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Subwavelength resonant dielectric nanoparticles with high refractive indices

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Abstract

This paper aims at understanding the nature of the subwavelength resonant frequencies of dielectric particles with high refractive indices. It is proved that for an arbitrary shaped particle, these subwavelength resonant frequencies can be expressed in terms of the eigenvalues of the Newtonian potential associated with its shape. The enhancement of the scattered field at the resonant frequencies is shown. The hybridization of the subwavelength resonant frequencies of a dimer consisting of high refractive index dielectric nanoparticles is also characterized.

Mathematics Subject Classification (MSC2000). 35R30, 35C20.

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1 Introduction

Nanoscale optics is usually associated with plasmonic resonant structures made of metals such as gold or silver. Plasmonic resonances of nanoparticles can be treated as an eigenvalue problem for the Neumann-Poincaré operator, see [1, 6, 7, 9]. However, plasmonic structures suffer from high losses inherent in metals and dissipation due to heating. Recent developments in nanoscale optical physics have led to a new branch of nanophotonics focused on the manipulation of optically induced subwavelength resonances in dielectric nanoparticles with high refractive indices [14, 16, 17]. Resonant high-index dielectric nanostructures form new building blocks which can be used to realize unique functionalities and novel photonic devices [14]. Their study has been established as a new research direction in nanophotonics. Nevertheless, despite strong experimental efforts, mathematical modeling of resonant high-index nanoparticles remains limited. Apart from the case where the particles are disks or spheres, their subwavelength resonant frequencies have not been characterized yet.

In this paper, we consider a dielectric high-index nanoparticle of arbitrary shape and characterize its subwavelength resonances in terms of the eigenvalues of the Newtonian potential

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associated with its shape. Our formula is closely related to the one established in [15]. Then, we provide an asymptotic formula for the field scattered by a dielectric nanoparticle and estimate the scattering enhancement near its resonant frequencies. We also consider the hybridization phenomenon of a dimer consisting of high refractive index dielectric nanoparticles. We derive asymptotic formulas for the hybridized resonant frequencies, which correspond to monopole and dipole modes.

For simplicity of presentation, we consider the Helmholtz equation as a model for the wave propagation. But one should emphasize that the approach developed here can be extended to the full Maxwell's equations. On the other hand, based on the asymptotic formula for the scattered field derived in this paper, one can characterize the temporal response of resonant dielectric nanoparticles and obtain a time-domain resonant-mode-expansion for the scattered field, which generalizes the time-domain asymptotic formula proved in [4] to the case of a resonant subwavelength particle. This can be easily done by reproducing the calculations presented in [11, Appendix B].

Our results in this paper provide a solid mathematical framework for the analysis of resonant dielectric nanoparticles. They make possible the direct calculation of resonant frequencies and the optimal design of dielectric nanoparticles that resonate at specified frequencies. They can also be applied in the design of dielectric metamaterials and are expected to adavance the applications described in [12,16,17], in particular those concerned with metasurfaces, double-negative all dielectric materials, super-focusing, and wavefront control at the deep subwavelength scale.

2 Resonant frequencies of dielectric nanoparticles with high refractive indices

Let $D \Subset \mathbb{R}^d$, for $d = 2, 3$, be a small particle of the form $D = z + \delta B$, where δ is its characteristic size, z its location, and B is a smooth bounded domain containing the origin. Let ω denote the frequency, let $\varepsilon \equiv \tau\varepsilon_c + \varepsilon_m$ inside D and $\varepsilon \equiv \varepsilon_m$ outside D. Here, $\varepsilon_c, \varepsilon_m$, and τ are positive constants. Let E^{in} be an incident plane wave with frequency ω.

Consider the Helmholtz equation

$$\begin{cases} (\Delta + \omega^2 \varepsilon) E = 0 \quad \text{in } \mathbb{R}^d, \\ E - E^{\text{in}} \text{ satisfies the Sommerfeld radiation condition.} \end{cases}$$

From

$$(\Delta + \omega^2 \varepsilon_m)(E - E^{\text{in}}) = -\omega^2 \tau \varepsilon_c E \mathbb{1}_D \quad \text{in } \mathbb{R}^d,$$

where $\mathbb{1}_D$ is the characteristic function of D, it follows that the following Lippmann-Schwinger representation formula holds:

$$(E - E^{\text{in}})(x) = -\omega^2 \tau \varepsilon_c \int_D E(y) \Gamma_m(x - y; \omega) dy \quad \text{for } x \in \mathbb{R}^d, \tag{1}$$

where Γ_m is the outgoing (i.e., subject to the Sommerfeld radiation condition) fundamental solution of $\Delta + \varepsilon_m \omega^2$ in free space.

Let $k_m = \omega\sqrt{\varepsilon_m}$. Let the volume integral operator $K_D^{k_m}$ be defined by

$$K_D^{k_m} : E \in L^2(D) \mapsto - \int_D E(y) \Gamma_m(x - y; \omega) dy \in L^2(D).$$

It is well known that, due to the weak singularity of the fundamental solution, $K_D^{k_m}$ is compact. When the norm of $\tau\omega^2\varepsilon_c K_D^{k_m}$ is smaller than 1, $I - \tau\omega^2\varepsilon_c K_D^{k_m}$ is invertible, so (3) can be rewritten as

$$E(x) = (I - \tau\omega^2 \varepsilon_c K_D^{k_m})^{-1}[E^{\text{in}}](x) \quad \text{for all } x \in D, \tag{2}$$

where I denotes the identity operator.

Assume that the characteristic size δ of the particle D is much smaller than the wavelength $2\pi/k_m$), and let $\omega \to 0$. The subwavelength resonance problem is then to find an $\omega \in \mathbb{C}$ close to 0 such that $(I - \tau\omega^2\varepsilon_c K_D^{k_m})^{-1}$ is singular, or equivalently, such that there exists $L^2(D) \ni E \not\equiv 0$ with

$$E(x) + \omega^2\tau\varepsilon_c \int_D E(y)\Gamma_m(x-y;\omega)dy = 0, \quad \text{for } x \in D; \tag{3}$$

see [10]. Such an ω would be a subwavelength resonance for the high refractive index dielectric particle D.

