Unconventional Fano effect and off-resonance field enhancement in plasmonic coated spheres

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We investigate light scattering by coated spheres composed of a dispersive plasmonic core and a dielectric shell. By writing the absorption cross-section in terms of the internal electromagnetic fields, we demonstrate it is an observable sensitive to interferences that ultimately lead to the Fano effect. Specially, we show that unconventional Fano resonances, recently discovered for homogeneous spheres with large dielectric permittivities, can also occur for metallic spheres coated with single dielectric layers. These resonances arise from the interference between two electromagnetic modes with the same multipole moment inside the shell and not from interactions between various plasmon modes of different layers of the particle. In contrast to the case of homogeneous spheres, unconventional Fano resonances in coated spheres exist even in the Rayleigh limit. These resonances can induce an off-resonance field enhancement, which is approximately one order of magnitude larger than the one achieved with conventional Fano resonances. We find that unconventional and conventional Fano resonances can occur at the same input frequency provided the dispersive core has a negative refraction index. This leads to an optimal field enhancement inside the particle, a result that could be useful for potential applications in plasmonics.

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1. INTRODUCTION

The Fano resonance, discovered in the realm of atomic physics by U. Fano in 1961 [1], is one distinctive characteristic of interacting quantum systems. The unique, asymmetric Fano lineshape has its origin in the wave interferences between a narrow discrete resonance and the continuum or a broad resonance. As a signature of quantum interference, the Fano effect has been extensively investigated in electronic transport at the nanoscale, in systems such as quantum dots, quantum wires, and tunnel junctions (for a review see Ref. [2]).

Being an interference phenomenon, Fano resonances also manifest themselves in classical optics. Historically, the first observation of a Fano resonance in optics was probably the Wood’s anomaly [3]. With the advent of metamaterials and plasmonic nanostructures, the Fano effect has recently become an important tool to control electromagnetic field. Fano resonances were shown to be at the origin of the so-called “dark-states” in light scattering by bound levels and their radiative decay is equivalent to the tunneling process from such levels [9]. The interference between the incident and re-emitted radiation gives rise to either enhancement (constructive interference) or suppression (destructive interference) of the electromagnetic field. Fano resonances were shown to be at the origin of the so-called “dark-states” in light scattering by coated spheres, where the local electromagnetic field enhancement occurs in off-resonant regions [10]. Light scattering by homogeneous particles can also exhibit unconventional Fano resonances in the extinction cross-section beyond the Rayleigh approximation; they result from the interference between different electromagnetic modes with the same multipole moment [11]. This contrasts to conventional Fano resonances, which arise from the spectral overlap of broad and narrow electromagnetic modes with different values of multipole moment [11].

In light scattering by small particles, Fano resonances are also known to play an important role. Despite its long history since the pioneer work of Mie in 1908 [5], light scattering still reveals challenging surprises, and many among them are related to the Fano effect. Indeed, anomalous light scattering phenomena such as giant optical resonances with an inverse hierarchy (quadrupole resonance is stronger than the dipole one), near-field pattern exhibiting vortices, and unusual size and frequency dependencies [6–8] were found to be described in terms of an analogy with Fano resonances of a quantum particle scattered by a potential with quasi-bound levels [6]. According to this analogy, localized plasmons (polaritons) excited by an incident radiation play the role of quasi-bound levels and their radiative decay is equivalent to the tunneling process from such levels [9]. The interference between the incident and re-emitted radiation gives rise to either enhancement (constructive interference) or suppression (destructive interference) of the electromagnetic field. Fano resonances were shown to be at the origin of the so-called “dark-states” in light scattering by coated spheres, where the local electromagnetic field enhancement occurs in off-resonant regions [10]. Light scattering by homogeneous particles can also exhibit unconventional Fano resonances in the extinction cross-section beyond the Rayleigh approximation; they result from the interference between different electromagnetic modes with the same multipole moment [11]. This contrasts to conventional Fano resonances, which arise from the spectral overlap of broad and narrow electromagnetic modes with different values of multipole moment [11].

The aim of this paper is to investigate conventional and unconventional Fano resonances and their connec-
tion to off-resonance field enhancement in coated spheres composed of a plasmonic core and a dielectric shell. We demonstrate that unconventional Fano resonances can also show up for coated spheres even in the Rayleigh limit, which is in contrast to the case of homogeneous spheres. These unconventional Fano resonances are at the origin of an off-resonance field enhancement within the scatterer, which can be even larger than the one achieved with conventional Fano resonances. In addition, we show that when the core has negative refractive index, conventional and unconventional Fano resonances can occur at the same input frequency, corresponding to the condition for an optimal field enhancement within the particle.

