$ time dependence is assumed throughout the manuscript.

II. Theoretical Analysis

Consider for simplicity a subwavelength nano-antenna (e.g., a core-shell spherical system) embedded in a transparent background with permittivity ε_0 and permeability μ_0, as depicted in Fig. 1. Let us assume first that the antenna works as a scatterer and it is excited by a uniform plane wave traveling along the z axis and with electric field polarized along x. Its scattering properties may be derived by expanding the radiated field in terms of multipoles. Under the hypothesis of subwavelength size, usually the dipolar fields are dominant when conventional materials are used, and the nano-antenna is conveniently described in terms of its electric and magnetic polarizabilities. In this case, the far-field distribution has the standard "donut" shape of a dipolar pattern, azimuthally uniform around the direction of polarization and with one null along the polarization axis. A quasi-uniform near field is also induced inside the nano-antenna under this assumption. Applying reciprocity, this implies

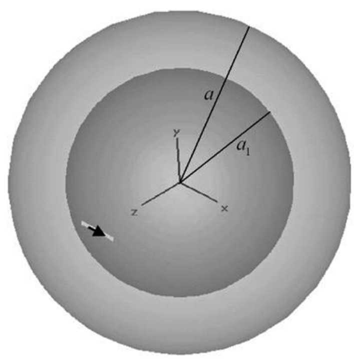

Fig. 1. Geometry of a simple nano-antenna: a spherical core-shell system embedded in a suitable Cartesian reference system and excited by a short feeding dipole placed at the interface between core and shell materials along the z axis.

that a small electric source embedded in this subwavelength object, as shown in Fig. 1, would make the whole system radiate as a short-dipole, with a weak and low-directive pattern. This is consistent with any small dielectric resonator antenna, or any small-scale radiator.

In [19], however, we have shown how pairing together materials with oppositely-signed constitutive parameters, i.e., employing ENG, MNG, or DNG materials together with conventional positive-parameter materials, would induce a local resonance in a subwavelength scale at the interface between oppositely- signed materials. This is also consistent with numerical and experimental results presented in the recent technical literature [38], [39]. To address this issue more formally, let us consider the core-shell spherical nano-antenna, as in [19], for which the closed-form condition for getting a TM_{n1} resonance, of polar order n, is found in [19] to be (1), shown at the bottom of the page.

Here, a_1 is the core radius (and shell inner radius), $a > a_1$ is the shell outer radius, ε_1 and ε_2 are the respective permittivities, $k_i = \omega\sqrt{\varepsilon_i \mu_i}$, with ω being the radian frequency of excitation and μ_i the permeability of the materials, and j_n, y_n are spherical Bessel functions of order n [40]. For the TE_{n1} resonances an analogous formula may be easily derived exchanging the role of permittivities and permeabilities. In the small radii subwavelength limit, when positive-index materials are considered for both layers, (1) can never be satisfied, regardless of n, consistent with the fact that a very small subwavelength structure made of conventional materials does not generally support any resonant mode. However, when negative-materials can be used for one of the shell or core materials, the following approximate condition for TM_{n1} resonance of order n may be found for the core-shell ratio of radii [19], under the small-argument assumption $k_i a \ll 1 (i = 0, 2)$

$$\frac{a_1}{a} \simeq \sqrt[2n+1]{\frac{[(n+1)\varepsilon_0 + n\varepsilon_2][(n+1)\varepsilon_2 + n\varepsilon_1]}{n(n+1)(\varepsilon_2 - \varepsilon_0)(\varepsilon_2 - \varepsilon_1)}}. \quad (2)$$

This condition implies that for specific pairs of "complementary" materials with oppositely-signed permittivities, and specific ratios of radii, resonances of any order n may in principle be induced in this nano-antenna, without a lower limit on its total size. (MNG materials may induce the dual TE_{n1} magnetic resonances). The presence of material losses at some point practically prevents the excitation of higher order n resonances in too small particles, since, as discussed in the following, the quality factor Q of these resonances increases with n and with reduction of the size of the particles, but the theoretical concept of envisioning these "localized" resonances may lead to exciting applications in several areas, as mentioned in [19].

The possibility of using the $n = 1$ resonance of a subwavelength DNG or ENG spherical shell for enhancing the radiation performance of small antennas has been first proposed in [28] and further discussed in [29], [30] and related works. In those cases, the dipolar resonance does not modify the radiation pattern that a small dipole would have when isolated, but the presence of a surrounding subwavelength DNG or ENG shell, at the resonance predicted by (1) and (2), is shown to extract significantly more power from the source for a fixed level of current on the exciting dipole [28]–[30]. It is interesting to note how in the lossless limit any higher order n resonance may be excited in the same subwavelength scale, by properly choosing the parameters of the core-shell materials, the ratio of radii in order to satisfy (2) and the position of the feeding dipole. Unlike the case of $n = 1$ resonance, for higher order n the field distributions in the near zone and inside the particle may be sharply varying in a very small scale. For instance, in Figs. 2 and 3 are plotted the magnitude and phase of the near-zone electric and magnetic field distributions in the two planes orthogonal, respectively, to the exciting electric and magnetic field vectors (respectively the H and E planes of the nano-antenna). In these figures, we show the first four orders TM_{n1} resonant modes in a spherical nano-antenna as in Fig. 1 composed of a lossless spherical shell of uniform permittivity $\varepsilon_2 = -5\,\varepsilon_0$ and outer radius $a = \lambda_0/20$ with a hollow core with $\varepsilon_1 = \varepsilon_0$. This may represent the case of a plasmonic material shell at THz, infrared or optical frequencies, at which such values of permittivities and sufficiently low losses are available naturally, or more in general the case of a metamaterial specifically constructed to achieve the desired value of permittivity at the frequency of interest. For each n, the inner radius a_1 is chosen such that (1) is satisfied. In each figure the core-shell geometry is also sketched and the field values are evaluated in the fully dynamic case under plane wave incidence when the nano-antenna works as a "reflector".

For $n = 1$, which is the conventional dipolar resonance, the field is much enhanced compared with the case of a standard dielectric particle of the same size,[1] due to the resonant exci-

[1]The field enhancement for the case at hand with respect to a regular dielectric particle with $\varepsilon_2 = +5\,\varepsilon_0$ is of about two orders of magnitude.

$$\begin{vmatrix} j_n(k_1 a_1) & j_n(k_2 a_1) & y_n(k_2 a_1) & 0 \\ [k_1 a_1 j_n(k_1 a_1)]'/\varepsilon_1 & [k_2 a_1 j_n(k_2 a_1)]'/\varepsilon_2 & [k_2 a_1 y_n(k_2 a_1)]'/\varepsilon_2 & 0 \\ 0 & j_n(k_2 a) & y_n(k_2 a) & y_n(k_0 a) \\ 0 & [k_2 a\, j_n(k_2 a)]'/\varepsilon_2 & [k_2 a\, y_n(k_2 a)]'/\varepsilon_2 & [k_0 a\, y_n(k_0 a)]'/\varepsilon_0 \end{vmatrix} = 0. \quad (1)$$

