First-order perturbation theory of eigenmodes for systems with interfaces

Z. Sztranyovszky,[1,2] W. Langbein,[1] and E. A. Muljarov[1]
[1]*School of Physics and Astronomy, Cardiff University, Cardiff CF24 3AA, United Kingdom*
[2]*School of Chemical Engineering, University of Birmingham, Birmingham B15 2TT, United Kingdom*

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We present an exact first-order perturbation theory for eigenmodes in systems with interfaces causing material discontinuities. We show that when interfaces deform, higher-order terms of the perturbation series can contribute to the eigenmode frequencies in first order in the deformation depth. In such cases, the first-order approximation is different from the usual diagonal approximation and its single-mode result. Extracting additional first-order corrections from all higher-order terms enables us to recover the diagonal formalism in a modified form. A general formula for the single-mode first-order correction to electromagnetic eigenmodes in systems with interfaces is derived, capable of treating dispersive, magnetic, and chiral materials of arbitrary shape.

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I. INTRODUCTION

Eigenmodes, which are solutions to a differential equation of Sturm-Liouville type with a set of boundary conditions, are used to describe physical phenomena across physics, including gravitational astronomy [1], acoustics [2], seismology [3], quantum mechanics (QM) [4], and electromagnetism (EM) [5]. The eigenmodes of open systems are also referred to as resonant states (RSs) [6], or quasinormal modes [7]. They determine the optical properties of a resonator, such as its scattering cross-section or Purcell enhancement [8]. For simple electromagnetic systems, such as a slab or a sphere, the RSs can be found analytically [9]. For more complicated shapes they can be found numerically [7,10] or via perturbative approaches [7,8]. The resonant-state expansion (RSE) is a method that treats perturbations in all perturbation orders by transforming Maxwell's equations into a matrix eigenvalue problem [6]. Its accuracy is controlled by the selection of eigenmodes in the basis.

For small changes of the system, it is sufficient to take only a few suited RSs in the basis, or even a single one in a nondegenerate case. The latter corresponds to the diagonal approximation in terms of the matrix equation, and in certain cases this can also be equivalent to the first-order approximation, although not necessarily, as we will show in this paper. Following the terminology of Ref. [11], we distinguish two different kinds of perturbations: *volume* perturbation (VP) and *boundary* perturbation (BP). A VP is a small change of the medium properties over a finite volume, for example, in QM a small change in the potential over the width of a quantum well, or in acoustics a small change of the density of the medium. A BP instead moves the spatial position of a medium interface with a discontinuity in medium properties, such as changing the width of a quantum well in QM. In EM, VPs could be a small change of a resonator's permittivity $\Delta\varepsilon$, for which first- and second-order results are well known [12,13], or a change of the medium surrounding the resonator [14,15]. The first-order results for VPs in EM correspond to the diagonal approximation in the RSE matrix equation, and they include an overlap integral of the eigenmode field with the perturbation (e.g., $\Delta\varepsilon$), in complete analogy with conventional QM [16]. For BPs, this approach is not suited, because the local change of the medium property is not small. Instead, the deformation depth $h(\boldsymbol{r})$, which is the shift of the surface at position $\boldsymbol{r}$, plays the role of a small perturbation parameter, and an interesting consequence arises from the boundary conditions. For an open system, these are *outgoing waves* [17], which cannot be expressed as a combination of Neumann and Dirichlet boundary conditions, making the approach of Ref. [11] inapplicable. The underlying cause for the different treatment of BPs required in EM is the discontinuity of the normal component of the electric field at a material boundary [18]. We note that similar effects can also occur in condensed matter physics when the effective mass in Schrödinger's equation is discontinuous, or in acoustics at the boundary between two media with different densities. In EM, the first-order correction to the RS frequency for a BP was treated for closed isotropic dielectric systems by using the electric displacement field normal to the surface [19], and for cavities with perfect electric conductor walls [20,21]. In case of isotropic open dispersive systems, it was also recognized that the VP diagonal matrix element does not give the correct first-order results, and an alternative treatment was found based on distinguishing the electric fields inside and outside the resonator [22], and Taylor expanding them [7]. The two approaches [7,19] are equivalent apart from the frequency-dependent permittivity and the different field normalization for open systems [7]. It is also known that in general zero-frequency [23] or zero-permittivity [24] longitudinal modes need to be included in the basis alongside the RSs; however, the contribution of these additional modes to the first-order results have not been considered in detail. Notably, the above BP methods [7,19] do not treat VPs, and the mentioned VP methods do not treat BPs.

The purpose of this article is twofold. Firstly, we show that when treating a BP by applying the standard perturbation theory valid for VP, all orders of the perturbation series can contribute linearly in h. Using EM for illustration, we demonstrate this surprising finding in terms of static (zero-frequency) modes of a nondispersive open optical system.

Secondly, we derive a unified treatment of small BP and VP, describing correctly the first-order RS wave number change, linear in h and $\Delta\varepsilon$, respectively. This treatment is generalized to include frequency dispersion, arbitrary media (including magnetic and chiral), and arbitrary shape. Illustrations for both spherical and nonspherical dispersive systems are provided.