Through a Taylor series expansion of the fundamental solution we obtain the following result.

Lemma 2.1. *Let $d = 3$. Let $K_D^{(0)}$ be the Newtonian potential on D, i.e., the operator defined by*

$$K_D^{(0)}[E](x) = -\int_D E(y)\Gamma(x-y)\,dy \quad \textit{for } x \in D,$$

with $\Gamma(x)$ being the fundamental solution of the Laplacian in $\mathbb{R}^3$. The operator $K_D^{k_m}$ can be rewritten as

$$K_D^{k_m} = \sum_{i=0}^{\infty} \omega^i K_D^{(i)}, \tag{4}$$

where the series converges in operator norm if ω is small enough.

Let $A_i = \tau\omega^2\varepsilon_c K_D^{(i)}$. By expanding with a Neumann series, we have

$$\begin{aligned}\Big(I - A_0 - \sum_{i=1}^{\infty}\omega^i A_i\Big)^{-1} &= \Big(I - (I-A_0)^{-1}\sum_{i=1}^{\infty}\omega^i A_i\Big)^{-1}(I-A_0)^{-1} \\ &= \sum_{k=0}^{\infty}\Big((I-A_0)^{-1}\sum_{i=1}^{\infty}\omega^i A_i\Big)^k (I-A_0)^{-1} \\ &= (I-A_0)^{-1} + (I-A_0)^{-1}\omega A_1 (I-A_0)^{-1} + O(\omega^3). \end{aligned} \tag{5}$$

Recall that $K_D^{(0)} : L^2(D) \to L^2(D)$ is a compact, self-adjoint operator. Let, for the sake of clarity of the presentation, λ_0 be a simple eigenvalue of $K_D^{(0)}$ associated with the normalized eigenfunction ϕ_0 in $L^2(D)$. We remark that the eigenvalues of $K_D^{(0)}$ are positive. For the analysis of the spectrum of the Newtonian potential, we refer the reader, for instance, to [13].

Let ω_0 be a frequency at which $I - A_0$ becomes singular. In particular, let

$$\omega_0 = 1/\sqrt{\tau\varepsilon_c\lambda_0}. \tag{6}$$

Note that ω_0 is small only for τ large enough. This shows that subwavelength resonances occur only for particles with high refractive indices.

For ω near ω_0, we have, by a pole-pencil operator decomposition, that

$$(I - A_0)^{-1}[\psi] = \frac{\langle \psi, \phi_0\rangle \phi_0}{1 - \tau\omega^2\varepsilon_c\lambda_0} + R(\omega)[\psi],$$

where $\omega \mapsto R(\omega)$ is analytic in a neighborhood of ω_0 and $\langle \cdot, \cdot \rangle$ denotes the scalar product on $L^2(D)$. Hence, considering only the first two terms in the expansion (5), we obtain from (2) that an approximation of the resonance must satisfy (see, for instance, [2,5])

$$\frac{\phi_0}{1 - \tau\omega^2\varepsilon_c\lambda_0} + \tau\omega^3\varepsilon_c \frac{\langle K_D^{(1)}[\phi_0], \phi_0\rangle}{(1 - \tau\omega^2\varepsilon_c\lambda_0)^2}\phi_0 = 0.$$

Therefore, we have the following approximation for the subwavelength resonances.

Proposition 2.2. *Let $d = 3$ and let τ be large enough. Let ω_0 be defined by (6), where λ_0 is an eigenvalue of the Newtonian potential $K_D^{(0)}$. Then, the $O(\omega^4)$-approximation of the subwavelength resonant frequencies ω_{s} of the dielectric particle D satisfies*

$$1 - \tau\omega_{\mathrm{s}}^2\varepsilon_c\lambda_0 = -\tau\omega_{\mathrm{s}}^s\varepsilon_c\langle K_D^{(1)}[\phi_0], \phi_0\rangle.$$

Note that, in three dimensions,

$$K_D^{(1)}[\phi] = -i\frac{\sqrt{\varepsilon_m}}{4\pi}\int_D \phi\, dy \quad \text{for all } \phi \in L^2(D).$$

Therefore, ω_{s} satisfies

$$1 - \tau\omega_{\mathrm{s}}^2\varepsilon_c\lambda_0 = \frac{i\tau}{4\pi}\omega_{\mathrm{s}}^s\sqrt{\varepsilon_m}\varepsilon_c\Big(\int_D \phi_0\, dy\Big)^2.$$

Since ω_{s} is close to ω_0, by approximating $\omega_{\mathrm{s}}^3 \simeq \omega_0^3$, and since by definition $\tau\varepsilon_c\lambda_0 = 1/\omega_0^2$, we obtain

$$1 - \frac{\omega_{\mathrm{s}}^2}{\omega_0^2} = \frac{i\tau}{4\pi}\omega_0^3\sqrt{\varepsilon_m}\varepsilon_c\Big(\int_D \phi_0\, dy\Big)^2.$$

Corollary 2.3. *Let $d = 3$. Then, the $O(\omega^4)$-approximation of the subwavelength resonant frequencies of the dielectric particle D can be computed as*

$$\omega_{\mathrm{s}} = \omega_0 - \frac{i}{8\pi}\frac{\omega_0^2}{\lambda_0}\sqrt{\varepsilon_m}\Big(\int_D \phi_0\, dy\Big)^2.$$

By using the Lippmann-Schwinger representation formula (3), we can also rewrite

$$E(x) - E^{\mathrm{in}}(x) \simeq -\omega^2\tau\varepsilon_c\Gamma_m(x-z;\omega)\frac{\langle E^{\mathrm{in}}, \phi_0\rangle(\int_D \phi_0)}{1-\tau\omega^2\varepsilon_c\lambda_0} + \tau\omega^3\varepsilon_c\frac{\langle K_D^{(1)}[\phi_0], \phi_0\rangle\langle E^{\mathrm{in}}, \phi_0\rangle(\int_D \phi_0)}{(1-\tau\omega^2\varepsilon_c\lambda_0)^2}.$$

By plugging the expression of ω_{s} obtained in Proposition 2.2 into the above approximation of the scattered field, we arrive at the following result.