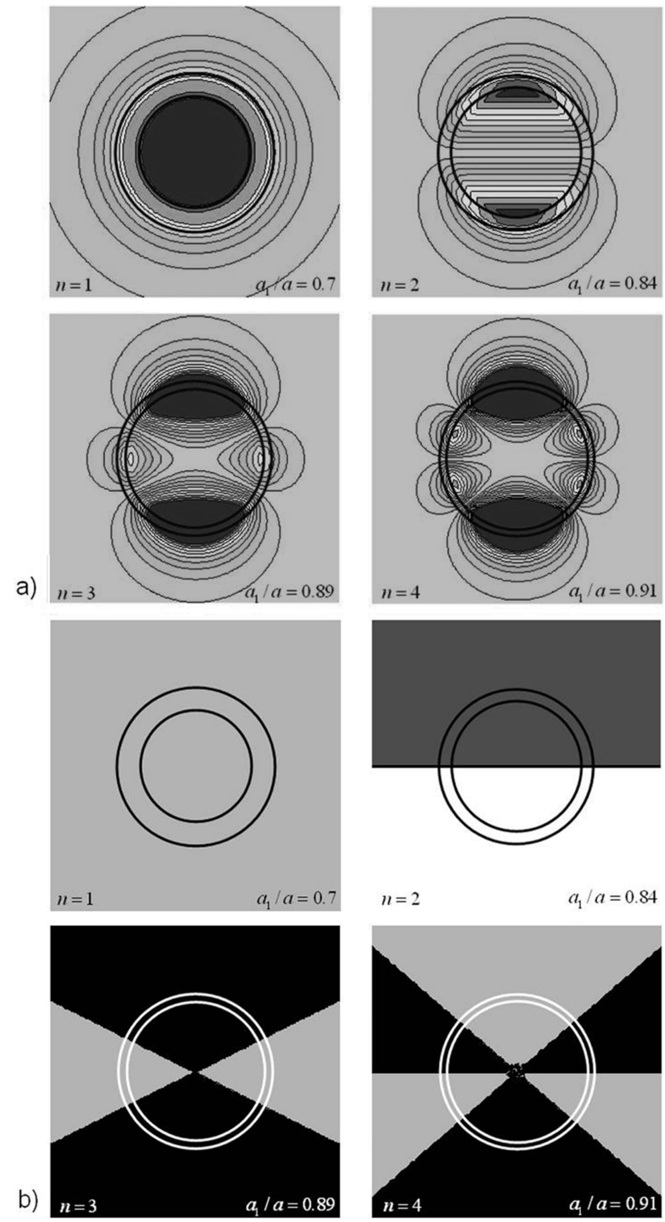

Fig. 2. (a) Magnitude and (b) phase of the near-zone electric field distributions for four compact, subwavelength spherical hollow shells of outer radius $a = \lambda_0/20$, with $\varepsilon_2 = -5\varepsilon_0$ for the shell and $\varepsilon_1 = \varepsilon_0$ for the core and the host background. For each n, the inner radius a_1 is chosen such that (2) is satisfied, in order to excite the different resonances of order n. (For the grayscale code, darker areas correspond to higher values of magnitude and phase. The phase plots are all normalized to the extremes of the phase interval). The feeding dipole is orthogonal to the plane of the figure (H plane).

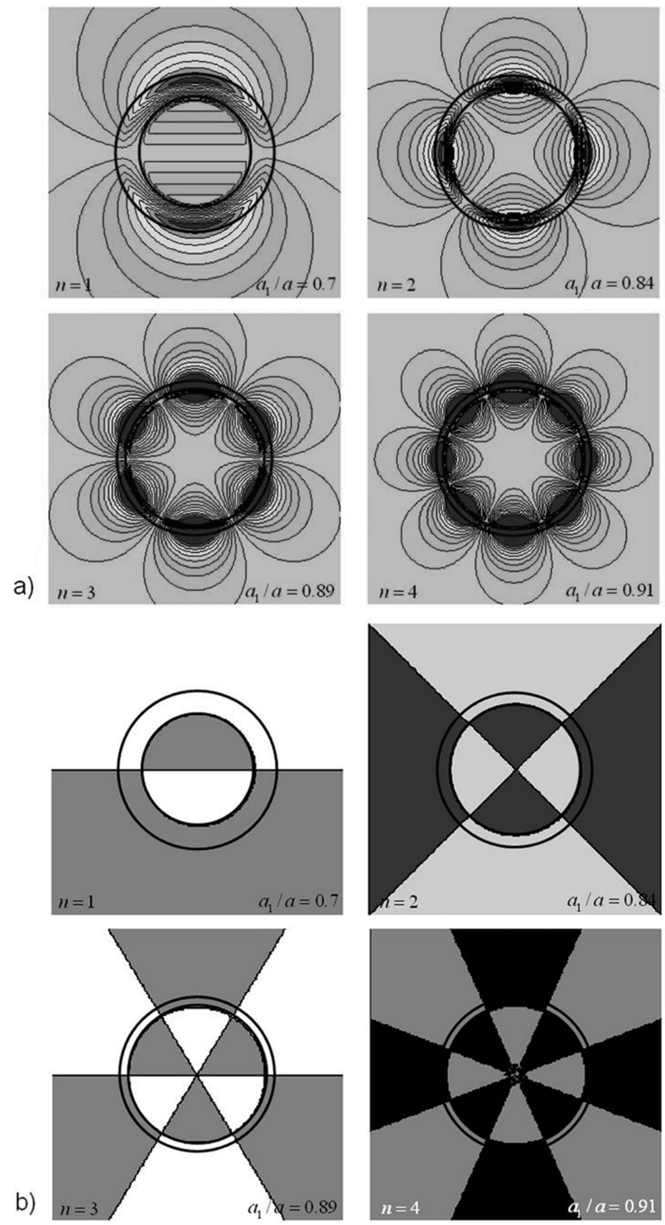

Fig. 3. Similar to Fig. 2, but in the orthogonal E plane: (a) magnitude and (b) phase of the near-zone magnetic field distributions for the same four compact, subwavelength spherical shells of Fig. 2.

tation of the nano-antenna, but one may notice how its angular distribution in the cross section shown in Fig. 2 is uniform inside and around the particle, as in the non-resonant case. This leads to the standard "donut-shape" radiation pattern from this object, which may be obtained if the feeding dipole source is located at its center. Changing the inner radius, however, one may in principle tune to the different resonances, which locally, in the near-field, look much more sharply varying both in amplitude and in phase as the order n is increased. Such increase also leads to higher Q factor, implying smaller bandwidths and higher sensitivity to the possible material losses and to variation of the design parameters. From Figs. 2 and 3, we can see how the field concentrates around the shell when the order n is increased, causing higher sensitivity to the properties of the narrow region where the field is localized.

Applying the reciprocity theorem, it is then possible to evaluate the radiation patterns of small sources, i.e., short dipoles, embedded in the near zone or inside such subwavelength structures, and to explore the radiation characteristics of these subwavelength systems. We clearly expect to obtain an increasingly more directive radiation pattern once the resonant order is increased, without a need to enlarge the resonant size of the structure, leading to possibility of having super-directive radiators. From the near-field configuration and the reciprocity theorem, it is evident that the higher order superdirective radiation

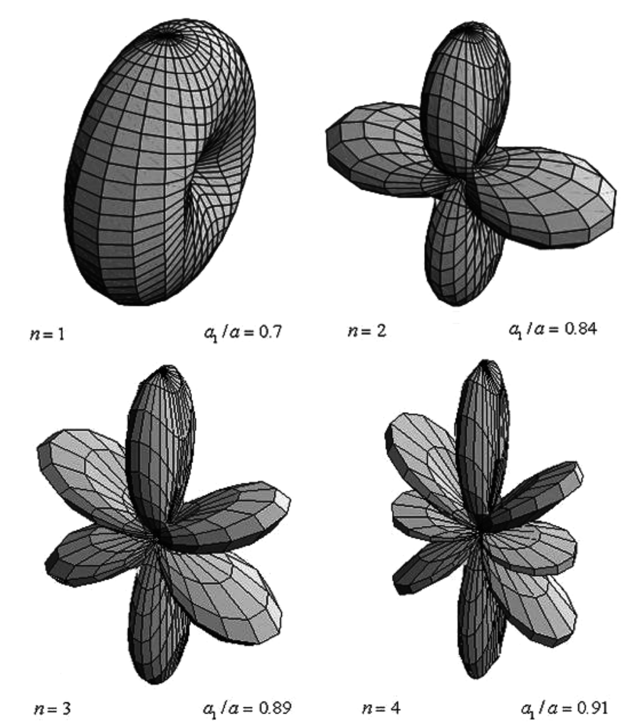

Fig. 4. Far field patterns for the subwavelength systems of Figs. 2 and 3, when they are driven by an infinitesimal dipole located at the interface of the shell and the core.