II. SECOND AND HIGHER ORDERS OF PERTURBATION THEORY: LINEAR CONTRIBUTION FOR BOUNDARY PERTURBATIONS

For clarity of presentation, we start by considering an unperturbed dielectric system described by a nondispersive permittivity tensor $\hat{\varepsilon}(\mathbf{r})$ with known RSs having wave numbers k_n and electric fields $\mathbf{E}_n(\mathbf{r})$. This system is perturbed by a change of the permittivity $\Delta\hat{\varepsilon}(\mathbf{r})$. In the RSE approach [6,13], the electric field $\mathbf{E}(\mathbf{r})$ of a perturbed RS is expanded as

$$\mathbf{E}(\mathbf{r}) = \sum_{\nu} c_\nu \mathbf{E}_\nu(\mathbf{r}), \tag{1}$$

leading to a matrix eigenvalue problem

$$(k - k_\nu)c_\nu = -k \sum_{\nu'} V_{\nu\nu'} c_{\nu'}, \tag{2}$$

which determines the exact values of the perturbed RS wave numbers k and the expansion coefficients c_ν in the limit of all unperturbed modes included in the summation. Here, index ν labels both the RSs ($\nu = n$, with $k_n \neq 0$) and static modes ($\nu = \lambda$, with $k_\lambda = 0$ [23]), and the matrix elements of the perturbation have the form

$$V_{\nu\nu'} = \int \mathbf{E}_\nu(\mathbf{r}) \cdot \Delta\hat{\varepsilon}(\mathbf{r}) \mathbf{E}_{\nu'}(\mathbf{r}) d\mathbf{r}, \tag{3}$$

where all fields $\mathbf{E}_\nu$ are properly normalized [6,25]. From the exact RSE equation (2) one can extract, in the spirit of a standard perturbation theory [16], corrections to the eigenvalue k_n in all orders, in a form of an infinite series [13]

$$k = k_n - k_n V_{nn} + k_n V_{nn}^2 + k_n^2 \sum_{\nu \neq n} \frac{V_{n\nu} V_{\nu n}}{k_n - k_\nu} + \ldots, \tag{4}$$

which suggests that

$$k^{(1)} = -k_n V_{nn} \tag{5}$$

is the first-order correction to the wave number k_n. In fact, each matrix element Eq. (3) is linear both in the permittivity perturbation $\Delta\hat{\varepsilon}$ and in the deformation depth h in lowest order.

The above first-order correction $k^{(1)}$ is illustrated in Fig. 1 for transverse-magnetic (TM) modes of a dielectric sphere of radius R in vacuum, with angular momentum $l = 1$, for a BP changing the radius of the sphere by h. The real part of the wave number $\mathrm{Re}(k)$ gives the position of the resonance, and the imaginary part $\mathrm{Im}(k)$ gives the decay rate, or the half width at half maximum. Clearly, for the fundamental mode, Eq. (5)

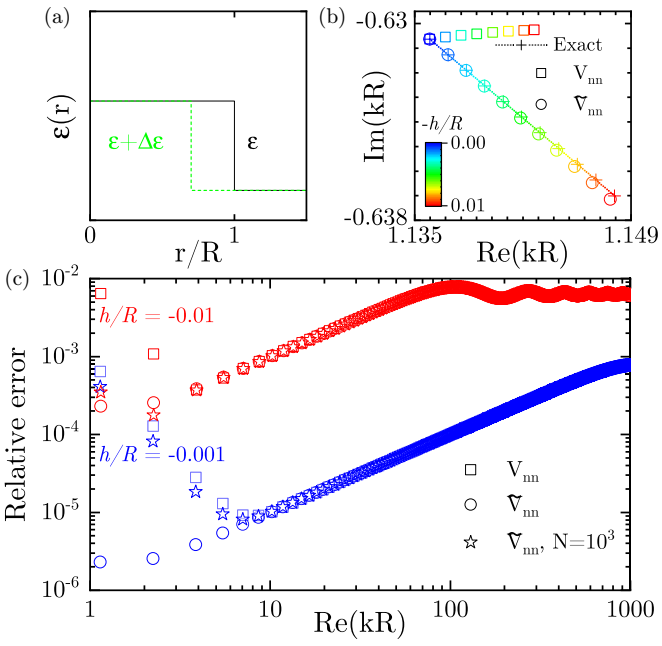

FIG. 1. (a) Schematic illustration of a BP as a permittivity perturbation of the sphere, with its radius R changing by h. (b) Effect of the BP on the wave number k of first RS with orbital number $l = 1$ in a sphere of permittivity $\varepsilon = 4$ surrounded by vacuum. (c) Relative error of RS wave numbers calculated without static modes (squares), with all static modes (circles), and with 1000 static modes included (stars), for two different BPs as given.

(squares), does not describe correctly the first-order changes of the RS wave number k, as a deviation linear in h is observed implying that first-order contributions to k are missing.

The origin of this mismatch lies in the role of static modes, which surprisingly can contribute linearly in h via the second-order sum, $k_n \sum_\lambda V_{n\lambda} V_{\lambda n}$, and also via all higher-order terms of the perturbation series Eq. (4). To take their cumulative effect into account, let us write the RSE equation (2) in terms of the RSs only, by using the $k_\lambda = 0$ degeneracy of static modes [26]

$$(k - k_n)c_n = -k \sum_{n'} \tilde{V}_{nn'} c_{n'}, \tag{6}$$

where

$$\tilde{V}_{nn'} = V_{nn'} - \sum_{\lambda\lambda'} V_{n\lambda} W_{\lambda\lambda'} V_{\lambda' n} \tag{7}$$

and the matrix $W_{\lambda\lambda'}$ is the inverse of $\delta_{\lambda\lambda'} + V_{\lambda\lambda'}$ with $\delta_{\lambda\lambda'}$ being the Kronecker delta. The full linear correction to the RS wave number is then given by

$$\tilde{k}^{(1)} = -k_n \tilde{V}_{nn}. \tag{8}$$

To evaluate the sum in Eq. (7), we use a Neumann series expansion $W = (I + V)^{-1} = I - V + V^2 - V^3 + \ldots$, where W (V) is a matrix with elements $W_{\lambda\lambda'}$ ($V_{\lambda\lambda'}$) and I is the identity matrix. Substituting it into Eq. (7) results in an infinite series

$$\tilde{V}_{nn'} = V_{nn'} - \sum_{\lambda} V_{n\lambda} V_{\lambda n'} + \sum_{\lambda\lambda'} V_{n\lambda} V_{\lambda\lambda'} V_{\lambda' n'} - \ldots, \tag{9}$$

which we evaluate below to first order in h, using the static pole residue of the dyadic Green's function (here, its electric part only, which is sufficient for permittivity perturbation).