Proposition 2.4. *For ω (real) near the resonant frequency ω_{s} and E^{in} such that $\langle E^{\mathrm{in}}, \phi_0\rangle_{L^2(D)} \neq 0$, the following monopole approximation of the dielectric nanoparticle D holds:*

$$E(x) - E^{\mathrm{in}}(x) \simeq -\frac{\lambda_0\Big(\frac{\omega_{\mathrm{s}}^2}{\omega^2} - 1\Big) - i\frac{\sqrt{\varepsilon_m}}{4\pi}(\int_D \phi_0)^2\Big(\omega - \frac{\omega_{\mathrm{s}}^3}{\omega^2}\Big)}{\Big(\lambda_0\Big(\frac{\omega_{\mathrm{s}}^2}{\omega^2} - 1\Big) - i\frac{\sqrt{\varepsilon_m}}{4\pi}(\int_D \phi_0)^2\,\frac{\omega_{\mathrm{s}}^3}{\omega^2}\Big)^2}\langle E^{\mathrm{in}}, \phi_0\rangle_{L^2(D)}\Gamma_m(x-z;\omega), \qquad (7)$$

for $|x-z| \gg 2\pi/(\omega\sqrt{\varepsilon_m})$.

Now, we turn to the two-dimensional case. In this case the problem is complicated by the logarithmic singularity of the operator $K_D^{k_m}$ as $\omega \to 0$ which gives rise to an averaging operator at leading order when asymptotically expanded. This means we cannot expect to frame the resonance frequency in terms of a single eigenvalue of the Newtonian potential. Instead, in two dimensions the resonance frequency takes account of an infinite number of eigenvalues of the Newtonian potential.

From the asymptotic expansion of the Hankel function $H_0^{(1)}$ of the first kind of order zero:

$$H_0^{(1)}(s) = \frac{2i}{\pi}\sum_{m=0}^{\infty}(-1)^m\frac{s^{2m}}{2^{2m}(m!)^2}\Big(\log(\hat{\gamma}s) - \sum_{j=1}^{m}\frac{1}{j}\Big),$$

where $2\hat{\gamma} = \exp(\gamma - i\pi/2)$ with γ being the Euler's constant (see, for instance, [8]), it follows that

$$K_D^{k_m}[E] = -\frac{1}{2\pi}(\log(\omega\sqrt{\varepsilon_m}\hat{\gamma}))\int_D E(y)\,dy + K_D^{(0)} + (\omega^2\log\omega) \quad \text{as } \omega \to 0, \tag{8}$$

where $K_D^{(0)}$ is the Newtonian potential in dimension two, that is, the operator defined on $L^2(D)$ by

$$K_D^{(0)}[E](x) = \int_D E(y)\Gamma(x-y)\,dy \quad \text{for } x \in D,$$

with Γ being the fundamental solution of the Laplacian in $\mathbb{R}^2$.

Expanding $K_D^{k_m}$ as in (5) and following the same calculations, we obtain the following characterization of subwavelength resonant frequencies in the two-dimensional case; see Appendix A.

Proposition 2.5. *Let $d = 2$ and τ large enough. Then, the $o(\omega^2)$-approximation of the subwavelength resonant frequencies ω_{s} of the dielectric particle D satisfies*

$$1 - \omega_{\mathrm{s}}^2\tau\varepsilon_c\Big(-\frac{|D|}{2\pi}\log(\omega_{\mathrm{s}}\hat{\gamma}\sqrt{\varepsilon_m}) + \langle K_D^{(0)}[\hat{\mathbb{1}}_D], \hat{\mathbb{1}}_D\rangle\Big) = 0,$$

where $|D|$ is the volume of D and $\hat{\mathbb{1}}_D = \mathbb{1}_D/\sqrt{|D|}$.

3 Hybridization of subwavelength resonant frequencies for a dimer of dielectric nanoparticles

Consider a dimer of two identical particles D_1 and D_2 with the same dielectric parameter as in the above section. Then the field $E - E^{\mathrm{in}}$ scattered by the two particles has the following representation formula:

$$(E - E^{\mathrm{in}})(x) = -\omega^2\tau\varepsilon_c\Big(\int_{D_1} E(y)\Gamma_m(x-y;\omega)dy + \int_{D_2} E(y)\Gamma_m(x-y;\omega)dy\Big) \quad \text{for } x \in \mathbb{R}^d. \tag{9}$$

Define the operators $K_{D_i}^{k_m}$ and $R_{D_i,D_j}^{k_m}$ for $i, j = 1, 2$, by

$$K_{D_i}^{k_m} : E|_{D_i} \in L^2(D_i) \mapsto -\int_{D_i} E(y)\Gamma_m(x-y;\omega)dy\big|_{D_i} \in L^2(D_i),$$

and

$$R_{D_i,D_j}^{k_m} : E|_{D_i} \in L^2(D_i) \mapsto -\int_{D_i} E(y)\Gamma_m(x-y;\omega)dy\big|_{D_j} \in L^2(D_j).$$

Then, from (9) we obtain the following system of operator equations:

$$\begin{pmatrix} 1 - \tau\omega^2\varepsilon_c K_{D_1}^{k_m} & -\tau\omega^2\varepsilon_c R_{D_2,D_1}^{k_m} \\ -\tau\omega^2\varepsilon_c R_{D_1,D_2}^{k_m} & 1 - \tau\omega^2\varepsilon_c K_{D_2}^{k_m} \end{pmatrix}\begin{pmatrix} E|_{D_1} \\ E|_{D_2} \end{pmatrix} = \begin{pmatrix} E^{\mathrm{in}}|_{D_1} \\ E^{\mathrm{in}}|_{D_2}. \end{pmatrix} \tag{10}$$