cannot be excited with a source placed at the center of the system (where in fact the field is zero for $n \geq 2$), and this is related to the fact that a certain gradient (i.e., spatial variation of the field) is needed in order to excite higher order resonances in the structure.[2] Placing a source at the interface between the core and the shell in this structure, as suggested in Fig. 1, is a viable method to excite such higher order resonances, and it may produce the maximum radiation gain for a fixed level of current on the infinitesimal source, as it can be easily shown analytically by calculating the position of the maxima of the near field distribution. These maxima occur at the interface between a positive-parameter material (in this case the host material), and a negative-parameter material (here the ENG shell), consistent with the analogous resonant phenomenon analyzed in the planar geometry in [15]. This also explains the localization of the field around these interfaces when the order n is increased, which by itself may offer interesting potential applications for subwavelength near-field localization.

The angular position of the source determines the angular distribution in the far field. The power radiation patterns $F(\theta, \phi)$ in the spherical coordinate system for any resonance of order n excited in the xz plane (i.e., by a dipole oriented in the x direction and placed off axis along z) may be written as

$$F(\theta,\phi) = \frac{\left[P_n^1(\cos\theta)\right]^2 \sin^2\phi + \left[\frac{d\,P_n^1(\cos\theta)}{d\theta}\right]^2 \cos^2\phi}{\sin^2\theta} \qquad (3)$$

where $P_n^m(x)$ are the associated Legendre polynomials [41].

[2]The spherical harmonic of order n, in fact, interacts with and it is excited by the $(n-1)$th gradient of the field at the location of the particle [42]. A uniform field, for instance, can excite only the $n = 1$ resonance. For reciprocity, the near-field of the $n = 1$ resonance is uniform around the origin [see Figs. 1(a) and 2(a)], whereas all the other higher order resonances have a certain gradient, with zero field at the particle center.

As the order n is increased, the beamwidth of the radiated maxima decreases and the overall directivity increases, consistent with the sharp variation in the near-field patterns shown in Figs. 2 and 3.

Fig. 4 shows the far-field radiation patterns for the particles of Figs. 2 and 3 considering an infinitesimal dipole placed at the interface between the metamaterial shell and the hollow core. It is evident from this figure how the beamwidth for higher order n decreases, together with an increase in the number of multiple beams radiated by the system. The overall directivity may be calculated as follows, since the maximum radiation is always obtained for any n at $\theta = 0$

$$\begin{aligned} D &= \frac{4\pi|F(0,\phi)|}{\int_0^\pi \int_0^{2\pi |F(\theta,\phi)|^2 \sin\theta d\phi d\theta}} = \\ &= \frac{4|F(0,\phi)|}{\int_0^\pi \frac{[P_n^1(\cos\theta)]^2 + [dP_n^1(\cos\theta)/d\theta]^2}{\sin\theta} d\theta} \\ &= \frac{2n+1}{2} \end{aligned} \qquad (4)$$

where we have used the following results, obtained after analytical manipulations from the properties of Legendre polynomials [43]

$$\begin{aligned} |F(0,\phi)| &= \frac{n^2(n+1)^2}{4} \\ \int_{-1}^{1} \frac{\left[P_n^1(x)\right]^2}{1-x^2} dx &= 2\int_0^1 \frac{\left[P_n^1(x)\right]^2}{1-x^2} dx \\ &= n(n+1) \\ \int_{-1}^{1} \frac{\left[d(P_n^1(x))/dx\right]^2}{1-x^2} dx &= \frac{n(n+1)(2n^2-1)}{2n+1}. \end{aligned} \qquad (5)$$

Interestingly enough, these values of directivity are higher than those evaluated in [2] for the same level of complexity of the antenna (i.e., the same maximum order n of multipole excitation), which was $2n/\pi$. This is due to the fact that the antennas considered here do not have a circularly symmetric radiation pattern, as in Chu's seminal work. The limitation on directivity described by (4), indeed, corresponds to these TM_{n1} spherical harmonics, analogous to the one evaluated in [2] for TM_{n0} modes.

The linear growth of directivity with the polar order n is also accompanied by a linear growth in the number of distinct radiated beams. In particular, in the $x-z$ plane (E-plane) there are in general $2n$ distinct beams and in the $y-z$ plane (H-plane) the number of maxima is $2(n-1)$, with the maximum at $\theta = 0$ in both planes. The amplitude of the other beams in the E plane smoothly decays moving away from the z axis, whereas the beams in the H plane are minor grating lobes (not visible in Fig. 4 because of their small relative amplitudes with respect to the main beam). The beamwidth at $\theta = 0$ may be expressed as $\mathrm{Beamwidth} \simeq ((\pi)/(2n) \times (\pi)/(n))$ on the two planes, which is exact for $n = 2$, and a good approximation for any $n > 2$. The corresponding beamwidth efficiency BE, defined as the ratio between the power radiated in the main beam (at $\theta = 0$) over the total radiated power, decreases on the E plane with an increase of n, due to the excitation of multiple highly-directive beams with nearly equal amplitude, and remains almost constant and equal to 0.5 in the H-plane, due to the low

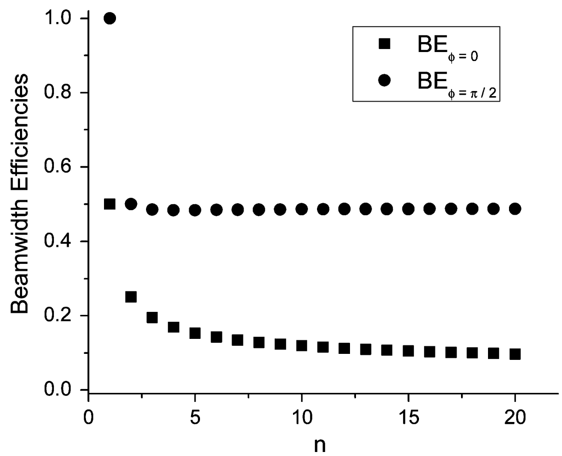

Fig. 5. Beamwidth efficiency for the main lobe (at $\theta = 0$) on the E and H planes varying the resonant order n.

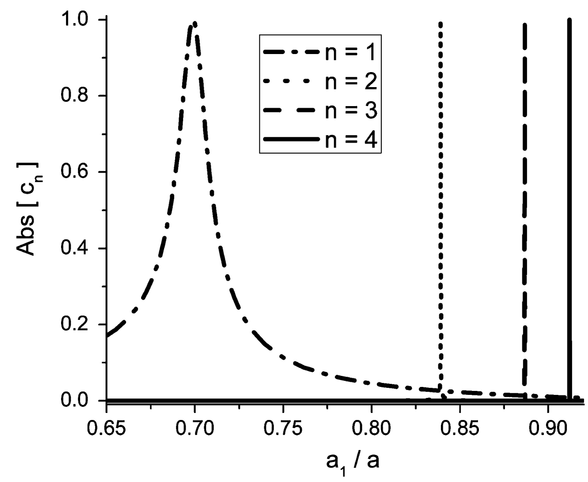

Fig. 6. Magnitude of the scattering coefficient c_n^{TM}, as defined in [19], for the structures of Figs. 2–4 versus the core-shell ratio of radii for a fixed outer radius.

level of grating lobes in this plane. A plot of these efficiencies in the two planes is reported in Fig. 5. It is worth noting that not only does the overall directivity increase with the resonant order n, but also the concentration of the radiated power in distinct narrow beams, with beamwidths much smaller than those expected by an aperture of such small size, grows much faster, which may be useful in different applications, as we discuss in Section VI.