For a spherically symmetric dielectric systems with permittivity $\varepsilon(r)$, the residue can be written explicitly as (see Appendix A 4)

$$\sum_{\lambda} \mathbf{E}_{\lambda}(\mathbf{r}) \otimes \mathbf{E}_{\lambda}(\mathbf{r}') = \frac{\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}}{\varepsilon(r)} \delta(\mathbf{r} - \mathbf{r}') + \hat{\mathbf{R}}(\mathbf{r}, \mathbf{r}'), \tag{10}$$

where the tensor $\hat{\mathbf{R}}(\mathbf{r}, \mathbf{r}')$ is a regular part of the residue, $\hat{\mathbf{r}}$ is the unit vector in the radial direction, and $\otimes$ denotes the dyadic product. Using Eq. (10) for each sum over static modes in Eq. (9), one can see that the δ function in Eq. (10) eliminates one volume integration, reducing each term in Eq. (9) to a single volume integral, proportional to h. Furthermore, the contribution of the regular part can be neglected in linear order in h, as it comes with an additional volume integral, and hence is of higher order. Summing over all orders, we arrive, after some algebra, at

$$\begin{aligned} \frac{\tilde{k}^{(1)}}{k_n} &= -\int \mathbf{E}_n \cdot \left[\hat{\mathbf{1}} + \frac{\Delta\hat{\varepsilon}(\mathbf{r})}{\varepsilon(r)} \hat{\mathbf{r}} \otimes \hat{\mathbf{r}}\right]^{-1} \Delta\hat{\varepsilon}(\mathbf{r}) \mathbf{E}_n d\mathbf{r} \\ &= -\int \left[\mathbf{E}_n^{\parallel} \cdot \Delta\varepsilon(\mathbf{r}) \mathbf{E}_n^{\parallel} + \mathbf{E}_n^{\perp} \cdot \frac{\varepsilon(r)\Delta\varepsilon(\mathbf{r})}{\varepsilon(r) + \Delta\varepsilon(\mathbf{r})} \mathbf{E}_n^{\perp}\right] d\mathbf{r}, \end{aligned} \tag{11}$$

where $\hat{\mathbf{1}}$ is the identity tensor, and the perturbation $\Delta\varepsilon(\mathbf{r})$ in the second line is assumed isotropic but not necessarily spherically symmetric. The superscript $\parallel$ ($\perp$) labels the vector component parallel (normal) to the interface of the basis system. More details of the derivation of the above equations and their extension to magnetic and chiral materials are provided in Appendix A.

Figure 1 demonstrates that Eq. (11) (circles) correctly describes the effect of the size perturbation in first order. In fact, comparing $h/R = -0.01$ and -0.001, one can see that the residual error scales quadratically, i.e., is of second order in h, as expected. We also show in Fig. 1 the error for the RS wave numbers calculated with an explicit use of $N = 10^3$ static modes [27] in $\tilde{V}_{nn}$ via Eq. (7) (stars). This demonstrates that as h gets smaller, even the use of a large number of static modes carries a significant truncation error in Eq. (10), leading to a poor representation of the δ function and an increased error of the eigenmodes compared to Eq. (11). For higher l, the static modes can still contribute to the RSs in first order, although the effect is less pronounced due to the higher frequencies of the RSs (see Appendix B 2). Note that, while the total number of static modes is countably infinite, the freedom of choosing such a set, granted by their wave number degeneracy, allows one to concentrate the effect of the boundary shift in a single-mode contribution [28].

As it is clear from Fig. 1 and the above derivation, the first-order term of the standard perturbation series Eq. (4) does not contain all first-order effects of the BP. Instead, additional first-order terms can be found in all orders of the perturbation theory. We emphasize that this occurs in any area of physics describing wave phenomena, and we give in Appendix D examples from condensed matter and acoustics illustrating this effect.

III. UNIFIED TREATMENT OF VOLUME AND BOUNDARY PERTURBATIONS FOR ARBITRARY SYSTEMS

The first-order result Eq. (11) was for clarity obtained for a spherically symmetric nondispersive system. We now generalize Eq. (11) to optical systems with (i) any geometry, (ii) magnetic and chiral materials, and (iii) arbitrary frequency dispersion. To do this, we write Maxwell's equations for the unperturbed system in the compact form [25]

$$[k_n \hat{\mathbb{P}}_0(k_n, \mathbf{r}) - \hat{\mathbb{D}}(\mathbf{r})] \vec{\mathbb{F}}_n(\mathbf{r}) = 0, \tag{12}$$