This paper is organized as follows. In Sec. II we describe light scattering by coated magnetic spheres within a generalization of the Lorenz-Mie scattering. In Sec. III we investigate the behavior of conventional and unconventional Fano resonances in coated spheres. The connection to off-resonance field enhancement is discussed in Sec. IV whereas Sec. V is devoted to the case of cores by a coated sphere with inner radius \(a\) and outer one \(b\). We assume that the coated sphere is made of linear, spatially homogeneous, and isotropic materials. The electric permittivities and magnetic permeabilities of the core \((0 \leq r \leq a)\) and the shell \((a \leq r \leq b)\) are \((\epsilon_1, \mu_1)\) and \((\epsilon_2, \mu_2)\), respectively. For simplicity, the scatterer is embedded in vacuum, with optical properties \((\epsilon_0, \mu_0)\) [see Fig. 1]. The extinction and scattering efficiencies (i.e., the respective cross-sections in units of \(\pi b^2\)) are: \(Q_{\text{ext}} = (2/y^2) \sum_{n=1}^{\infty} (2n + 1) \text{Re}(a_n + b_n)\) and \(Q_{\text{sca}} = (2/y^2) \sum_{n=1}^{\infty} [(a_n^2 + b_n^2)], \) where \(y = kb\) is the size parameter of the outer sphere \((k\) being the incident wave number \)]3]. By energy conservation, the absorption efficiency is \(Q_{\text{abs}} = Q_{\text{ext}} - Q_{\text{sca}}\). In particular, the radar backscattering efficiency is \(Q_{\text{backsc}} = (1/y^2) \sum_{n=1}^{\infty} (-1)^n (2n + 1)(a_n - b_n)^2\). In all the Q-factors, the Aden-Kerker scattering coefficients \(a_n\) and \(b_n\) are \[12\]:

\[
\begin{align*}
an &= \frac{(\bar{D}_n/m_2 + n/y) \psi_n(y) - \psi_{n-1}(y)}{(\bar{D}_n/m_2 + n/y) \xi_n(y) - \xi_{n-1}(y)}, \\
b_n &= \frac{(m_2 \bar{G}_n/m_2 + n/y) \psi_n(y) - \psi_{n-1}(y)}{(m_2 \bar{G}_n/m_2 + n/y) \xi_n(y) - \xi_{n-1}(y)},
\end{align*}
\]

where one defines the auxiliary functions

\[
\begin{align*}
\bar{D}_n &= \frac{D_n(m_2y) - A_n \chi'_n(m_2y)/\psi_n(m_2y)}{1 - A_n \chi'_n(m_2y)/\psi_n(m_2y)}, \\
\bar{G}_n &= \frac{D_n(m_2y) - B_n \chi'_n(m_2y)/\psi_n(m_2y)}{1 - B_n \chi'_n(m_2y)/\psi_n(m_2y)}, \\
\bar{A}_n &= \psi_n(m_2x) [\tilde{m}_2 D_n(m_1x) - \tilde{m}_1 D_n(m_2x)] \\
&= \frac{\tilde{m}_2 D_n(m_1x) \chi_n(m_2x) - \tilde{m}_1 \chi_n(m_2x)}{\tilde{m}_2 D_n(m_2x) - \tilde{m}_1 D_n(m_1x)}, \\
\bar{B}_n &= \psi_n(m_2x) [\tilde{m}_2 D_n(m_2x) - \tilde{m}_1 D_n(m_1x)] \\
&= \frac{\tilde{m}_2 D_n(m_2x) \chi_n(m_2x) - \tilde{m}_1 \chi_n(m_2x)}{\tilde{m}_2 D_n(m_2x) - \tilde{m}_1 D_n(m_1x)},
\end{align*}
\]

with \(x = ka\) the size parameter of the inner sphere and \(D_n(\rho) \equiv \frac{\text{d}[\ln \psi_n(\rho)]/\text{d} \rho}{\psi_n(\rho)}\). The functions \(\psi_n(\rho) = \rho J_n(\rho), \chi_n(\rho) = -\rho Y_n(\rho)\) and \(\xi_n(\rho) = \psi_n(\rho) - \chi_n(\rho)\) are the Riccati-Bessel, Riccati-Neumann and Riccati-Hankel functions, respectively, with \(j_n\) and \(y_n\) being the spherical Bessel and Neumann functions. The refractive and impedance indices are \(m = q_0/k = \sqrt{\epsilon_0 \mu_0/(\epsilon_0 \mu_0)}\) and \(\tilde{m}_q = \mu_0 m_q/\mu_3 = \sqrt{q_0/(\epsilon_0 \mu_0)}\), respectively \[14\] (with \(q = 1\) for the core and \(q = 2\) for the shell).

Figure 1. The scatterer geometry: a sphere of radius \(a\) and optical properties \((\epsilon_1, \mu_1)\) coated with a spherical shell of thickness \((b - a)\) and optical properties \((\epsilon_2, \mu_2)\); the embedding medium is \((\epsilon_0, \mu_0)\).

The angle-averaged electric \((\langle |E|^2 \rangle)\) and magnetic \((\langle |H|^2 \rangle)\) fields within the coated sphere can be explicitly calculated \[15\]. Here, we define \((\langle F \rangle) = \int_{-\pi/2}^{\pi/2} \text{d}(\cos \theta) \langle |F|^2 \rangle/4\pi\), where \(F = F(r, \cos \theta, \phi)\) is either the electric \((E)\) or the magnetic \((H)\) fields in the spherical coordinate system \((r, \theta, \phi)\). Using the exact expression for the electric field inside the core region \((0 \leq r \leq a)\), we obtain \[15\] \[16\]:

\[
\begin{align*}
\frac{\langle |E|^2 \rangle}{|E_0|^2} &= \frac{1}{2} \sum_{n=1}^{\infty} \left\{ (2n + 1)\epsilon_n^2 |j_n(\rho_1)|^2 \\
&+ |d_{n+1}|^2 n |j_{n+1}(\rho_1)|^2 + (n + 1) |j_{n-1}(\rho_1)|^2 \right\},
\end{align*}
\]