This enhanced directivity phenomenon is clearly due to the sharply varying amplitude and phase of the near-field distribution inside the shell (compare Fig. 2 and 3 with Fig. 4 for the different orders n), somewhat analogous to what happens in a classic super-directive array with elements fed with opposite phases and sharply varying amplitudes in a very close space. The advantage of the subwavelength plasmonic core-shell here, however, is that the resonant mode of order n may be set up in the system "automatically" when it is excited by the dipole at the core-shell interface, much like exciting a resonant cavity tuned to the proper resonant mode of interest. One may argue that a similar mode may be obtained in a core-shell structure with conventional materials, when (1) is satisfied. However, with conventional materials, this can happen only when the size of structure is not small, but instead comparable with the effective aperture required for obtaining the corresponding level of directivity and beamwidth with a uniform illumination. On the other hand, the use of negative-parameter materials, like the shell in this proposed system, allows satisfaction of (1) [or its simplified approximate version in (2)] by exploiting a subwavelength plasmonic resonance of the structure while keeping its size small.

III. Bandwidth and Loss Limitations

Following (4), the overall directivity increases linearly with the order n of excitation and the beamwidth of the radiated beams decreases even faster, in principle providing the possibility for superdirective radiation in the cases under consideration. The price to be paid, however, relies on the resonance Q of such systems. The Q factor of the resonances indeed drastically increases with the resonant order n, and this is clearly evident from the plot of the resonant order amplitudes in terms of the ratio of radii of the core-shell lossless particles, reported in Fig. 6. The figure shows how the scattering coefficient c_n^{TM}, as described in [19], whose absolute maximum is unity at the resonance in a lossless particle, varies when the inner core radius is modified. By reciprocity, this is consistent with the different radiation patterns predicted in Fig. 3 when the different resonances are excited. This implies a decrease in the bandwidth of operation and sensitivity to material losses when these devices are utilized as directive radiators, i.e., higher directivity and narrower beamwidth may be achieved at the cost of lower bandwidths and less robustness to loss and parameter changes. Although Fig. 6 refers to a variation of the ratio of radii at a fixed frequency, rather than directly to a frequency shift, parameter changes have similar effects in detuning the nano-antenna operation. A more extensive discussion of the Q factor of these systems in terms of frequency bandwidth is given in the following, where material dispersion is also considered.

As already mentioned, the presence of material losses may reduce or drastically affect the possibility of inducing such higher order resonances in a subwavelength particle. When considering realistic systems in which losses play a role, due to the high Q of these resonances the sensitivity to losses may be very high, and the peaks of higher order radiation may be overcome by the presence of the residual dipolar or lower-order terms. As derived in [44], the possibility of exciting nano-resonances in subwavelength systems holds as long as the absorption in the particle is negligible, which quantitatively can be written as

$$-\mathrm{Im}[\varepsilon/\varepsilon_0] \ll (k_0 a)^{2n+1} \frac{n+1}{n^2[(2n-1)!!]^2} \tag{6}$$

consistent with the fact that the Q factor increases with n accordingly. Such a condition on the material absorption shows that it will be challenging to excite higher order resonances for a small sphere, unless extremely low-loss plasmonic materials are available or their size is adequately increased (we should point out, however, that in exciting higher order plasmonic resonances we may not be necessarily interested in designing extremely subwavelength antennas, but instead these concepts may be applied in order to conceive antennas whose size, not being necessarily subwavelength, is still much smaller

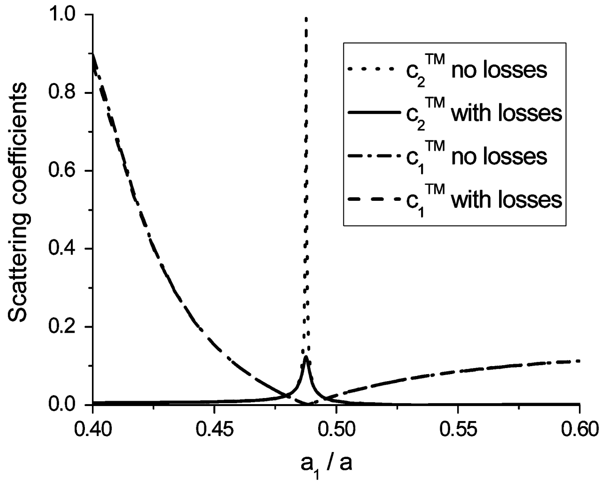

Fig. 7. Dependence of the scattering coefficient c_1^{TM}, relative to the dipolar radiation, and the c_2^{TM}, associated with the quadrupolar field, for an antenna with $\varepsilon_1 = 8\varepsilon_0, \varepsilon_2 = -6.5\,\varepsilon_0$ and $a = \lambda_0/10$. When losses are introduced $\varepsilon_2 = (-6.5 - j\,0.01)\varepsilon_0$.

than the size required to obtain analogous levels of directivity and beamwidth with uniformly illuminated apertures). In other words, realistic designs in which the beamwidth is narrower than the one expected from an aperture of the same size with uniform field distribution may be designed following the ideas presented here.

In this sense, a useful technique to reduce the sensitivity to losses may be suggested by the combined excitation of a higher order mode resonance together with the simultaneous cancellation of lower-order terms, following the technique we have proposed in [45] for canceling a given multipolar contribution to the scattering using metamaterials. For instance, a core-shell system with $\varepsilon_1 = 8\varepsilon_0, \varepsilon_2 = -6.5\,\varepsilon_0$ and $a = \lambda_0/10$ would have the interesting property of supporting the quadrupole resonance for the same exact ratio of core-shell radii at which the dipolar radiation is totally cancelled. This would create a subwavelength quadrupolar nano-antenna with relatively less sensitivity to losses, as Fig. 7 shows. The solid and dotted lines (representing lossy and lossless cases, respectively) correspond to the coefficient relative to quadrupolar radiation, which is achieved for this antenna at the ratio $a_1/a = 0.49$, at the same exact ratio for which the dipolar radiation coefficient (dashed and dot-dashed lines) is identically zero, due to the cancellation effect introduced in [45]. The plot shows how the radiation is still dominated by the c_2^{TM} coefficient, with the lower-order term c_1^{TM} being identically zero, even when presence of some losses is considered, which in this subwavelength design drastically affects the Q factor of the quadrupolar resonance and reduces the radiated power of the system and its efficiency. In the lossy case, the permittivity of the shell material considered in the simulation is $\varepsilon_2 = (-6.5 - j\,0.01)\varepsilon_0$. At some level, this concept will again be limited when losses are too high or the size of the antenna becomes too small, and then the quadrupolar radiation will become negligible. However, a quadrupolar beamwidth from a source with aperture extension smaller than $\lambda_0/2$ is expected in this configuration, and it may not otherwise be obtainable using different techniques.

IV. Feeding the Nano-Antennas and Q-Factor

In order to forecast a practical realization, the problem of feeding these nano-antennas is of extreme importance. Even if it is clear that the maximum gain for a fixed current amplitude is obtained when the source is placed near the interface, the interest lies in the possibility of matching a real feed network for this radiating system in order to avoid a high return loss. To this end, we have simulated an example of these radiators with finite integration technique software [46], to explore the full-wave behavior of this type of radiators in a realistic environment, with possible imperfections in the shape and considering the presence of a realistic feed.