where

$$\hat{\mathbb{P}}_0 = \begin{pmatrix} \hat{\boldsymbol{\varepsilon}} & -i\hat{\boldsymbol{\xi}} \\ i\hat{\boldsymbol{\zeta}} & \hat{\boldsymbol{\mu}} \end{pmatrix}, \quad \hat{\mathbb{D}} = \begin{pmatrix} \hat{\mathbf{0}} & \nabla\times \\ \nabla\times & \hat{\mathbf{0}} \end{pmatrix}, \quad \vec{\mathbb{F}}_n = \begin{pmatrix} \mathbf{E}_n \\ i\mathbf{H}_n \end{pmatrix}, \tag{13}$$

and $\hat{\mathbf{0}}$ is the 3×3 zero matrix. $\hat{\mathbb{P}}_0(k, \mathbf{r})$ is a 6×6 tensor describing the system, which consists of frequency-dispersive tensors of permittivity $\hat{\boldsymbol{\varepsilon}}(k, \mathbf{r})$, permeability $\hat{\boldsymbol{\mu}}(k, \mathbf{r})$, and bi-anisotropy $\hat{\boldsymbol{\xi}}(k, \mathbf{r})$ and $\hat{\boldsymbol{\zeta}}(k, \mathbf{r})$. $\vec{\mathbb{F}}_n(\mathbf{r})$ is a 6×1 vector comprising $\mathbf{E}_n(\mathbf{r})$ and $\mathbf{H}_n(\mathbf{r})$, the electric and magnetic fields of the RS with the wave number k_n. Applying a perturbation $\Delta\hat{\mathbb{P}}(k, \mathbf{r})$ of the generalized permittivity, the electromagnetic field and the wave number of this RS change, respectively, to $\vec{\mathbb{F}}(\mathbf{r})$ and k, which in turn satisfy perturbed Maxwell's equations

$$[k\hat{\mathbb{P}}(k, \mathbf{r}) - \hat{\mathbb{D}}(\mathbf{r})] \vec{\mathbb{F}}(\mathbf{r}) = 0 \tag{14}$$

with $\hat{\mathbb{P}}(k, \mathbf{r}) = \hat{\mathbb{P}}_0(k, \mathbf{r}) + \Delta\hat{\mathbb{P}}(k, \mathbf{r})$ of the perturbed system. For clarity of presentation, we assume below isotropic and reciprocal materials; anisotropy is considered in Appendix F, and a further generalization to nonreciprocal materials is possible [29]. Multiplying Eq. (12) with $\vec{\mathbb{F}}$ and Eq. (14) with $\vec{\mathbb{F}}_n$, integrating both equations over volume V_1, which contains the original system volume and the perturbation, taking the difference between the results, and applying the divergence theorem to the terms with $\hat{\mathbb{D}}$ operators [25], we obtain

$$\begin{aligned} &\int_{V_1} \vec{\mathbb{F}}_n(\mathbf{r}) \cdot [k_n \hat{\mathbb{P}}_0(k_n, \mathbf{r}) - k\hat{\mathbb{P}}(k, \mathbf{r})] \vec{\mathbb{F}}(\mathbf{r}) d\mathbf{r} \\ &\quad = i \oint_{S_1} [\mathbf{E}_n(\mathbf{r}) \times \mathbf{H}(\mathbf{r}) - \mathbf{E}(\mathbf{r}) \times \mathbf{H}_n(\mathbf{r})] \cdot d\mathbf{S}, \end{aligned} \tag{15}$$

where S_1 is the boundary of V_1.

To extract from Eq. (15) the first-order correction $\tilde{k}^{(1)}$ to the wave number, we introduce a real vector field $\hat{\mathbf{n}}(\mathbf{r})$, which is normal to both the surface of the perturbed and unperturbed system, and is normalized at each point as $|\hat{\mathbf{n}}(\mathbf{r})| = 1$ (if there are other surfaces with material discontinuities, $\hat{\mathbf{n}}$ should be chosen normal also to them, see Fig. 2 for illustration). Then we introduce a normal component of the perturbed field,

$$\vec{\mathbb{F}}^{\perp}(\mathbf{r}) = \begin{pmatrix} \mathbf{E}^{\perp}(\mathbf{r}) \\ i\mathbf{H}^{\perp}(\mathbf{r}) \end{pmatrix} = \begin{pmatrix} \hat{\mathbf{n}}(\mathbf{r})[\hat{\mathbf{n}}(\mathbf{r}) \cdot \mathbf{E}(\mathbf{r})] \\ \hat{\mathbf{n}}(\mathbf{r})[\hat{\mathbf{n}}(\mathbf{r}) \cdot i\mathbf{H}(\mathbf{r})] \end{pmatrix}. \tag{16}$$

The tangential component is then given by $\vec{\mathbb{F}}^{\parallel} = \vec{\mathbb{F}} - \vec{\mathbb{F}}^{\perp}$. Now, according to Maxwell's boundary conditions, fields $\vec{\mathbb{F}}^{\parallel}(\mathbf{r})$ and $\hat{\mathbb{P}}(k, \mathbf{r})\vec{\mathbb{F}}^{\perp}(\mathbf{r})$ are continuous everywhere. Similarly, the unperturbed fields $\vec{\mathbb{F}}_n^{\parallel}(\mathbf{r})$ and $\hat{\mathbb{P}}_0(k_n, \mathbf{r})\vec{\mathbb{F}}_n^{\perp}(\mathbf{r})$,