The scattering resonance problem is to find ω such that the operator in (10) is singular, or equivalently such that there exists $L^2(D_1) \times L^2(D_2) \ni (E_1, E_2) \not\equiv (0,0)$ such that

$$\begin{pmatrix} 1 - \tau\omega^2\varepsilon_c K_{D_1}^{k_m} & -\tau\omega^2\varepsilon_c R_{D_2,D_1}^{k_m} \\ -\tau\omega^2\varepsilon_c R_{D_1,D_2}^{k_m} & 1 - \tau\omega^2\varepsilon_c K_{D_2}^{k_m} \end{pmatrix}\begin{pmatrix} E|_{D_1} \\ E|_{D_2} \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix}. \tag{11}$$

Note that here we have a coupled system of subwavelength resonators. As in [2, 3], the following results hold.

Proposition 3.1. *Let $d = 3$. The subwavelength resonant frequency ω_{s} is hybridized into two subwavelength resonant frequencies $\omega_{\mathrm{s}}^{\pm}$ approximately given by*

$$\omega_{\mathrm{s}}^{\pm} = \omega_0 \pm \frac{1}{2}\tau\omega_0^3\varepsilon_c\sqrt{(R_{D_1,D_2}^{\omega_{\mathrm{s}}}[\phi_0^{(1)}], \phi_0^{(2)})(R_{D_2,D_1}^{\omega_{\mathrm{s}}}[\phi_0^{(2)}], \phi_0^{(1)})}, \qquad (12)$$

where $\phi_0^{(i)}$, for $i = 1, 2$, is the eigenfunction associated to the eigenvalue λ_0 of the Newtonian potential of D_i. Moreover, in the far-field, the dimer of dielectric particles behaves as the sum of a monopole and a dipole.

Now, let $d = 2$ and consider for simplicity a dimer of two identical disks D_1 and D_2 with the same dielectric parameters as in the above section.

Define the operators $K_{D_i}^{k_m}$ and $R_{D_i,D_j}^{k_m}$ for $i, j = 1, 2$, by

$$\begin{aligned} K_{D_i}^{k_m} &: E|_{D_i} \in L^2(D_i) \mapsto -\int_{D_i} E(y)\Gamma_{k_m}(x-y)dy\big|_{D_i} \in L^2(D_i), \\ R_{D_i,D_j}^{k_m} &: E|_{D_i} \in L^2(D_i) \mapsto -\int_{D_i} E(y)\Gamma_{k_m}(x-y)dy\big|_{D_j} \in L^2(D_j). \end{aligned}$$

Define the operators $M_{D_i}^{k_m}$ and $N_{D_i,D_j}^{k_m}$ for $i, j = 1, 2$, by

$$\begin{aligned} M_{D_i}^{k_m} &:= \hat{K}_{D_i}^{k_m} + K_{D_i}^{(0)}, \\ N_{D_i,D_j}^{k_m} &:= \hat{K}_{D_i,D_j}^{k_m} + R_{D_i,D_j}^{(0)}, \end{aligned}$$

where

$$\begin{aligned} K_{D_i}^{(0)} &: E|_{D_i} \in L^2(D_i) \mapsto \int_{D_i} E(y)\Gamma(x-y)dy|_{D_i} \in L^2(D_i), \\ \hat{K}_{D_i}^{k_m} &: E|_{D_i} \in L^2(D_i) \mapsto \log(\hat{\gamma}k_m)\hat{K}_{D_i}[E]|_{D_i} \in L^2(D_i), \\ \hat{K}_{D_i} &: E|_{D_i} \in L^2(D_i) \mapsto -\frac{1}{2\pi}\int_{D_i} E(y)dy|_{D_i} \in L^2(D_i), \\ R_{D_i,D_j}^{(0)} &: E|_{D_i} \in L^2(D_i) \mapsto \int_{D_i} E(y)\Gamma(x-y)\big|_{D_j} \in L^2(D_j), \\ \hat{K}_{D_i,D_j}^{k_m} &: E|_{D_i} \in L^2(D_i) \mapsto \log(\hat{\gamma}k_m)\hat{K}_{D_i}[E]|_{D_j} \in L^2(D_j), \\ \hat{K}_{D_i,D_j} &: E|_{D_i} \in L^2(D_i) \mapsto -\frac{1}{2\pi}\int_{D_i} E(y)dy|_{D_j} \in L^2(D_j). \end{aligned}$$

We refer to Appendix C for the proof of the following proposition.

Proposition 3.2. *Let $d = 2$ and τ large enough. Then the monopole and dipole hybridized resonances of the dimer of two identical disks D_1 and D_2 of radius δ are approximately given by*

$$1 - \omega^2\tau\varepsilon_c\Big(-\frac{\delta^2}{2}\log(\omega\hat{\gamma}\sqrt{\varepsilon_m})(1 \pm 1) + \langle K_{D_1}^{(0)}[\hat{\mathbb{1}}_{D_1}]\hat{\mathbb{1}}_{D_1}\rangle \pm \langle R_{D_2,D_1}^{(0)}[\hat{\mathbb{1}}_{D_2}], \hat{\mathbb{1}}_{D_1}\rangle \Big) = 0.$$

The following corollary gives more explicit formulae for the hybridized resonances in the case when D_1 and D_2 are unit disks.