Fig. 8 reports the numerical results for a subwavelength silver spherical shell fed at the interface $r = a_1$ by a short dipole with a $50\,\Omega$ port at the frequency $f_0 = 675$ THz, i.e., at the quadrupolar resonance (for $n = 2$) of the system. The geometry considered here has $a_1 = 42$ nm, $a = 46$ nm, $\varepsilon_1 = \varepsilon_0$ and $\varepsilon_2 = \varepsilon_{\mathrm{Ag}}$. We have assumed a realistic Drude model for silver with $\varepsilon_{\mathrm{Ag}} = (\varepsilon_\infty - (\omega_p^2)/(\omega^2 - j\omega\omega_\tau))\varepsilon_0$ with $\omega_p = 2\pi \cdot 2175\mathrm{THz}, \omega_\tau = 2\pi \cdot 4.35\mathrm{THz}$ and $\varepsilon_\infty = 5$, considering material dispersion and realistic losses, following [47]. In our numerical model the feed is represented by an electric dipole constituted by a 5nm idealized conducting wire with a $50\,\Omega$ port feed at its center. The free-space wavelength at the frequency of interest is 440 nm.

The near-field pattern shown in Fig. 8(a) (electric field distribution on the H plane orthogonal to the exciting dipole) and in Fig. 8(b) (magnetic field distribution on the E plane containing the exciting dipole) is consistent with the plots in the top right panels of Figs. 2 and 3, despite the numerical discretization "noise" of the software, the presence of a real feeding network and of material loss and dispersion. Likewise, the far-field gain pattern of Fig. 8(c) does also agree with far-zone pattern shown in the top right panel of Fig. 4. Although the matching is not ideal due to the high reactive (stored) field excited around the feed, the quadrupolar pattern is dominant and clearly recognizable in the near as well as in far field. In our simulations achieving good matching and high efficiency was not the main issue, and therefore they have not been properly optimized, since we wanted just to show the possibility of exciting such highly-directive resonances in subwavelength structures with available materials at optical frequencies. We speculate however that with a proper optimization of the feeding point and an adequate modeling of the optical feeding technique, which may be constituted by a near-field scanning optical microscope (NSOM) exciting a small nano-particle placed close to the resonant nano-antenna, one may be able to further improve the matching characteristics of this antenna, similar to what was found in [29] for the dipolar resonance at lower frequencies. These problems are under consideration and they will be the subject of further investigation.

These numerical results may offer the possibility of synthesizing and feeding infrared and optical nano-antennas with enhanced directivity following the concepts studied here, by building nano-shells of noble metals with proper design to provide the higher order resonances predicted by (2). Some experiments in this sense have already been performed by Halas and her group on shells of metal, even though in that case the

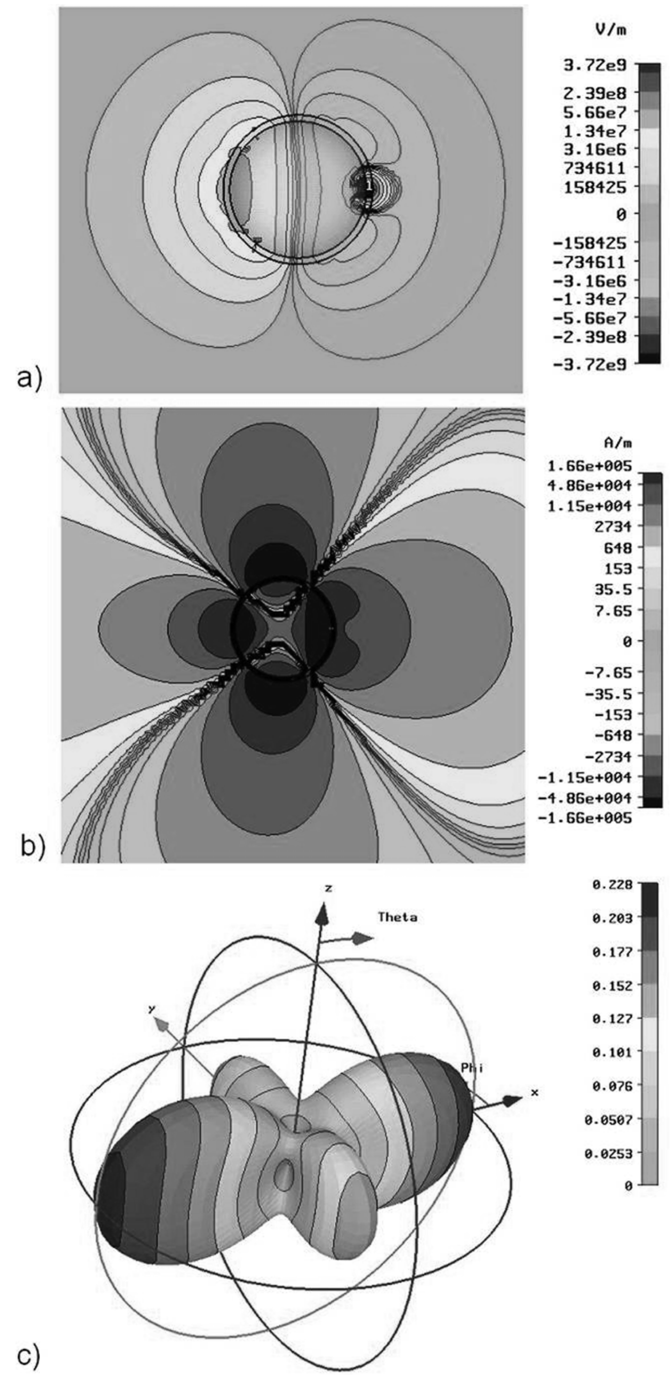

Fig. 8. Numerical simulation of the quadrupolar shell of Figs. 2–4 utilizing a suitable Drude model for the silver used to synthesize the ENG shell at optical frequencies, feeding the nano-antenna of Fig. 1 with a 50- Ω port at the inner interface between core and shell. (a) Near-field electric distribution on the H-plane (consistent with Fig. 2). (b) Near-field magnetic distribution on the E-plane (consistent with Fig. 3). (c) Far-field gain pattern (consistent with Fig. 4).

particles were not as subwavelength in dimensions [34]–[37] as we propose here. Nevertheless, however, the quadrupole contribution has been isolated here even in the subwavelength case and expectedly these concepts appear to be limited only by the intrinsic material losses. At lower frequencies, or when natural materials are not available with the desired properties, proper synthesis of metamaterials may be envisioned.

We note how the Drude model considered in the example of Fig. 8 for the permittivity of silver takes into account adequately the necessary conditions for passive and causal materials satisfying the condition $\partial(\omega\varepsilon)/\partial\omega > 0$ even in the regions of frequency where $\varepsilon < 0$ [48]. The frequency dispersion of the materials should indeed be taken into account (as we have) in any consideration on the realistic performance of these radiators, as we discuss in the following.

Considering the Q of these structures, material dispersion plays a fundamental role. The resonance Q of an antenna is strictly related, as in any resonant structure, to the inverse of the bandwidth of operation [12]. Since for driving an antenna one in general needs to compensate the lack of balance in the near-zone electric and magnetic energy densities with a proper feeding network, the standard definition of an antenna Q for an ideal lossless antenna is given by [6]

$$Q = \frac{2\omega \max[W_e, W_m]}{P_{\mathrm{rad}}} \tag{7}$$

where W_e and W_m are, respectively, the electric and magnetic energy densities associated with the non-radiating stored fields around the antenna and P_{rad} is the radiation power. Since the resonance of an antenna is reached when $W_e = W_m$, it is in fact assumed that at the operating frequency the matching network adds a reactive energy in such a way to bring the system into resonance.