where \(\rho_1 = m_1 kr\) and \(E_0\) is the incident wave amplitude. The expression for the magnetic field is obtained by replacing \((E_1, E_0)\) with \((H_1, H_0)\) (where \(H_0 = E_0 \sqrt{\epsilon_0/\mu_0}\)).
and $(c_n, d_n)$ with $(\tilde{m}_1 d_n, \tilde{m}_1 c_n)$ in Eq. (4). For the shell region $(a \leq r \leq b)$ the angle-averaged electric field is [12]:

$$
\frac{\langle |E_r|^2 \rangle_{\Omega}}{|E_0|^2} = \frac{1}{2} \sum_{n=1}^{\infty} \left\{ (2n+1) \left[ |f_n|^2 |j_n(\rho_2)|^2 + |v_n|^2 |y_n(\rho_2)|^2 \right] + |g_n|^2 \left[ n |j_{n+1}(\rho_2)|^2 + (n+1) |j_{n-1}(\rho_2)|^2 \right] + |w_n|^2 \left[ n |y_{n+1}(\rho_2)|^2 + (n+1) |y_{n-1}(\rho_2)|^2 \right] + 2 \text{Re} \left[ (2n+1) f_n^* v_n j_n(\rho_2) y_n(\rho_2^*) + g_n w_n^* n y_{n+1}(\rho_2) y_{n-1}(\rho_2^*) \right] \right\},
$$

where $\rho_2 = n \omega k r$. The expression for the angle-averaged magnetic field within the shell is obtained by replacing $(E_r, E_0)$ with $(H_2, H_0)$, $(f_n, g_n)$ with $(\tilde{m}_2 g_n, \tilde{m}_2 f_n)$, and $(v_n, w_n)$ with $(\tilde{m}_2 v_n, \tilde{m}_2 w_n)$ in Eq. (5). In terms of the auxiliary coefficients $c_n, d_n, f_n, v_n, g_n$ and $w_n$ read [12]

$$
c_n = m_1 f_n \left[ \psi_m(m_2 y) - B_n \chi_n(m_2 y) \right] \left[ m_2 \psi_m(m_1 x) \right]^{-1},
$$

$$
d_n = m_1 g_n \left[ \psi_m'(m_2 y) - A_n \chi_n(m_2 y) \right] \left[ m_2 \psi_m(m_1 x) \right]^{-1},
$$

$$
f_n = \frac{m_2}{(\tilde{m}_2 G_n + n/y) \xi_n(y) - \xi_n^{-1}(y)},
$$

$$
g_n = \frac{m_2}{(\tilde{m}_2 A_n \chi_n(m_2 y) - \psi_n(m_2 y)) \xi_n(y) - \tilde{m}_2 \xi_n^{-1}(y)},
$$

$$
v_n = B_n f_n,
$$

$$
w_n = A_n g_n.
$$

The above expressions for the angle-averaged fields inside a coated sphere agree with Ref. [15] in the nonmagnetic case ($\mu_1 = \mu_2 = \mu_0$).

### III. FANO-LIKE RESONANCES IN COATED SPHERES

The identification of conventional Fano resonances require observables that are sensitive to wave interference. As a result, in light scattering by single homogeneous spheres, Fano-like resonances do not occur in the extinction $Q_{\text{ext}}$ and scattering $Q_{\text{sca}}$ efficiencies (and consequently in the absorption efficiency, $Q_{\text{abs}}$) since these quantities are proportional to the sum of intensities [4]. Also, since both $Q_{\text{ext}}$ and $Q_{\text{sca}}$ are averaged among all possible directions and polarizations, the resonance properties are averaged as well. For this reason, conventional Fano resonances in homogeneous spheres are expected to occur only in the cross-sections at a particular direction, e.g. the backscattering cross-section. However, for coated spheres the wave interferences, which ultimately lead to Fano-like resonances, can show up in the absorption efficiency $Q_{\text{abs}}$ even though $Q_{\text{ext}}$ and $Q_{\text{sca}}$ do not explicitly contain interference terms. To demonstrate this, note that one can explicitly write $Q_{\text{abs}}$ in terms of internal electromagnetic modes [13, 16, 17]. Defining the volume $V_q = 4\pi (l_q^2 - l_q^2)/3$, where $(l_1, l_2) = (0, a)$ for the core $(q = 1)$ and $(l_1, l_2) = (a, b)$ for the shell $(q = 2)$, one calculates the volume-averaged field intensities using $\langle |F|^2 \rangle_{V_q} = 3 \int_{l_q} d^2 \psi / r^2 \langle |F|^2 \rangle_{V_q}$. Since $\epsilon_q = \epsilon_q' + i \epsilon_q''$ and $\mu_q = \mu_q' + i \mu_q''$ and using the relations between $Q_{\text{abs}}$ and the absorbed powers [17], $P_{\text{abs}} = \omega \mu_q' \int_{V_q} d^3 r |E|^2 |/2$, we obtain:

$$
Q_{\text{abs}} = \frac{4}{3} y \left[ \left( \frac{e_q''}{e_0} \frac{\langle |E|^2 \rangle_{V_q}}{|E_0|^2} + \frac{\mu_q''}{\mu_0} \frac{\langle |H|^2 \rangle_{V_q}}{|H_0|^2} \right) S^3 + \left( \frac{e_q''}{e_0} \frac{\langle |E|^2 \rangle_{V_q}}{|E_0|^2} + \frac{\mu_q''}{\mu_0} \frac{\langle |H|^2 \rangle_{V_q}}{|H_0|^2} \right) (1 - S^3) \right],
$$

with $S = a/b$ being the thickness ratio. Inspecting the expression for the internal field [Eq. (8)], it is clear that Eq. (15) explicitly contains cross-terms such as $f_n v_n^*$ and $g_n w_n^*$ that physically represent interference between internal modes within the shell. This result suggests that Fano-like resonances could be detected in $Q_{\text{abs}}$ as well for coated spheres.