Corollary 3.3. *Let $d = 2$ and τ large enough. Then the monopole and dipole hybridized resonances of a dimer of two identical unit disks D_1 and D_2 are given by*

$$\omega_{\mathrm{m}}(\tau) = -\frac{\sqrt{2}i}{\sqrt{\tau\varepsilon_c W(\Phi(\tau))}},$$
$$\omega_{\mathrm{d}}(\tau) = \frac{\sqrt{2}}{\sqrt{\tau\varepsilon_c\left(\frac{1}{4} - \frac{2}{\pi}\langle R^{(0)}_{D_2,D_1}[\mathbb{1}_{D_2}], \mathbb{1}_{D_1}\rangle\right)}},$$

where

$$\Phi(\tau) = -\frac{2}{\tau\varepsilon_c}\exp\left(2\log(\sqrt{\varepsilon_m}\hat{\gamma}) - \frac{1}{4} - \frac{2}{\pi}\langle R^{(0)}_{D_2,D_1}[\mathbb{1}_{D_2}], \mathbb{1}_{D_1}\rangle\right),$$

and W is the lower branch of the Lambert W function defined in the interval $[-1/e, 0)$.

Remark 3.4. Note that as $\tau \to \infty$ the monopole resonance $\omega_{\mathrm{m}} = O(1/\sqrt{\tau\log(\tau)})$ and hence it decays at the same rate as the single particle resonance ω_{s}, however the dipole resonance ω_{d} decays slightly slower, i.e., as $O(1/\sqrt{\tau})$. ■

4 Numerical illustrations

Let $\varepsilon_m = \varepsilon_c = 1$. Let D, D_1, and D_2 be unit disks with D centered at the origin, D_1 centered at $(-2, 0)$, and D_2 centered at $(2, 0)$ with D being the geometry for the single particle problem (3) and $D_1 \cup D_2$ being the geometry for the dimer problem (11).

The asymptotic resonances ω_{s}, ω_{m}, and ω_{d} are given by the formulas in Proposition B.1 and Corollary 3.3, with the $\langle R^{(0)}_{D_2,D_1}[\mathbb{1}_{D_2}], \mathbb{1}_{D_1}\rangle$ term in the hybridized resonances computed numerically using Python's `nquad` routine after first putting it in polar coordinates with respect to the center of D_1. We numerically compute reference solutions to the single particle problem and the dimer problem using boundary integral equation formulations expanded on multipole bases to obtain reference resonances $\omega_{\mathrm{s,ref}}$ (single particle), $\omega_{\mathrm{m,ref}}$ (monopole), and $\omega_{\mathrm{d,ref}}$ (dipole). In Figure 1 we plot the asymptotic resonances along with the corresponding reference resonances and predicted rates of convergence.

In Table 1 we give values of ω_{s} and $\omega_{\mathrm{s,ref}}$ and their corresponding relative errors for $\tau \in \{2^j\}_{j=3}^7$.

τ	$\mathrm{Re}(\omega_{\mathrm{s,ref}})$	$\mathrm{Re}(\omega_{\mathrm{s}})$	$\mathrm{Im}(\omega_{\mathrm{s,ref}})$	$\mathrm{Im}(\omega_{\mathrm{s}})$	Rel. err.
2^3	2.8012e−01	2.8043e−01	1.4476e−01	1.4022e−01	1.44e−02
2^4	1.9685e−01	1.9649e−01	8.4484e−01	8.3198e−01	6.23e−03
2^5	1.3602e−01	1.3581e−01	4.9465e−01	4.9189e−01	2.40e−03
2^6	9.3188e−02	9.3111e−02	2.9242e−02	2.9221e−02	8.22e−04
2^7	6.3606e−02	6.3587e−02	1.7497e−02	1.7521e−02	4.68e−04

Table 1: The real and imaginary parts of ω_{s} and $\omega_{\mathrm{s,ref}}$ along with the relative error for $\tau \in \{2^j\}_{j=3}^7$.

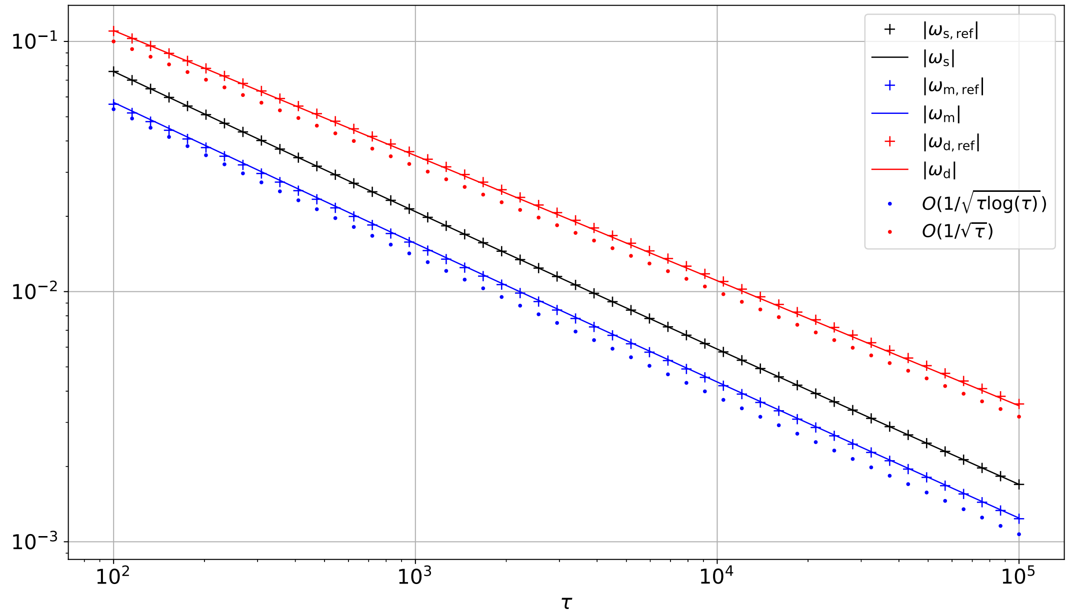

Figure 1: The asymptotic resonances ω_{s} (single particle), ω_{m} (monpole) and ω_{d} (dipole) given by the formulas in Propositions B.1 and Corollary 3.3 and the corresponding resonances obtained using reference solutions.