The increase of the Q of an antenna with its inverse size and its angular complexity (corresponding here to the multipole order n) has been discussed for decades [1]–[12], and the classical result that for omnidirectional (on their H plane) antennas the Q grows dramatically when the directivity is increased beyond the limit represented by a uniform field distribution on the aperture of the same size (which is $D = 4a/\lambda_0$) is very well accepted [1].

The scattered power from the induced multipoles in the nano-antenna of Fig. 2 due to an incident plane wave with electric field $\mathbf{E} = E_0 e^{-jk_0 z}\hat{\mathbf{x}}$ is easily calculated with the results of (4): for the antenna at resonance dominated by the multipole of order n, the far-field scattered power density is given by

$$P(\theta,\phi) = \frac{|E_0|^2}{2\eta_0}|F(\theta,\phi)| \tag{8}$$

and therefore

$$P_{\mathrm{rad}} = \frac{|E_0|^2 n^2 (n+1)^2}{\eta_0 [2n+1]}. \tag{9}$$

The electric and magnetic stored energy densities associated with the non-radiating fields, that contribute to the numerator of (7) may be calculated following [6] (in this case the limitation of this method for the multiple mode scenario, which was pointed out in [11], does not hold, since the excited resonant mode is only one). Since we are under the hypothesis of resonant excitation of one multipole, which dominates the near as well as the far-field of the structure, we should integrate all over the space the corresponding electric and magnetic energy densities calculated from the field distributions as given in [19], subtracted from it the contributions to radiation, given by P_{rad}/c, (c is the velocity of light in free space) [6]. This represents the stored energy density for each multipole, concentrated around the antenna, and rapidly decaying when r is increased. If we limit ourselves to the contribution of this integral given in the outer space $r > a$, the result for the Q is given by formula (8) in [6], since this is independent of the azimuthal order m (there it was calculated for $m = 0$, whereas here $m = 1$). In the limit of small antennas, i.e., $k_0 a < 1$, which is of interest here, we get

$$Q = \frac{n(2n+1)!!^2}{(k_0 a)^{2n+1}} \tag{10}$$

that confirms how the Q factor increases dramatically when the antenna size is reduced for a fixed directivity, or when the directivity is increased for a fixed size, consistent with [11] and with the result in (6).

In (10) the contribution of the energy stored inside the antenna, i.e., in the region $r < a$, has not been considered, and that is why (10) represents a generalized lower limit for the super-directive antennas presented here (and applies also to the case $n = 1$, which is usually referred to as the classic Chu limit for small antennas). Although it is possible to evaluate the inner contribution to the energy density in an exact way (since the field distribution is written in closed form [19]), we do not enter into the details of this calculation here. It is sufficient to note for now that if on the one hand the region $r < a$ is electrically small in extension, on the other hand the field amplitude there reaches its peak values, rapidly decaying outside the shell. This value may get very high when a larger n is selected, as seen from the plots in Fig. 2. It is interesting to note, therefore, that the quantities $|\mathbf{E}|^2$ and $|\mathbf{H}|^2$, as well as their integral all over the nano-antenna volume, may reach values comparable in magnitude with those obtained for the outside region and used to evaluate (10).

The energy density for lossless dispersive metamaterials is in fact proportional to $[\partial(\omega\varepsilon)/\partial\omega]\,|\mathbf{E}|^2$, which is always a positive quantity [48] and it adds a further contribution to the numerator of (7). For lossy and dispersive materials the correct definition of energy density has been derived in [49], and it is still an always-positive quantity. It has been shown in [30], for instance, how for the case $n = 1$ and a Drude-dispersive material the resonance in the quasi-static limit of $k_0 a \ll 1$ has a Q factor equal to 1.5 times the Chu limit. This can be extended to higher order resonances, and it will be the subject of further investigations, but for now it is clear that (10) still represents a lower limit to the Q factor of the super-directive antennas and it should be taken into account in any effort to practically realize them. In the evaluation of the Q factor, losses may also play a role, lowering down the high Q, possibly together with the superdirective properties of these structures.

V. Further Limitations and Alternative Solutions

In the previous sections, we have discussed the ideal case in which the required material response for synthesizing subwavelength radiators with enhanced directivity is already available at the desired frequency of interest, showing how under such assumption this possibility may be physically sound and realistic. In this sense, the use of plasmonic infrared and optical materials or metamaterials may allow inducing the necessary near-field configurations in a natural way, in contrast with other superdirective techniques. However, up to now we have assumed that such special materials with the required negative parameters exist and they can be tailored in the specific way needed for the purpose at the desired frequency of operation. When we work at frequencies for which some materials do not already naturally possess an isotropic permittivity with the required value, we may be required, however, to synthesize them artificially. Currently the metamaterial science and technology is progressing fast, but still a brief discussion on the current possibility of manufacturing this setup at different frequencies is suitable here. Super-directivity generally requires apertures and antennas that are small, possibly smaller than the wavelength of operation. When dealing with metamaterials, on the other hand, the inclusions composing the medium should be themselves much smaller than both the wavelength and the volume under consideration. Basically, the inclusions should act as the artificial "atoms" and "molecules" composing the metamaterial, and therefore it may be less suitable to consider as a "bulk medium" a composite structure made out of a limited number of relatively large inclusions. This opens up the question of the practical realizability of the radiators described here, particularly for lower frequencies, i.e., microwaves, where plasmonic materials are not naturally present. Even though the design of electric resonant inclusions may be relatively straightforward, and it may offer better performance in terms of the range of negativity that the effective permittivity of the bulk metamaterial may reach, to the best of our knowledge at the present time, efforts to build them in a subwavelength scale in the microwave regime have been more intricate than the design of subwavelength magnetic resonant inclusions. In other words, MNG materials may rely on inclusions that may be made at smaller scale (albeit with narrower bandwidth).

Clearly, the theory previously presented, which was tailored for ENG materials, may be easily applied to MNG materials in the same way: by duality such systems would work equally well by exciting TE_{n1} resonant modes. Also Q and directivity do not depend on the polarization of operation. However, the dispersion of MNG in general follows a Lorentz model, rather than a Drude dispersion, which has generally a narrower bandwidth of negativity of the parameters and a sharper variation with frequency. The problem of "overlapping" of the different resonant frequencies for different polar orders n might arise, thus not allowing the effective presence of an isolated higher order peak. It may be shown, however, (not reported here for sake of brevity) that resonant peaks of different orders may remain distinct also when MNG metamaterials are employed, supporting the present theory also in this possibility of realization.

Dealing with realization issues, another important aspect may be encountered by the possible anisotropy of the employed materials. This is particularly important when artificial materials are considered. A totally 3-D isotropic metamaterial may be harder to realize, even though efforts are being spent in realizing isotropic negative effective constitutive parameters with different technologies (e.g., [50], [51]). In the Appendix we analyze in detail the problem of an anisotropic spherical core-shell system in the quasi-static limit, in order to study the dependence of the previous results on the assumption of an isotropic shell. We show in particular the conditions on the material anisotropy under which results similar to those presented in the previous sections may be applied to anisotropic nano-antennas. These results may be similarly extended to more complex geometries, i.e., elliptical or spheroidal.