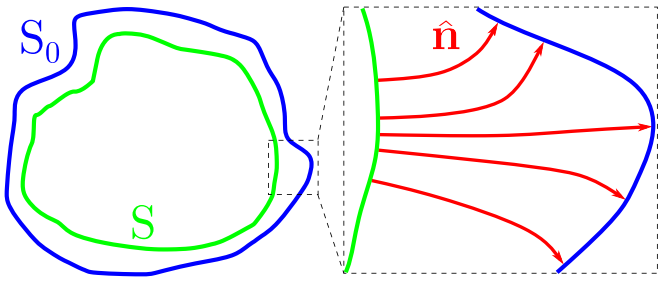

FIG. 2. Sketch of the unperturbed and perturbed systems occupying the volumes V_0 and V and having surfaces S_0 and S, respectively. The vector field $\hat{\mathbf{n}}(\mathbf{r})$ is normal to both S_0 and S.

introduced in the same manner, are also continuous. Then, approximating $\vec{\mathbb{F}}^{\parallel}(\mathbf{r}) \approx \vec{\mathbb{F}}^{\parallel}_n(\mathbf{r})$ and $\hat{\mathbb{P}}(k,\mathbf{r})\vec{\mathbb{F}}^{\perp}(\mathbf{r}) \approx \hat{\mathbb{P}}_0(k_n,\mathbf{r})\vec{\mathbb{F}}^{\perp}_n(\mathbf{r})$, which is sufficient for determining the wave number k to first order, we use in Eq. (15)

$$\vec{\mathbb{F}}(\mathbf{r}) = \vec{\mathbb{F}}^{\parallel}_n(\mathbf{r}) + \hat{\mathbb{P}}^{-1}(k,\mathbf{r})\hat{\mathbb{P}}_0(k_n,\mathbf{r})\vec{\mathbb{F}}^{\perp}_n(\mathbf{r}), \tag{17}$$

where $\hat{\mathbb{P}}^{-1}$ is the inverse of $\hat{\mathbb{P}}$. Finally, applying a Taylor expansion $k\hat{\mathbb{P}}(k,\mathbf{r}) = k_n\hat{\mathbb{P}}(k_n,\mathbf{r}) + [k\hat{\mathbb{P}}(k,\mathbf{r})]'(k-k_n) + \ldots$ and $\vec{\mathbb{F}}(\mathbf{r}) = \vec{\mathbb{F}}_n(\mathbf{r}) + \vec{\mathbb{F}}'_n(\mathbf{r})(k-k_n) + \ldots$ for the field outside the systems and keeping only terms linear in $k-k_n$, we arrive, after some algebra (see Appendix E), at

$$\frac{\tilde{k}^{(1)}}{k_n} = \frac{-\int[\vec{\mathbb{F}}^{\parallel}_n \cdot \Delta\hat{\mathbb{P}}\vec{\mathbb{F}}^{\parallel}_n + \vec{\mathbb{F}}^{\perp}_n \cdot \hat{\mathbb{P}}_0\hat{\mathbb{P}}^{-1}\Delta\hat{\mathbb{P}}\vec{\mathbb{F}}^{\perp}_n]d\mathbf{r}}{\int_{V_0}\vec{\mathbb{F}}_n\cdot[k\hat{\mathbb{P}}_0]'\vec{\mathbb{F}}_n d\mathbf{r} + i\oint_{S_0}[\mathbf{E}_n\times\mathbf{H}'_n - \mathbf{E}'_n\times\mathbf{H}_n]\cdot d\mathbf{S}}, \tag{18}$$

where V_0 is the unperturbed system volume, S_0 is its boundary, and the prime indicates the derivative with respect to k, with all quantities taken at $k = k_n$. Equation (18) is a generalization of Eq. (11), which is valid for small perturbations both *inside and outside* the basis system, including deformation outwards. Note that we have not assumed so far any specific normalization of $\vec{\mathbb{F}}_n(\mathbf{r})$. The analytic normalization introduced in [6,25] ensures that the denominator in Eq. (18) is equal to 1.

It is important to note that Eqs. (11) and (18) contain the exact first-order correction both in terms of the permittivity change ($\Delta\hat{\varepsilon}$ or $\Delta\hat{\mathbb{P}}$) and in the deformation depth h. They also include higher-order corrections, which are not exact. For simplicity we assumed nondegenerate modes in the above derivation. To find the first-order correction to degenerate modes, a matrix equation similar to Eq. (6), including only degenerate states, will need to be diagonalized.

The above derivation provides a clue for understanding the demonstrated phenomenon that the standard perturbation series Eq. (4) can have contributions to the RS wave number, which are linear in h in all perturbation orders. The zeroth-order approximation of the field Eq. (17), which is the key point of the derivation, is different from the standard expansion Eq. (1) used for a single mode. The failure to extract the correct first order from a series like Eq. (4) technically arises from approximating discontinuous functions with continuous ones. Further illustrations of this fact and a link to the completeness of the basis functions are provided in Appendix C.