5 Concluding remarks

In this paper, we have provided the first mathematical model of resonant high-index dielectric nanoparticles. We have characterized their subwavelength resonances in terms of the eigenvalues of the associated Newtonian potential. We have also discussed the hybridization phenomenon of a dimer of dielectric nanoparticles with high refractive indices. Our results in this paper pave the way for the analysis, design, and manipulation of resonant dielectric nanostructures and their use as metamaterials. In particular, they can be used for mathematically and numerically modelling super-focusing in dielectric nanostructures, double-negative dielectric materials, and dielectric metasurfaces. Moreover, following [4,11], formula (7) can be easily generalized to the time-domain in order to characterize the temporal response of resonant dielectric nanoparticles and accelerate computations involving the temporal responses of subwavelength dielectric resonators.

A Proof of Proposition 2.5

Expansion (8) can be rewritten as

$$K_D^{k_m} = \hat{K}_D^{k_m} + K_D^{(0)} + O(\omega^2 \log(\omega)),$$

where

$$\begin{aligned}
\hat{K}_D^{k_m}[E] &= \log(\hat{\gamma} k_m)\hat{K}_D[E],\\
\hat{K}_D[E] &= -\frac{1}{2\pi}\int_D E(y)dy,\\
K_D^{(0)}[E] &= \int_D E(y)\Gamma(x-y)dy = -\frac{1}{2\pi}\int_D E(y)\log|\cdot - y|dy.
\end{aligned}$$

Using this expansion the resonance problem of finding $\omega \in \mathbb{C}$ such that there exists a solution $L^2(D) \ni E \not\equiv 0$ to (3) becomes

$$(I - \omega^2 \tau \varepsilon_c (\hat{K}_D^{k_m} + K_D^{(0)}))[E](x) = O(\omega^4 \log(\omega)).$$

Denote by

$$M_D^{k_m} := \hat{K}_D^{k_m} + K_D^{(0)},$$

which is self-adjoint as it is the sum of bounded self-adjoint operators.

Note that when

$$\omega = \frac{1}{\sqrt{\tau \varepsilon_c \lambda_0}},$$

where λ_0 belongs to $\sigma(K_D^{(0)})$, the spectrum of $K_D^{(0)}$, the following equation has a non-trivial solution:

$$(I - \omega^2 \tau \varepsilon_c K_D^{(0)})[E] = 0.$$

Analogously, when

$$\omega = \frac{1}{\sqrt{\tau \varepsilon_c \nu(\omega)}}, \tag{13}$$

where $\nu(\omega) \in \sigma(M_D^{k_m})$ the following equation, which is an approximation of our resonance problem up to order $O(\omega^4 \log(\omega))$, has a non-trivial solution:

$$(I - \omega^2 \tau \varepsilon_c M_D^{k_m})[E] = 0. \tag{14}$$

Consider the eigenvalue problem for $M_D^{k_m}$:

$$M_D^{k_m}[\Psi] = \nu(\omega)\Psi,$$

where $\Psi = \Psi(\omega)$ is normalized on $L^2(D)$. Using the expansions

$$\begin{aligned}\Psi(\omega) &= \Psi_0 + O(\frac{1}{\log \omega}),\\ \nu(\omega) &= \log(\omega)\nu_0 + \nu_1 + O(\frac{1}{\log \omega}),\end{aligned}$$

we have

$$(\log(\omega)\hat{K}_D + \log(\hat{\gamma}\sqrt{\varepsilon_m})\hat{K}_D + K_D^{(0)})[\Psi_0] = (\log(\omega)\nu_0 + \nu_1)[\Psi_0] + O(\frac{1}{\log \omega}).$$

Equating terms of $O(\log(\omega))$ gives

$$\hat{K}_D[\Psi_0] = \nu_0 \Psi_0.$$

As $\hat{K}_D$ is independent of $x \in D$, Ψ_0 must be a constant function, which we normalize on $L^2(D)$, i.e., $\Psi_0 = \hat{\mathbb{1}}_D$.

This gives

$$\nu_0 \hat{\mathbb{1}}_D = \hat{K}_D[\hat{\mathbb{1}}_D] = -\frac{|D|}{2\pi}\hat{\mathbb{1}}_D,$$

so $\nu_0 = -|D|/(2\pi)$. Next, equating terms of $O(1)$ we have

$$\begin{aligned}\nu_1 \hat{\mathbb{1}}_D &= (\log(\hat{\gamma}\sqrt{\varepsilon_m})\hat{K}_D + K_D^{(0)})[\hat{\mathbb{1}}_D]\\ &= -\frac{|D|}{2\pi}\log(\hat{\gamma}\sqrt{\varepsilon_m})\hat{\mathbb{1}}_D + K_D^{(0)}[\hat{\mathbb{1}}_D],\end{aligned}$$

and so, after taking the inner product with $\hat{\mathbb{1}}_D$ we get

$$\nu_1 = -\frac{|D|}{2\pi}\log(\hat{\gamma}\sqrt{\varepsilon_m}) + \langle K_D^{(0)}[\hat{\mathbb{1}}_D], \hat{\mathbb{1}}_D\rangle.$$

This means that

$$\nu(\omega) = -\frac{|D|}{2\pi}\log(\hat{\gamma}k_m) + \langle K_D^{(0)}[\hat{\mathbb{1}}_D], \hat{\mathbb{1}}_D\rangle + O(\frac{1}{\log\omega}). \tag{15}$$

Using the expansion of $\nu(\omega)$ in (15) we obtain from (13) that

$$1 - \omega_s^2\tau\varepsilon_c\bigg(- \frac{|D|}{2\pi}\log(\omega_s\hat{\gamma}\sqrt{\varepsilon_m}) + \langle K_D^{(0)}[\hat{\mathbb{1}}_D], \hat{\mathbb{1}}_D\rangle\bigg) = O(\frac{\omega^2}{\log\omega}) = o(\omega^2). \tag{16}$$