When infrared and optical nano-antennas are designed using natural materials, the material anisotropy is not expected to play a dominant role, since noble metals and polar dielectrics are not usually characterized by relevant anisotropies. An aspect to be aware of, however, when designing such nano-antennas is that the permittivity of thin shells of such materials may not necessarily coincide with the tabulated values for the bulk material [53]. Even though change from the bulk values is expected only when the thickness of the material samples is comparable with the mean free path of the conduction electrons in the material, this may represent a limitation of the present analysis in the minimum size of the nano-antennas that may be designed. For the numerical examples presented here, however, the geometries employed are above the limits for which this problem may arise. Scaling down the size further, generally the material loss factor becomes dominant in the permittivity response of the nano-particles and such nano-resonances may be strongly affected.

To conclude this section, we mention a few words about the possible realization of a highly-directive nano-antenna with current technology. As already highlighted, at microwave frequencies the use of metamaterials may be forecasted. The presence of tiny inclusions, much smaller than the wavelength of operation, would however be necessary in order to squeeze the dimensions of the radiator and simultaneously obtain the required effective medium properties for the material. In this scenario, the use of MNG metamaterials might seem to be more plausible and promising with the current technology, due to the possibility of reducing the inclusion size for MNG construction well below the wavelength of operation by employing multiple split-ring resonators (SRR) or capacitively loaded SRRs or similar inclusions [54]. At higher frequencies, more of interest for the present paper, natural plasmonic materials and their combinations may be proposed for this purpose. In this sense, covering dielectric nanospheres with thin nano-scale layers of metal, i.e., gold shell over silicon-dioxide spherical structures of the size of a few tens of nanometers [34] is indeed achievable with the current technology.

VI. SUPER-RESOLUTION DEVICE FOR OPTICAL DETECTION OF NANO-PARTICLES

As a potential application of such nano-devices at infrared and optical frequencies, here we discuss of the possibility of employing these higher order resonances for a super-resolution device. Super-resolution, as defined in [10], is obtained when the beamwidth of a radiated beam is narrower compared with that of the Rayleigh limit, given by $\lambda_0/(2a)$, where $(2a)$ is the aperture length (in this case the particle diameter). As we consider higher multipolar order, the directivity is increased linearly with n, as shown in (4), and the beamwidth of the main beam decreases even faster, as already noticed. This opens up interesting possibilities for super-resolving subwavelength details in the far-field, as discussed in [6] and in the introduction of this paper.

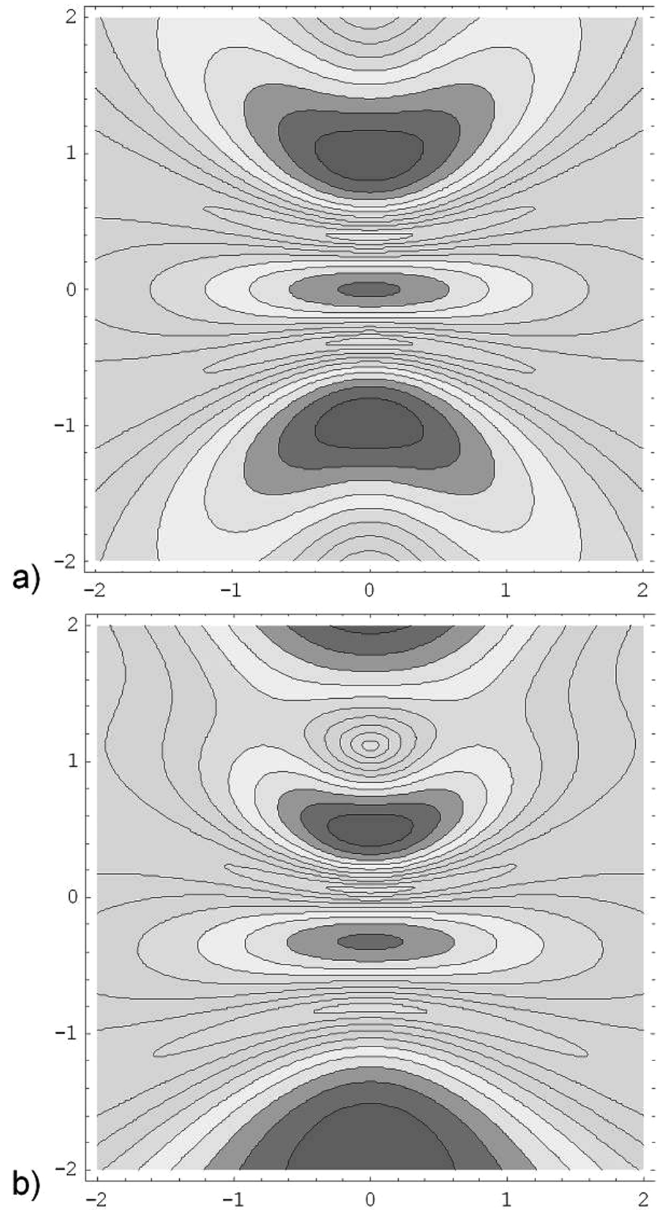

Fig. 9. Subwavelength localization of an infinitesimal nano-particle placed inside the superdirective antenna of Fig. 1, bottom right. The two panels report the field amplitude on a plane located at a distant L from the antenna center, when the nano-antenna operates at its $n = 4$ resonance, showing how the far-field pattern is noticeably shifted when an infinitesimal source at the interface of the core and shell is moved angularly by the amount of $0.028\lambda_0$ from (a) to (b). The distances shown on the two axes are normalized to distance L.

Due to the symmetry of the radiation pattern and the presence of closely spaced multiple beams, the setup presented here does not allow to resolve a subwavelength detail in a complex image displaced in the near-field of the nano-antenna, but it is indeed possible to distinguish clearly the position of a subwavelength object in the near-field, or inside the sphere, with a detail that is much smaller than half wavelength. Let us consider as an example an infinitesimal source of radiation at the core-shell interface of the sphere of Fig. 1, at the $n = 4$ resonance (bottom right panel). Since the inner radius of the system is $a_1 = 0.091\lambda_0$, a slight angular movement of the object (i.e., the source) would tilt the whole radiated pattern in the far-field. Fig. 9, as an example, shows the case of a nano-object placed at the interface between the core-shell, moved by an angle $\alpha = \pi/10$ from the position $(\theta = \pi/2, \phi = 0)$ to the position $(\theta = \pi/2 + \alpha, \phi = 0)$, and the far-field pattern is measured at a distance L on a planar screen orthogonal to the x axis. We note how an ultra small subwavelength change in the position of the object (the absolute shift in position is of $0.028\lambda_0$) generates on the far-field image plane a noticeable shift of the measured peak, which may be of several wavelengths, depending on L (the axes show values of the coordinates in the image plane normalized to L). The presence of the other sidelobe beams in this configuration limits this effect, particularly when multiple sources are present, and therefore a real subwavelength imager in the far-field is not possible in this configuration. However, we speculate that with other configurations involving similar concepts, but with less symmetry, i.e., apertures filled with metamaterials or different non symmetrical shapes for the antennas, this super-resolution effect might also lead to the possibilities for subwavelength imaging of complex objects. This is currently under investigation in our group.

VII. Conclusion

To conclude, we have presented here some theoretical results on how metamaterial resonator subwavelength antennas may be employed to synthesize efficient superdirective radiators. The possibility of inducing sharply varying fields in a very small region when higher order resonant modes are excited in a subwavelength particle is here exploited to conceptualize a superdirective antenna which might overcome the standard limitations of the feeding system present in classic superdirective arrays or apertures. Details of the physics behind this phenomenon and practical limitations have been described and analyzed with a certain detail, together with some ideas for practical implementation and a potential application of this technique to realize a super-resolving device. It should be noted that similar concepts could be applied to such antennas as receiving systems. A beam captured from a small angular direction may indeed be concentrated in a small subwavelength region of space and detected with a proper receiver embedded in the antenna. Moreover, in this case the angular position of the transmitter may exhibit subdiffraction characteristics and sensitivity.