For single homogeneous spheres, unconventional Fano resonances result from the interference between different electromagnetic modes with the same multipole moment so that they show up in $Q_{\text{ext}}$ [11]. These Fano-like resonances are expected to exist either in homogeneous spheres with large dielectric permittivity, i.e. beyond the Rayleigh approximation, or in particles with spatial dispersion [11]. In the former case, such resonances may occur due the interference of e.g. two dipole modes, the resonant one (excited at a large dielectric permittivity) and the off-resonant Rayleigh one. The unconventional Fano-like resonances behavior in coated spheres, to the best of our knowledge, has been not reported so far.

To establish the conditions for the occurrence of Fano-like resonances in coated spheres, let us examine resonances in the multipolar moments $a_n$ and $b_n$, which are related to the surface modes or localized plasmon (polariton) resonances (LPRs). In Lorenz-Mie scattering, these LPRs are usually associated with a strong enhancement of the electromagnetic field intensity near the scatterer surface. Analytically, they are determined when denominators of the scattering coefficients $a_n$ and $b_n$ vanish [3]. For nonmagnetic ($\epsilon_1 = \mu_1 = \mu_0$) homogeneous spheres of radius $R$ in the Rayleigh limit ($kR \ll 1$ and $|m_1|kR \ll 1$), the electric surface modes are $\epsilon_1 = -(n+1)\epsilon_0/n$, with $n$ being the mode number. The lowest-order mode ($n = 1$) corresponds to the mode of uniform polarization throughout the sphere. The frequency in which it occurs, $\epsilon_1 (|\omega|) = -2\epsilon_0$, is often referred to as Fröhlich frequency [2]. For coated spheres in the Rayleigh limit ($y \ll 1$, $|m_1|x \ll 1$ and $|m_2|y \ll 1$), the application of the Fröhlich condition for the first mode ($n = 1$)
results in the following dipole scattering coefficient:

\[
\begin{equation}
\alpha_1 \approx \frac{2\gamma^3}{3} \left[ \frac{(\epsilon_0 - \epsilon_2)(\epsilon_1 + 2\epsilon_2) - (\epsilon_0 + 2\epsilon_2)(\epsilon_1 - \epsilon_2)}{(2\epsilon_0 + \epsilon_2)(\epsilon_1 + 2\epsilon_2) - 2(\epsilon_0 - \epsilon_2)(\epsilon_1 - \epsilon_2)} S^3 \right] \\
= -\frac{2\gamma^3}{3} \left[ \frac{\epsilon_2/\epsilon_0 - (1 - S^3)/(1 + 2S^3)}{\epsilon_2/\epsilon_0 + 2(1 - S^3)/(1 + 2S^3)} \right] \left( \frac{\epsilon_1 - \epsilon_1^{(\text{ant})}}{\epsilon_1 - \epsilon_1^{(\text{res})}} \right),
\end{equation}
\]

(16)

where we have kept up to \(O(\gamma^5)\). In Eq. (16), \(\epsilon_1^{(\text{ant})}\) and \(\epsilon_1^{(\text{res})}\) are the \(\epsilon_1\) values which make the numerator (an- tiresonance) or the denominator (resonance) of \(\alpha_1\) to vanish, respectively. Taking into account that only the core is dispersive \([\epsilon_1 = \epsilon_1(\omega)]\) in our geometry, the Fröhlich mode is determined for \(\epsilon_1(\omega_T) = \epsilon_1^{(\text{res})}\). Assuming that the media are weakly absorbing \((\epsilon_0' \ll \epsilon_0'')\), we obtain

\[
|\alpha_1(\omega)|^2 \propto \left[ \frac{\epsilon_1(\omega) - \epsilon_1^{(\text{res})}}{\epsilon_1(\omega) - \epsilon_1^{(\text{ant})}} \right]^2 \frac{[X(\epsilon_1) + \eta(\epsilon_1)]^2}{X(\epsilon_1)^2 + 1},
\]

where \(X(\epsilon_1) \equiv (\epsilon_1 - \omega_0)/\sqrt{\gamma^2 - 2\gamma(\epsilon_1 - \omega_0)}\) and \(\eta(\epsilon_1) \equiv \gamma/\sqrt{\gamma^2 - 2\gamma(\epsilon_1 - \omega_0)}\), with \(\omega_0 = \epsilon_1^{(\text{res})} + \epsilon_1^{(\text{ant})}/2\) and \(\gamma = \epsilon_1^{(\text{res})} - \epsilon_1^{(\text{ant})}/2\). A small mismatch between the resonance and anti-resonance parameters, \(\gamma/\epsilon_0 \ll 1\) (achieved for \(S \ll 1\)), implies that \(X(\epsilon_1) \approx (\epsilon_1 - \omega_0)/\gamma\) and \(\eta \approx \gamma/\gamma\), for \(\epsilon_1(\omega) \approx \omega_0\). In this situation, \(|\alpha_1|^2\), as a function of \(\epsilon_1(\omega)\), is a Fano-like resonance, characterized by the following (normalized) lineshape [3]:

\[
F_\eta(X) = \frac{1}{1 + \eta^2} \left( \frac{X + \eta^2}{X^2 + 1} \right),
\]

(17)

with \(\eta\) being the asymmetry parameter.