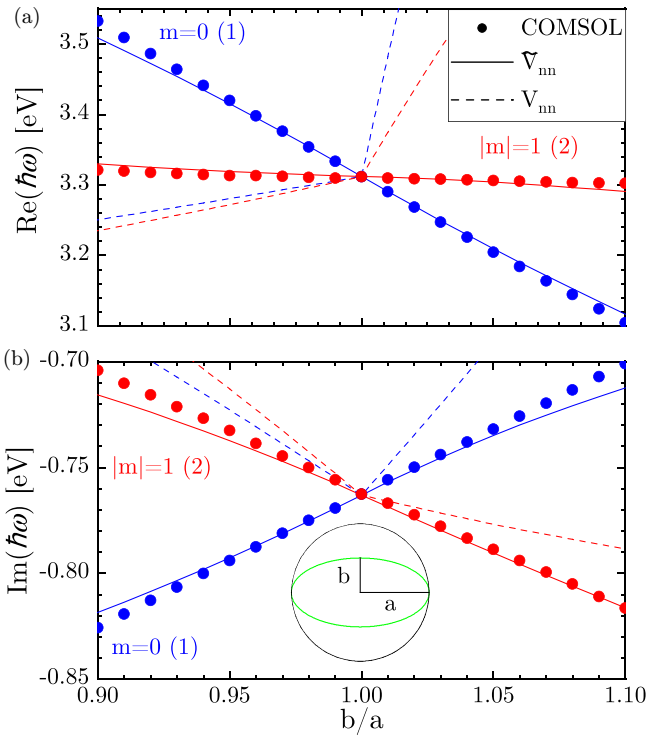

FIG. 3. (a) Real and (b) imaginary parts of the wave number of the dipolar surface plasmon mode of a silver sphere perturbed to an ellipsoid, as sketched in the inset of (b). The mode degeneracy is shown in brackets, and m is its magnetic quantum number. The COMSOL data is taken from [7].

IV. ILLUSTRATIONS FOR DISPERSIVE AND NONSPHERICAL SYSTEMS

We now demonstrate the first-order formula Eq. (18) on a system with frequency dispersion. Figures 3(a) and 3(b) show, respectively, the real and imaginary parts of k for the dipolar surface plasmon mode of a silver sphere being distorted into an ellipsoid. The permittivity of silver is given by the Drude model: $\epsilon(\omega) = 1 - \omega_p^2/(\omega^2 + i\omega\gamma)$ with $\hbar\omega_p = 9$ eV and $\hbar\gamma = 0.021$ eV, as used in [7]. Calculation of the matrix elements of the perturbation are detailed in Appendix G. The perturbation theory Eq. (18) (solid lines) agrees in first order in h with eigenfrequencies calculated numerically with COMSOL Multiphysics [30] (circles), for both inward ($a < R$) and outward ($a > R$) perturbations of the silver sphere. The results using $k^{(1)}$ are also shown for comparison (dashed lines), and are clearly incorrect. This example was chosen identical to the one used in [7], where the first-order RSE was taken as $k^{(1)}$ given by Eq. (5), even though earlier papers [23,26] indicate that static modes could contribute in first order. Thus the statement in Ref. [7] that the RSE is providing an incorrect first-order result was premature.

Finally, we consider the perturbation of a nonspherical system. In such systems the modes cannot be separated into TM and transverse-electric (TE) polarizations, nor into angular momentum numbers l, and exact analytic solutions for the modes are not available. We use a cylinder [23] of height h, diameter $d = h$, and permittivity $\epsilon = 9$. We calculate the unperturbed modes numerically using COMSOL Multiphysics

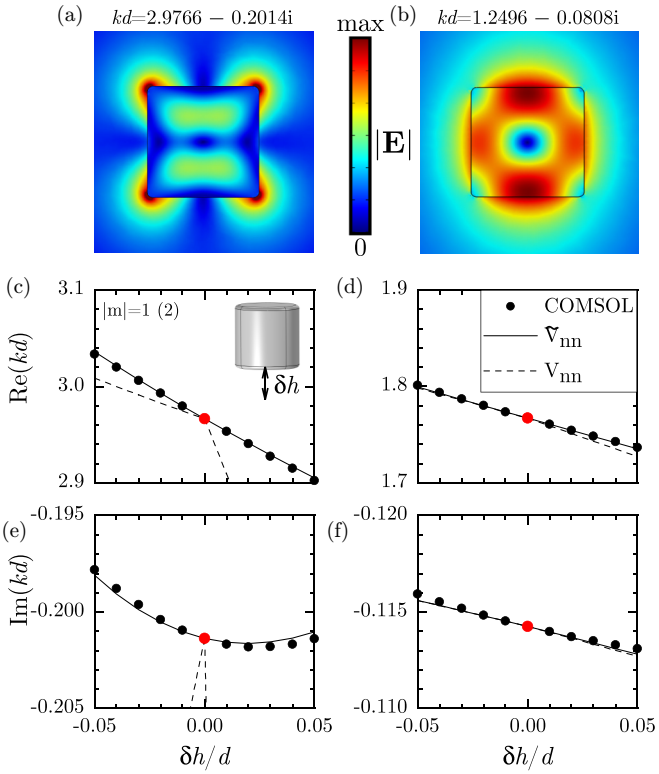

FIG. 4. Left: (a) Field amplitude of a mode of a cylinder with $kd = 2.9766 - 0.2014i$ in a plane containing the cylinder axis. (c) Real and (e) imaginary parts of the mode wave number versus height change δh for first-order perturbations (lines) and COMSOL (dots), with the unperturbed mode highlighted in red and a sketch of the cylinder and its height perturbation in (c). Right: As left but for a mode with $kd = 1.2496 - 0.0808i$. All modes have a magnetic quantum number of $|m| = 1$ and are twice degenerate (2) as noted in (c).

[30], and we introduce a small rounding of radius $d/20$ of the edges to improve numerical stability. We consider two modes with magnetic quantum number $|m| = 1$, shown in Figs. 4(a) and 4(b), and their first-order perturbation by a height change. The mode in panel (a) has a large field component normal to the surface, resulting in a clear discontinuity in the field amplitude. This leads to a large difference between the first-order results $k^{(1)}$ and $\tilde{k}^{(1)}$ [Figs. 4(c) and 4(e)], with only $\tilde{k}^{(1)}$ being consistent with the numerical results. The mode in panel (b) instead has a small field component normal to the surface, as indicated by the smaller discontinuity of the field amplitude, and while $k^{(1)}$ is close to $\tilde{k}^{(1)}$ [Figs. 4(d) and 4(f)], only $\tilde{k}^{(1)}$ is consistent with the numerical results.