Appendix

We assume here to have a quasi-static impinging electric field, directed along z, and exciting the core-shell geometry of Figs. 2–3. The corresponding potential $V_i(r,\theta) = -E_0 r^n P_n(\cos\theta)$, with $P_n(x)$ being the Legendre polynomial, may excite the "quasi-static" nth order resonance, similarly to the previous analysis. Here, however, we suppose that the two layers have permittivities $\varepsilon_{\mathrm{core}}$ and $\varepsilon_{\mathrm{shell}}$, respectively, in general full tensors to take into account of the possible anisotropy (an analogous analysis may be carried out for a magnetostatic potential and for the permeability tensors by invoking duality).

If the potential inside the core is expressed as $V_{\mathrm{core}} = c_{\mathrm{core}} r^n P_n(\cos\theta)$, the one in the shell as $V_{\mathrm{shell}} = [c_{\mathrm{shell}} r^n + d_{\mathrm{shell}} r^{-n-1}] P_n(\cos\theta)$, and the one in the region outside as $V_{\mathrm{out}}(r,\theta) = [-E_0 r^n + c_{\mathrm{out}} r^{-n-1}] P_n(\cos\theta)$, then the boundary conditions at the two interfaces may be matched to give the following conditions for the resonance:

$$\begin{aligned}
& c_{\mathrm{core}} a_1^{n-1} = c_{\mathrm{shell}} a_1^{n-1} + d_{\mathrm{shell}} a_1^{-n-2} \\
& c_{\mathrm{shell}} a^{n-1} + d_{\mathrm{shell}} a^{-n-2} \\
& = -E_0 a^{n-1} + c_{\mathrm{out}} a^{-n-2}. \\
& c_{\mathrm{core}} a_1^{n-1} \left[n\varepsilon_{\mathrm{core}}^{rr} P_n(\cos\theta) \right. \\
& \quad \left. - \sin\theta \varepsilon_{\mathrm{core}}^{r\theta} P_n'(\cos\theta) \right] = \\
& = c_{\mathrm{shell}} a_1^{n-1} \left[n\varepsilon_{\mathrm{shell}}^{rr} P_n(\cos\theta) \right. \\
& \quad \left. - \sin\theta\, \varepsilon_{\mathrm{shell}}^{r\theta} P_n'(\cos\theta) \right] + \\
& \quad + d_{\mathrm{shell}} a_1^{-n-3} \left[(-n-2)\varepsilon_{\mathrm{shell}}^{rr} P_n(\cos\theta) \right. \\
& \quad \left. - \sin\theta\, \varepsilon_{\mathrm{shell}}^{r\theta} P_n'(\cos\theta) \right] \\
& \varepsilon_0 P_n(\cos\theta) \left[E_0 a^{n-1} n - c_{\mathrm{out}} a^{-n-3}(n+2) \right] = \\
& = c_{\mathrm{shell}} a^{n-1} \left[n\varepsilon_{\mathrm{shell}}^{rr} P_n(\cos\theta) \right. \\
& \quad \left. - \sin\theta\, \varepsilon_{\mathrm{shell}}^{r\theta} P_n'(\cos\theta) \right] + \\
& \quad + d_{\mathrm{shell}} a^{-n-3} \left[(-n-2)\varepsilon_{\mathrm{shell}}^{rr} P_n(\cos\theta) \right. \\
& \quad \left. - \sin\theta \varepsilon_{\mathrm{shell}}^{r\theta} P_n'(\cos\theta) \right].
\end{aligned} \tag{A.1}$$

In these formulas, $\varepsilon_{\mathrm{core}}^{rr}$ and $\varepsilon_{\mathrm{core}}^{r\theta}$ are the first two elements of the first row of $\varepsilon_{\mathrm{core}}$, and similarly for $\varepsilon_{\mathrm{shell}}$, as written in the spherical reference system (r,θ,ϕ). They can be in general functions of θ and ϕ. By inspection you may notice that when isotropic and homogeneous media are considered, or more in general when the sufficient conditions

$$\begin{aligned}
\varepsilon_{\mathrm{core}}^{r\theta} = \varepsilon_{\mathrm{shell}}^{r\theta} = 0 \,\text{and}\, \frac{\partial \varepsilon_{rr}^{\mathrm{core}}}{\partial\theta} &= \frac{\partial \varepsilon_{rr}^{\mathrm{core}}}{\partial\phi} \\
= \frac{\partial \varepsilon_{rr}^{\mathrm{shell}}}{\partial\theta} &= \frac{\partial \varepsilon_{rr}^{\mathrm{shell}}}{\partial\phi} = 0
\end{aligned} \tag{A.2}$$

are satisfied, the equations lead to an algebraic system independent of space variables, that grants for the resonance the usual conditions for the resonances

$$\frac{a_1}{a} \simeq \sqrt[2n+1]{\frac{\left[(n+1)\varepsilon_0 + n\varepsilon_{rr}^{\mathrm{shell}}\right]\left[(n+1)\varepsilon_{rr}^{\mathrm{shell}} + n\,\varepsilon_{rr}^{\mathrm{core}}\right]}{n(n+1)\left(\varepsilon_{rr}^{\mathrm{shell}} - \varepsilon_0\right)\left(\varepsilon_{rr}^{\mathrm{shell}} - \varepsilon_{\mathrm{core}}^{rr}\right)}} \tag{A.3}$$

analogous to (2) but in which the radial permittivities take the place of the corresponding homogeneous values. Notice that the necessary conditions (A.2) are not easily obtainable in the spherical geometry, unless the medium is isotropic. Even a simple homogeneous uniaxial medium with a straight optical axis, for instance, has necessarily a nonzero $\varepsilon^{r\theta}$. If the optical axis is along z, which is the direction of the incident field, for instance, i.e., $\boldsymbol{\varepsilon} = \varepsilon_t(\hat{\mathbf{x}}\hat{\mathbf{x}} + \hat{\mathbf{y}}\hat{\mathbf{y}}) + \varepsilon_z \hat{\mathbf{z}}\hat{\mathbf{z}}$, in spherical coordinates $\varepsilon^{rr} = \varepsilon_t \sin^2\theta + \varepsilon_z \cos^2\theta$ and $\varepsilon^{r\theta} = (\varepsilon_t - \varepsilon_z)\sin(2\theta)/2$. An example of metamaterial with the required "radial" permittivity anisotropy may be possibly obtained by bending a uniaxial material in such a way that its axis of anisotropy is locally directed in the radial direction at every point.

The necessary and sufficient conditions for the induced resonances to be described by (A.3) is that the last two equations in (A.1) lose their dependence on the spatial variables, and this may be obtained in some special cases. For instance, when the anisotropy is only in the inner core, the condition becomes

$$\left[n\varepsilon_{\text{core}}^{rr}P_n(\cos\theta) - \sin\theta\varepsilon_{\text{core}}^{r\theta}P_n'(\cos\theta)\right] \propto P_n(\cos\theta) \quad \text{(A.4)}$$

which may be satisfied for $n = 1$ by a simple uniaxial material with optical axis directed along z, as the one previously considered. This is physically explained by the fact that the field induced in an anisotropic sphere is uniform and directed along its optical axis for the $n = 1$ quasi-static resonance, provided that also the impinging field is directed along the same direction [52]. Clearly in this case the anisotropy would not play a significant role. In this case, in fact, the quasi-static condition for the resonance becomes $\varepsilon_z = -2\varepsilon_0$.

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