In Fig. 2(a), the scattering efficiency \(Q_{sca}\) for a coated sphere in the Rayleigh limit \((y \ll 1, |m_1|x \ll 1\) and \(|m_2|y \ll 1\) is plotted as a function of the electric permittivity of the core \((\epsilon_1, \mu_1 = \mu_0)\) in the vicinity of the dipole resonance. This plot confirms that \(|a_1|^2\) exhibits an unconventional Fano resonance, which is well described by the Fano lineshape, Eq. (17). Also, the plot in Fig. 2(a) demonstrates that, for coated spheres without spatial dispersion, the existence of unconventional Fano resonances does not necessarily require large values of the dielectric permittivities, a situation that is typically difficult to achieve in natural media. This result is in contrast to the case of homogeneous spheres, where such resonances are predicted to occur only beyond the Rayleigh limit [11]. Indeed, the plot in Fig. 2(a) shows that unconventional Fano resonances in light scattering by coated spheres can exist even in the Rayleigh limit \((y \ll 1, |m_1|x \ll 1\) and \(|m_2|y \ll 1\)\), corroborating the previous analytical result.

The effect of finite dissipation is investigated in Fig. 2(b), where \(Q_{sca}\) is shown as a function of the real part of the core electric permittivity \((\epsilon_1')\), for different values of its imaginary part \((\epsilon_1'')\), in the vicinities of the dipole resonances that result in the Fano lineshape in \(|a_1|^2\). From this plot, it is clear that these resonances are quite robust against losses so that they are not completely washed out in the presence of finite dissipation.

For homogeneous spheres, unconventional Fano resonances in \(|a_1|^2\) are expected to manifest themselves not only in the the scattering efficiency \(Q_{sca}\), but also in the extinction efficiency \(Q_{ext}\) even in the presence of finite dissipation [11]. For coated spheres in the Rayleigh limit with \(S \ll 1\), the plot in Fig. 2(a) reveals that extinction is dominated by absorption, i.e. typically \(Q_{sca} \ll Q_{abs}\), so that \(Q_{ext} \approx Q_{abs}\), even in the weakly absorption approximation. We have numerically verified that \(Q_{sca} \approx (kb)^4 Q_{abs}\) in off-resonant regions. A similar effect is well-known for sufficiently small homogeneous spheres of radius \(R\), where \(Q_{abs} \propto kR\) and \(Q_{sca} \propto (kR)^4\) [5]. As a result, unconventional Fano resonances in \(Q_{sca}\) tend to be unnoticed in \(Q_{ext}\), which, like \(Q_{abs}\), exhibits a Lorentzian profile in the vicinities of the dipole resonance in \(|a_1|^2\). Hence, the observation of unconventional Fano resonances in extinction for weakly absorbing coated spheres with \(S \ll 1\) requires that \(Q_{ext} \approx Q_{sca}\). This condition can be achieved imposing that \(ka \ll 1\) for the core and that \(kb\) is large enough to only amplify the dipole resonance, but small enough to ensure the validity of the first order approximation.

We emphasize that the unconventional Fano resonances in coated spheres are not a result of interferences between plasmon modes in the different layers of the system (core and shell), as it is generally expected for multilayered particles, such as nanoshells [4]. In contrast, here the Fano-like resonance in \(Q_{sca}\) in the Rayleigh limit [Fig. 2(a)] occurs due to self-interferences between partial waves within the shell. Mathematically, they arise from partial waves generated from both Bessel and Neumann special functions. Within the core, wave interferences are generated only from Bessel functions. The existence of these wave interference terms within the shell, that eventually lead to Fano-like resonances in \(Q_{sca}\), is confirmed by the analytical expressions for the angle-averaged field intensities inside the core and shell, Eqs. (4) and (6), respectively. Indeed, Eq. (6) explicitly encodes interferences between internal field coefficients, which are associated with the scattering ones by the boundary condition

\[
a_n = \frac{\psi_n(y)}{\xi_n(y)} \left[ \frac{\psi_n(m_2y) - A_n \chi_n(m_2y)}{\mu_2 \xi_n(y)} \right] g_n .
\]

(18)

The corresponding condition for \(b_n\) is obtained by replacing \((a_n, A_n)\) with \((b_n, B_n)\) and \((g_2, g_0)\) with \((m_2, f_0)\). Inspecting Eq. (13), one can see that \(|a_n|^2\) contains interference terms between the internal coefficients \(g_n\) and \(u_n = A_n g_n\) of the shell. The presence of these interference terms, such as e.g. \(g_1^* a_1 = A_1 |g_1|^2\), shows that an off-resonant \(A_1\) may play the role of a broad resonance that interferes with a sharp one in \(g_1\). Since the denominators of \(a_1\) and \(g_1\) are the same [see Eqs. (4) and (12)], an unconventional Fano resonance in \(|a_1|^2\) may occur for coated spheres even in the Rayleigh limit. Indeed, for a dielectric shell and dispersive core in the Rayleigh limit, one has \(\gamma/\epsilon_0 \ll 1\) if \(S \ll 1\); in this case, \(A_1\) plays the
role of the broad resonance in the Fano effect.