V. CONCLUSIONS

We have shown that a first-order perturbation theory of the eigenfrequencies in open systems requires separate considerations for volume perturbations and interface shifts. While volume perturbations lead to first-order diagonal matrix elements capturing the complete first-order effect, moving interfaces, which host discontinuities of the underlying medium properties, lead to additional first-order contributions arising from higher-order terms. In case of electromagnetism, this is due to the coupling to the countably infinite number of degenerate static modes. The underlying mechanism is clarified by explicitly treating the static pole of the Green's dyadic, and a first-order perturbation theory expression valid for both medium changes and interface shifts is provided.

Information on the data created during this research, including how to access it, is available from the Cardiff University data archive Ref. [31].

ACKNOWLEDGMENTS

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APPENDIX A: FIRST-ORDER APPROXIMATIONS BASED ON THE RESONANT-STATE EXPANSION

In this section we provide, based on the RSE, a more general and detailed derivation of the correct first-order change of the RS wave number due to a small perturbation. To do this, we first recap how the RSE is treating perturbations in the permittivity, permeability, and chirality. Then we evaluate the perturbation series for the perturbed wave number from the RSE matrix equation. Based on the closure relation of the modes, we discuss qualitatively how the second-order term can lead to a linear contribution in the deformation depth $h(\boldsymbol{r})$. We then show explicitly, for spherically-symmetric basis systems, the exact conversion from second to first order based on the residue of the Green's function at the static pole. Finally, we extract the exact first-order correction from all higher orders.

1. Resonant-state expansion

A review and derivation of the general RSE method, treating perturbations to arbitrary order, can be found in [26] for nondispersive systems. Here we recap the elements of the RSE required in the following derivations.

The full electromagnetic Green's function $\mathbb{G}$ can be written as a 6×6 dyadic, made up of four 3×3 dyadic elements,

$$\mathbb{G} = \begin{pmatrix} \hat{\mathrm{G}}^{EE} & \hat{\mathrm{G}}^{EH} \\ \hat{\mathrm{G}}^{HE} & \hat{\mathrm{G}}^{HH} \end{pmatrix}. \tag{A1}$$

The full electromagnetic field can be written as a six-element vector

$$\vec{\mathbb{F}} = \begin{pmatrix} \mathbf{E} \\ i\mathbf{H} \end{pmatrix}, \tag{A2}$$

comprising both the electric and magnetic fields. The Green's dyadic inside the system can be written in its spectral form as an infinite sum

$$\begin{aligned} \mathbb{G}(\mathbf{r}, \mathbf{r}') &= \sum_{\nu} \frac{\vec{\mathbb{F}}_\nu(\mathbf{r}) \otimes \vec{\mathbb{F}}_\nu(\mathbf{r}')}{k - k_\nu} \\ &= \sum_{n} \frac{\vec{\mathbb{F}}_n(\mathbf{r}) \otimes \vec{\mathbb{F}}_n(\mathbf{r}')}{k - k_n} + \sum_{\lambda} \frac{\vec{\mathbb{F}}_\lambda(\mathbf{r}) \otimes \vec{\mathbb{F}}_\lambda(\mathbf{r}')}{k}. \end{aligned} \tag{A3}$$

Here n labels the resonant states (RSs), with $k_\nu = k_n \neq 0$, and λ labels the static modes, with $k_\nu = k_\lambda = 0$. The latter are used to represent the static pole of the Green's dyadic. The modes have to be normalized as in [25,26] (differing from the normalization in [6,13] by a factor of $\sqrt{2}$). Using Eq. (A3) allows us to map Maxwell's equations onto the following matrix eigenvalue problem:

$$(\varkappa - k_\nu)c_\nu = -\varkappa \sum_{\nu'} V_{\nu\nu'} c_{\nu'}, \tag{A4}$$

which links the perturbed and the basis systems. Here ν is going over the indices n and λ, $\varkappa$ is the eigenvalue of the perturbed system, c_ν is the expansion coefficient for the perturbed field, and

$$V_{\nu\nu'} = \int \vec{\mathbb{F}}_\nu(\mathbf{r}) \cdot \Delta\mathbb{P}(\mathbf{r}) \vec{\mathbb{F}}_{\nu'}(\mathbf{r}) d\mathbf{r}, \tag{A5}$$

which is a volume integral containing the perturbation. The perturbation is given by

$$\Delta\mathbb{P}(\mathbf{r}) = \begin{pmatrix} \Delta\hat{\varepsilon}(\mathbf{r}) & -i\Delta\hat{\xi}(\mathbf{r}) \\ i\Delta\hat{\zeta}(\mathbf{r}) & \Delta\hat{\mu}(\mathbf{r}) \end{pmatrix}, \tag{A6}$$

where 3×3 tensors $\Delta\hat{\boldsymbol{\varepsilon}}(\mathbf{r})$ and $\Delta\hat{\boldsymbol{\mu}}(\mathbf{r})$ express the change in permittivity and permeability, respectively, while $\Delta\hat{\boldsymbol{\xi}}(\mathbf{r})$ and $\Delta\hat{\boldsymbol{\zeta}}(\mathbf{r})$ describe the change of the bi-anisotropy tensors. For nonmagnetic systems, Eq. (A5) simplifies to $V_{\nu\nu'} = \int \mathbf{E}_\nu(\mathbf{r}) \cdot \Delta\hat{\varepsilon}(\mathbf{r})\mathbf{E}_{\nu'}(\mathbf{r}) d\mathbf{r}$. Perturbed eigenvalues $\varkappa$ are found by truncating the matrix equation (A4) at some finite size, and then diagonalizing it.