B The resonance for a unit disk

Let D be the unit disk. We can obtain a fully explicit expression for (16) in this case as the eigenvalues of the Newtonian potential have a direct relationship with the zeros of the Bessel function of order zero [13]. First we note that $\langle K_D^{(0)}[\hat{\mathbb{1}}_D], \hat{\mathbb{1}}_D\rangle = 1/\pi\langle K_D^{(0)}[\mathbb{1}_D], \mathbb{1}_D\rangle$. Now, let J_l be the Bessel function of order l and define $\mu_j^{(0)}$ by

$$J_0(\mu_j^{(0)}) = 0, \quad j = 1, 2, \ldots.$$

According to [13], the eigenvalues of the Newtonian potential for the unit disk are given by

$$\lambda_{0j} = \frac{1}{(\mu_j^{(0)})^2}, \quad j = 1, 2, \ldots,$$

with the associated orthornormal set of eigenfunctions $\{e_j\}_{j=1}^{\infty}$ given by

$$e_j(r) = \beta_j J_0\Big(\mu_j^{(0)} r\Big),$$

where

$$\beta_j = \frac{1}{\sqrt{\pi}J_1(\mu_j^{(0)})}.$$

Note that

$$\langle \mathbb{1}_D, e_j\rangle^2 = \frac{4\pi}{(\mu_j^{(0)})^2}.$$

Then we have

$$\begin{aligned}\langle K_D^{(0)}[\mathbb{1}_D], \mathbb{1}_D\rangle &= \sum_{j=1}^{\infty}\langle \mathbb{1}_D, e_j\rangle^2\lambda_{0j} = 4\pi\sum_{j=1}^{\infty}\frac{\lambda_{0j}}{(\mu_j^{(0)})^2} \\ &= 4\pi\sum_{j=1}^{\infty}\frac{1}{(\mu_j^{(0)})^4} = \frac{\pi}{8},\end{aligned}$$

where we used the identity $\sum_{j=1}^{\infty} 1/(\mu_j^{(0)})^4 = 1/32$ [19].

Therefore

$$\langle K_D^{(0)}[\hat{\mathbb{1}}_D], \hat{\mathbb{1}}_D\rangle = \frac{1}{8},$$

and (16) can be written as

$$1-\frac{\omega^2\tau\varepsilon_c}{2}\bigg(-\log(\hat{\gamma}k_m)+\frac{1}{4}\bigg)=o(\omega^2). \tag{17}$$

The precise dependence of ω on the contrast parameter τ can be found by writing the solution to (17) in terms of the Lambert W function [18].

Proposition B.1. *The resonance for a unit disk as the contrast $\tau\to\infty$ is given by*

$$\omega_s(\tau)=-\frac{2i}{\sqrt{\tau\varepsilon_c W(\Phi(\tau))}}+o\bigg(\frac{1}{\tau\log(\tau)}\bigg), \tag{18}$$

where

$$\Phi(\tau)=-\frac{4}{\tau\varepsilon_c}\exp\bigg(2\log(\sqrt{\varepsilon_m}\hat{\gamma})-\frac{1}{2}\bigg),$$

and W is the lower branch of the Lambert W function defined in the interval $[-1/e,0)$.

Proof. Denote by

$$\begin{aligned}
\alpha_0(\tau)&:=\frac{\tau\varepsilon_c}{2},\\
\alpha_1(\tau)&:=\alpha_0\bigg(\log(\sqrt{\varepsilon_m}\hat{\gamma})-\frac{1}{4}\bigg),\\
\alpha_2(\tau)&:=-\frac{2}{\alpha_0}\exp\bigg(\frac{2\alpha_1}{\alpha_0}\bigg).
\end{aligned}$$

Then we can write (17) as

$$1+\alpha_0\omega^2\log(\omega)+\alpha_1\omega^2=o(\omega)^2,$$

which leads to

$$-2\log(\omega)=\frac{2+2\alpha_1\omega^2}{\alpha_0\omega^2}+o(1).$$

Then

$$\frac{1}{\omega^2}=\exp\bigg(\frac{2+2\alpha_1\omega^2}{\alpha_0\omega^2}\bigg)+o(1).$$

Next we have

$$\frac{-2}{\alpha_0\omega^2}\exp\bigg(-\frac{2}{\alpha_0\omega^2}\bigg)=-\frac{2}{\alpha_0}\exp\bigg(\frac{2\alpha_1}{\alpha_0}\bigg)+o(\exp(-1/\omega^2)).$$

The Lambert W function is a map $ze^z\to W(ze^z)=z$ [18] and as the expression above is in this form, an application of this map leads to

$$\begin{aligned}
\frac{-2}{\alpha_0\omega^2}&=W\bigg(\frac{2}{\alpha_0}\exp\bigg(-\frac{2\alpha_1}{\alpha_0}\bigg)\bigg)+o(1)\\
&=W(\alpha_2)+o(1).
\end{aligned}$$

Note that the Lambert W function is double-valued in the interval $[-1/e,0)$, which is the interval we need to consider when $\tau\to\infty$, and we should choose the lower branch, denoted by W_{-1} in the literature, to obtain a physically meaningful resonance.

Now

$$\omega^2=-\frac{2}{\alpha_0 W(\alpha_2)}+o(\omega^2),$$

and so

$$\omega = \pm \frac{i\sqrt{2}}{\sqrt{\alpha_0 W(\alpha_2)}} + o(\omega^2). \tag{19}$$

Noting that $\alpha_2 = O(1/\tau)$, as $\tau \to \infty$ we have the expansion [18]

$$W(\alpha_2) = \log(-\alpha_2) - \log(-\log(-\alpha_2)) + \frac{\log(-\log(-\alpha_2))}{\log(-\alpha_2)} + \dots,$$

which implies that $W(\alpha_2) = O(\log(\tau))$. As $\alpha_0 = O(\tau)$ this means we have

$$\omega + o(\omega^2) = O\left(\frac{1}{\sqrt{\tau \log(\tau)}}\right),$$

and squaring both sides gives

$$\omega^2 + o(\omega^3) = O\left(\frac{1}{\tau \log(\tau)}\right).$$

Thus, upon taking the term with positive real part in (19) we obtain (18). □

A coarser approximation which gives a clearer qualitative indication of the dependence of the resonance frequency on the system variables is given in the following corollary.