IV. CONNECTION TO OFF-RESONANCE FIELD ENHANCEMENT

For coated spheres consisting of metal-dielectric composites, it was demonstrated that conventional Fano resonances are at the origin of an off-resonant field enhancement. Due to the Fano effect, the maximal field enhancement inside the scatterer does not necessarily correspond to a resonance in the extinction efficiency, contrary to the common belief [10]. In the plot in Fig. 3(a), we investigate whether unconventional Fano resonances can also induce a similar effect. Since $Q_{\text{ext}} = Q_{\text{sca}} + Q_{\text{abs}}$, only the efficiencies $Q_{\text{sca}}$ and $Q_{\text{abs}}$ are calculated for a non-absorbing metallic sphere ($\epsilon_1 < 0; \mu_1 = \mu_0$) coated with a dielectric shell ($\epsilon_2 = 3.4 + 0.004i; \mu_2 = \mu_0$) as a

Figure 2. Scattering efficiency $Q_{\text{sca}}$ in the vicinities of the electric dipole resonance for a plasmonic sphere ($\epsilon_1, \mu_1 = \mu_0$) coated with a lossy dielectric shell ($\epsilon_2/\epsilon_0 = 3.4 + 0.001i; \mu_2/\mu_0 = 1$) as a function of $\epsilon_1$ in the the small-particle limit (size parameters $ka = 10^{-2}$, $kb = 10^{-3}$). (a) The dipole resonance for a lossless core ($\epsilon_1 = \epsilon_1^\prime$) is well described by the Fano lineshape, Eq. (17) (dotted line). The inset shows the absorption efficiency as a function of $\epsilon_1$ for a lossless core ($\epsilon_1 = \epsilon_1^\prime$) in the vicinities of the electric dipole resonances.

Figure 3. Fano-like resonances and off-resonance field enhancement for a metallic non-absorbing sphere ($\epsilon_1/\epsilon_0 < 0; \mu_1 = \mu_0$) coated with a lossy dielectric shell ($\epsilon_2/\epsilon_0 = 3.4 + 0.004i; \mu_2/\mu_0 = 1$) with size parameters $ka = 0.2$ and $kb = 1$. (a) $Q_{\text{sca}}$ and $Q_{\text{abs}}$ as a function of $\epsilon_1$. The dotted vertical line indicates the position of the unconventional Fano resonance. The inset shows the backscattering efficiency $Q_{\text{back}}$ as a function of $\epsilon_1$ at the vicinities of the quadrupole resonance. (b) Angle-averaged electric field intensity as a function of the distance to the center of the coated sphere for $\epsilon_1/\epsilon_0 = -7.91$ (sharp dipole resonance), $\epsilon_1/\epsilon_0 = -7.85$ (unconventional Fano resonance), $\epsilon_1/\epsilon_0 = -7.32$ (Fano dip), and $\epsilon_1/\epsilon_0 = -5.27$ (conventional Fano resonance).
function of $\epsilon_1$. The asymmetric lineshape of $Q_{sca}$ in the
vicinities of the electric dipole resonance is a result of the
unconventional Fano effect, as it results from the interference
of two dipole resonances (i.e. a resonance in $|a_1|^2$).
The unconventional Fano resonance frequency occurs for
$\epsilon_1/\epsilon_0 = -7.85$, a value in between the ones correspond-
ing to maximal and minimal scattering (constructive and
destructive interferences): $\epsilon_1/\epsilon_0 = -7.91$ (sharp dipole
resonance) and $\epsilon_1/\epsilon_0 = -7.32$ (broad dipole resonance),
respectively. For simplicity, we refer to the minimum
scattering in the Fano curve as the Fano dip. At the same
time, the value $\epsilon_1/\epsilon_0 = -7.85$ corresponds to a peak in
$Q_{abs}$ which, by Eq. (15), is related to maximal field en-
hancement inside the system. This result demonstrates
that unconventional Fano resonances can also lead to off-
resonance field enhancement. This conclusion is corro-
borated by the analysis of the plot in Fig. 3(b), which
shows the angle-averaged electric field as a function of
the distance to the center of the coated sphere for dif-
ferent values of $\epsilon_1$. The maximal field enhancement does
not correspond to maximal scattering, but rather it is
achieved for $\epsilon_1/\epsilon_0 = -7.85$, precisely the position of the
unconventional Fano resonance and the peak in $Q_{abs}$.

The plot in Fig. 3(a) also reveals the existence of a
conventional Fano resonance at $\epsilon_1/\epsilon_0 = -5.27$. It
appears in the backscattering efficiency $Q_{back}$ [see inset of
Fig. 3(a)] and results from the interference between a
broad dipole resonance with a sharp quadrupole one.
This conventional Fano resonance not only appears in
$Q_{back}$, as expected from Ref. [10], but also is indicated as
a peak in $Q_{sca}$, further confirming the analysis of Sec. III.
From Eq. (15) and since it does not appear as a peak in $Q_{sca}$,
this conventional Fano resonance is related to an off-resonance field enhancement inside the
scatterer. For these parameters, one can see from the
plot in Fig. 3(b) that the off-resonance enhancement of the
electric field related to conventional Fano resonances is
approximately one order of magnitude smaller than the
one achieved with the unconventional Fano effect.
One can partially explain this result by the fact that
the unconventional Fano effect involves two dipole res-
onances while the conventional one involves dipole and
quadrupole (which is of higher order) resonances. How-
evertheless, the quadrupole resonance would appear as a
peak in $Q_{sca}$. This indicates that, for a lossless dielec-
tric shell, a finite absorption in the dispersive core is nec-
esary to minimize the scattering at the quadrupole reso-
nance in small-particle limit. Also, for a metallic core and
dielectric shell, an off-resonance field enhancement near
the quadrupole resonance emerges only for $S \ll 1$ [10],
which is the same condition we have obtained for uncon-
ventional Fano resonances in coated spheres. This leads
to the possibility of combining these two interference ef-
fects.

Although Fano-like resonances in $Q_{back}$ exist even
for homogeneous spheres [4], “hidden” quadrupole res-
onances in $Q_{sca}$ (they appear only in $Q_{abs}$ and leads to
an off-resonance field enhancement) are usually achieved
in layered particles, as we show for coated spheres in the
following. In the Aden-Kerker solution, for a given $kb$
in off-resonant regions, $ka \ll 1$ leads to $|A_n| \ll 1$ and
$|B_n| \ll 1$, where $A_n$ and $B_n$ are defined in Eqs. (5) and
(6). Omitting the leading order $\mathcal{O}(x^5)$ in Rayleigh limit,
we obtain the approximations:

$$ A_1 \approx 2(m_2x)^5 \left( \frac{e_1 - e_2}{e_1 + 2e_2} \right) \left( \frac{e_1 - 2e_2}{e_1 + 2e_2} \right) $$

$$ A_2 \approx -\frac{(m_2x)^5}{15} \left( \frac{e_1 - e_2}{2e_1 + 3e_2} \right). $$

Resonances in $A_1$ and $A_2$ are obtained when the real
parts of their denominators vanish: $\epsilon_1(\omega) = -2\epsilon_2'$
and $\epsilon_1(\omega) = -3\epsilon_2'/2$, respectively. If we have no absorption ($\epsilon_1'' = \epsilon_2'' = 0$) for finite $x = ka$, both $A_1$ and $A_2$ diverge
at the resonance and there is no “hidden” quadrupole reso-
nance in $Q_{sca}$. Instead, a sharp resonance is observed
in $|a_2|^2$. Consider now a weakly absorbing media such that
the first term in Eq. (11) does not depend on $ka$
at the dipole resonance: $x^5 \approx (\epsilon_1'' + 2\epsilon_2'')$. This leads to
$x^5 \ll (2\epsilon_1'' + 3\epsilon_2'')$ and Eq. (20) is partially suppressed at
the quadrupole resonance, whereas the second term in-
side brackets in Eq. (19) yields $A_1 \sim 1/x$ in off-resonant
regions (broad dipole resonance). Therefore, in this
approximation, the dipole resonance is huge compared to
the quadrupole one in $Q_{sca}$ and one may achieve inter-
ference between $a_1$ and $a_2$ in $Q_{back}$. This result provides
a simple condition to obtain the so-called “dark states”
in the small-particle limit. Indeed, in the plot in Figs. 3,
we have assumed $x^3 = (\epsilon_1'' + 2\epsilon_2'') = 8 \times 10^{-5}$, and similar
conditions can always be fulfilled for dispersive cores.

V. OPTIMAL FIELD ENHANCEMENT WITH NEGATIVE REFRACTIVE INDEX MATERIALS

Many applications in photonics depends crucially on
their capacity of enhancing light confinement in small
volumes with low losses. In the previous section, we have
demonstrated that unconventional Fano resonances lead
to a large off-resonance enhancement of the electromag-
netic field intensity, even larger than the one achieved
for conventional Fano resonances. To achieve an opti-
mal field enhancement inside the coated sphere, let us
investigate the conditions for simultaneous occurrence
of conventional and unconventional Fano resonances at
the same frequency. With this aim, we consider a plas-
monic dispersive core $\{\epsilon_1(\omega), \mu_1(\omega)\}$ coated with a non-
dispersive dielectric shell $\{\epsilon_2, \mu_2 = \mu_0\}$ and impose that a
dipolar antiresonance in $a_1$ coincides with a quadrupolar
resonance in a conventional Fano resonance), and

\[ \frac{Q_{\text{scs}}}{Q_{\text{abs}}} = \frac{Q_{\text{scs}}}{Q_{\text{abs}}} \]

electric field intensity as a function of the radial distance for a dielectric shell \( \epsilon_2 < \epsilon_0 > 0 \) with thickness parameter \( 0 < S < 1 \), the second Fröhlich mode occurs in the interval \( -3\epsilon_2'/2 < \epsilon'_1(\omega_F) < -3\epsilon_0/2 \). Since \( \epsilon'_2\epsilon'_2 > 0 \) and \( \epsilon'_1 < 0 \), it follows from this conditions that \( \mu'_1 < 0 \). As a result, we conclude that for \( ka < 1 \) and a dielectric shell \( m'_2 > 0 \) a sharp plasmon resonance in \( a_2 \) will only coincide with an antiresonance in \( a_1 \) if the dispersive core has a negative refractive index \( m'_1 < 0 \).