Corollary B.2. *As $\tau \to \infty$ it holds that*

$$\omega_{\mathrm{s}}(\tau) = \frac{2}{\sqrt{\tau \varepsilon_c \log(\tau)}} \frac{1}{\varphi(\tau)} + o\left(\frac{1}{\sqrt{\tau \log(\tau)}}\right),$$

where

$$\varphi(\tau) = \left(1 - \frac{\log(\frac{\varepsilon_m}{\varepsilon_c}\hat{\gamma}^2) + \frac{1}{2}}{2\log(\tau)}\right).$$

Proof. We have

$$\begin{aligned} W(\alpha_2) &= \log(-\alpha_2) + \dots \\ &= -\log(\tau) + \log\left(\frac{\varepsilon_m}{\varepsilon_c}\hat{\gamma}^2\right) + \frac{1}{2} + \dots. \end{aligned}$$

Then

$$\begin{aligned} \sqrt{W(\alpha_2)} &= \sqrt{-\log(\tau\delta^2)}\sqrt{\left(1 - \frac{\log\left(\frac{\varepsilon_m}{\varepsilon_c}\hat{\gamma}^2\right) + \frac{1}{2}}{\log(\tau\delta^2)}\right) + \dots} \\ &= i\sqrt{\log(\tau)}\left(1 - \frac{\log\left(\frac{\varepsilon_m}{\varepsilon_c}\hat{\gamma}^2\right) + \frac{1}{2}}{2\log(\tau)}\right) + \dots. \end{aligned}$$

Substituting this expression for $\sqrt{W(\alpha_2)}$ into (18) asserts the claim. □

C Proof of Proposition 3.2

We have $|D_1| = |D_2| = \pi\delta^2$. Since

$$
\begin{aligned}
K_{D_i}^{k_m} &= M_{D_i}^{k_m} + O(\omega^2 \log(\omega)),\\
R_{D_i,D_j}^{k_m} &= N_{D_i,D_j}^{k_m} + O(\omega^2 \log(\omega)),
\end{aligned}
$$

it holds that

$$
\begin{pmatrix} I - \tau\omega^2\varepsilon_c M_{D_i}^{k_m} & -\tau\omega^2\varepsilon_c N_{D_2,D_1}^{k_m} \\ -\tau\omega^2\varepsilon_c N_{D_1,D_2}^{k_m} & I - \tau\omega^2\varepsilon_c M_{D_2}^{k_m} \end{pmatrix} \begin{pmatrix} E|_{D_1} \\ E|_{D_2} \end{pmatrix} = \begin{pmatrix} O(\omega^4 \log(\omega)) \\ O(\omega^4 \log(\omega)) \end{pmatrix}. \tag{20}
$$

Denote by

$$
\begin{aligned}
\hat{\nu}(\omega) &= -\frac{\delta^2}{2}\log(\hat{\gamma}k_m) + \langle K_{D_1}^{(0)}[\hat{\mathbb{1}}_{D_1}], \hat{\mathbb{1}}_{D_1}\rangle\\
&= -\frac{\delta^2}{2}\log(\hat{\gamma}k_m) + \langle K_{D_2}^{(0)}[\hat{\mathbb{1}}_{D_2}], \hat{\mathbb{1}}_{D_2}\rangle,\\
\hat{\eta}(\omega) &= \langle N_{D_1,D_2}^{k_m}[\hat{\mathbb{1}}_{D_1}], \hat{\mathbb{1}}_{D_2}\rangle\\
&= \langle N_{D_2,D_1}^{k_m}[\hat{\mathbb{1}}_{D_2}], \hat{\mathbb{1}}_{D_1}\rangle,
\end{aligned}
$$

with these equalities holding due to the symmetry of the dimer. Furthermore, the symmetry of the dimer also means that

$$
\hat{K}_{D_i,D_j}^{k_m}[\hat{\mathbb{1}}_{D_i}] = \hat{K}_{D_j}^{k_m}[\hat{\mathbb{1}}_{D_j}]. \tag{21}
$$

Denote by $\nu(\omega)$ the eigenvalues of the operators $M_{D_i}^{k_m}$ such that

$$
\langle M_{D_1}^{k_m}[\Psi_{D_1}], \Psi_{D_1}\rangle = \nu(\omega) = \langle M_{D_2}^{k_m}[\Psi_{D_2}], \Psi_{D_2}\rangle,
$$

for eigenfunctions $L^2(D_i) \ni \Psi_{D_i}(\omega) = \Psi_{D_i,0} + O(\frac{1}{\log\omega}) = \hat{\mathbb{1}}_{D_i} + O(\frac{1}{\log\omega})$.

From Section A we know that

$$
\langle M_{D_i}^{k_m}[\Psi_{D_i}], \Psi_{D_i}\rangle = \nu(\omega) = \hat{\nu}(\omega) + o(1). \tag{22}
$$

Denote by $\eta(\omega) = \langle N_{D_i,D_j}^{k_m}[\Psi_{D_i}], \Psi_{D_j}\rangle$. Note also that

$$
\eta(\omega) = \hat{\eta}(\omega) + o(1).
$$

Therefore, we have the following implicit equation for the hybridized resonances,

$$
1 - \omega^2\tau\varepsilon_c\Big(- \frac{\delta^2}{2}\log(\omega\hat{\gamma}\sqrt{\varepsilon_m})(1 \pm 1) + \langle K_{D_1}^{(0)}[\hat{\mathbb{1}}_{D_1}]\hat{\mathbb{1}}_{D_1}\rangle \pm \langle R_{D_2,D_1}^{(0)}[\hat{\mathbb{1}}_{D_2}], \hat{\mathbb{1}}_{D_1}\rangle\Big) = o(\omega^2).
$$

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