Figure 4(a) shows the plots of the scattering \( Q_{\text{scs}} \) and absorption \( Q_{\text{abs}} \) efficiencies for non-absorbing sphere with negative refraction index \( \epsilon_1/\epsilon_0 < 0; \mu_1/\mu_0 = -21 \) coated with a lossy dielectric shell \( \epsilon_2 = 3.4 + 0.004i; \mu_2 = \mu_0 \) as a function of \( \epsilon_1 \). We demonstrate that the dipole and quadrupole resonance can be brought together provided the core has a negative index, confirming the previous analytical result. Hence conventional and unconventional Fano resonances can occur at the same value of \( \epsilon_1 \). As a consequence, there is a huge field enhancement near these resonances, as confirmed by the analysis of the plot in Fig. 4(b), and one may achieve the maximum field stored inside the particle with minimum scattering (Fano dip in \( \epsilon_1/\epsilon_0 = -4.27 \)).

VI. CONCLUSIONS

In this paper we have investigated Fano-like resonances in light scattering by coated spheres composed of a dispersive plasmonic core and a dielectric shell. Using the Aden-Kerker solution, we have derived an analytical expression for the absorption efficiency \( Q_{\text{abs}} \) as a function of the internal fields. This expression explicitly contains interference terms between internal Aden-Kerker coefficients of the shell that are not washed out by the average among all possible directions and polarizations. This result shows that Fano-like resonances, which result from interference between electromagnetic modes inside the scatterer, can be identified in the total cross-sections, which contrasts to the common belief that field interferences, and hence Fano resonances, can only be identified in differential scattering spectra (e.g. the radar backscattering efficiency). Specially, we have demonstrated that unconventional Fano resonances, recently discovered for homogeneous spheres [11], can also occur for coated spheres. These resonances arise from the interference between two electromagnetic modes with the same multipole moment within the shell and not from interactions between various plasmon modes of different
layers of the particle. In contrast to the case of homogeneous spheres, the existence of unconventional Fano resonances for coated spheres do not require large electric permittivities so that they can occur even in the Rayleigh limit. As for conventional Fano resonances, unconventional Fano resonances in coated spheres can induce an off-resonance field enhancement. This enhancement is, nevertheless, approximately one order of magnitude larger than the one achieved with conventional Fano resonances for lossy scatterers, since unconventional Fano resonances involves multipole moments with the same order. Finally, we have examined the conditions for an optimal field enhancement inside the scatterer. We show that unconventional and conventional Fano resonances can occur simultaneously provided the core has a negative refraction index, leading to a maximal field enhancement. We believe that our results could be relevant not only for a better understanding of Fano resonances in light scattering but also to the development of novel applications in photonics.

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[1] U. Fano, Phys. Rev. 124, 1866 (1961).
[2] A. E. Miroshnichenko, S. Flach, and Y. S. Kivshar, Rev. Mod. Phys 82, 2257 (2010).
[3] R. Wood, Proc. R. Soc. London Ser. A 18, 269 (1902).
[4] B. Luk’yanchuk, N. I. Zheludev, S. A. Maier, N. J. Halas, P. Nordlander, H. Giessen, and C. T. Chong, Nature Mater. 9, 707 (2010).
[5] C. F. Bohren and D. R. Huffman, Absorption and Scattering of Light by Small Particles (Wiley, New York, 1983).
[6] M. I. Tribelsky and B. S. Luk’yanchuk, Phys. Rev. Lett. 97, 263902 (2006).
[7] M. Bashevoy, V. Fedotov, and N. Zheludev, Opt. Express 13, 8372 (2005).
[8] Z. B. Wang, B. S. Luk’yanchuk, M. H. Hong, Y. Lin, and T. C. Chong, Phys. Rev. B 70, 035418 (2004).
[9] M. I. Tribelsky, S. Flach, A. E. Miroshnichenko, A. V. Gorbach, and Yu S. Kivshar, Phys. Rev. Lett. 100, 043903 (2008).
[10] A. E. Miroshnichenko, Phys. Rev. A 81, 053818 (2010).
[11] M. I. Tribelsky, A. E. Miroshnichenko, and Y. S. Kivshar, Europhys. Lett. 97, 44005 (2012).
[12] A. L. Aden and M. Kerker, J. Appl. Phys. 22, 1242 (1951).
[13] T. J. Arruda, F. A. Pinheiro, and A. S. Martinez, J. Opt. 14, 065101 (2012).
[14] F. A. Pinheiro, A. S. Martinez, and L. C. Sampaio, Phys. Rev. Lett. 84, 1435 (2000); 85, 5563 (2000).
[15] T. Kaiser, S. Lange, and G. Schweiger, Appl. Opt. 33, 7789 (1994).
[16] T. J. Arruda and A. S. Martinez, J. Opt. Soc. Am. A 27, 992 (2010); 27, 1679 (2010).
[17] R. Ruppin, J. Opt. 13, 095101 (2011).