2. Perturbation series up to second order

Instead of solving the complete matrix equation (A4), one can extract a perturbation series for $\varkappa$ in terms of powers of the matrix elements $V_{\nu\nu'}$ of the perturbation. This is done below by applying a standard procedure, namely, by taking a perturbed solution in the form

$$\begin{aligned} \varkappa &= \varkappa^{(0)} + \varkappa^{(1)} + \varkappa^{(2)} + \ldots, \\ \vec{\mathbb{F}} &= \sum_\nu c_\nu \vec{\mathbb{F}}_\nu, \\ c_\nu &= c_\nu^{(0)} + c_\nu^{(1)} + c_\nu^{(2)} + \ldots, \end{aligned} \tag{A7}$$

where the superscript indicates the related power of the matrix elements. Here we consider terms only up to second order, simply for the purpose of illustrating how can a second-order term be turned into a first-order correction. Later we extract first-order corrections from all higher-order terms.

Substituting the above expansions into Eq. (A4), we find the first-order corrections to the wave number and the wave function of state n,

$$\begin{aligned} \varkappa^{(1)} &= -k_n V_{nn}, \\ c^{(1)}_{\nu\neq n} &= -\frac{k_n V_{\nu n}}{(k_n - k_\nu)}. \end{aligned} \tag{A8}$$

Using these one can find the second-order correction to the wave number,

$$\varkappa^{(2)} = k_n V_{nn}^2 + k_n^2 \sum_{\nu\neq n} \frac{V_{n\nu} V_{\nu n}}{k_n - k_\nu}, \tag{A9}$$

giving the perturbed wave number up to second order as

$$\varkappa = k_n - k_n V_{nn} + k_n V_{nn}^2 + k_n^2 \sum_{\nu\neq n} \frac{V_{n\nu} V_{\nu n}}{k_n - k_\nu} + \ldots, \tag{A10}$$

where the sum in the last term includes all other RSs ($k_\nu \neq 0$) and all static modes ($k_\nu = 0$). The first three terms in Eq. (A10) can also be understood as a diagonal (i.e., $\nu = n$) approximation to the matrix equation (A4), expanded to second order in V_{nn},

$$\varkappa \approx \frac{k_n}{1 + V_{nn}} = k_n - k_n V_{nn} + k_n V_{nn}^2 + \ldots \tag{A11}$$

However, the diagonal approximation Eq. (A11) does not contain the full first-order information. In fact, the last term in Eq. (A10) can contain implicit first-order corrections arising from the static-modes as we will show in the following subsection.

3. Converting second-order terms to first order based on closure relation

Let us consider only the static-mode contribution to the sum in Eq. (A10), by setting $\nu = \lambda$, and $k_\nu = 0$. The last term then reduces to

$$\begin{aligned} \sum_\lambda V_{n\lambda} V_{\lambda n} = \sum_\lambda \iint \vec{\mathbb{F}}_n(\mathbf{r}) \cdot \Delta\mathbb{P}(\mathbf{r}) \vec{\mathbb{F}}_\lambda(\mathbf{r}) \\ \otimes \vec{\mathbb{F}}_\lambda(\mathbf{r}') \Delta\mathbb{P}(\mathbf{r}') \vec{\mathbb{F}}_n(\mathbf{r}') d\mathbf{r} d\mathbf{r}', \end{aligned} \tag{A12}$$

leaving out the factor k_n. Using the closure relation [26]

$$\mathbb{P}(\mathbf{r}) \sum_\nu \vec{\mathbb{F}}_\nu(\mathbf{r}) \otimes \vec{\mathbb{F}}_\nu(\mathbf{r}') = \mathbb{I}\delta(\mathbf{r} - \mathbf{r}'), \tag{A13}$$

where $\mathbb{I}$ is the 6×6 identity matrix, one can express the sum in Eq. (A12) as

$$\sum_\lambda \vec{\mathbb{F}}_\lambda(\mathbf{r}) \otimes \vec{\mathbb{F}}_\lambda(\mathbf{r}') = \mathbb{P}^{-1}(\mathbf{r})\delta(\mathbf{r} - \mathbf{r}') - \sum_n \vec{\mathbb{F}}_n(\mathbf{r}) \otimes \vec{\mathbb{F}}_n(\mathbf{r}'). \tag{A14}$$

Inserting this into Eq. (A12) and integrating out the delta function gives

$$\begin{aligned} \sum_\lambda V_{n\lambda} V_{\lambda n} = \int \vec{\mathbb{F}}_n(\mathbf{r}) \cdot \Delta\mathbb{P}(\mathbf{r}) \mathbb{P}^{-1}(\mathbf{r}) \Delta\mathbb{P}(\mathbf{r}) \vec{\mathbb{F}}_n(\mathbf{r}) d\mathbf{r} \\ - \sum_{n'} V_{nn'} V_{n'n}. \end{aligned} \tag{A15}$$

The first term is clearly second order in $\Delta\mathbb{P}(\mathbf{r})$. However, $\Delta\mathbb{P}(\mathbf{r})$ may not be small, and if we consider a perturbation that is a thin layer of thickness $h(\boldsymbol{r})$ and approximate the volume integral with a surface integral multiplied with $h(\boldsymbol{r})$ [this is equivalent to taking the zeroth-order Taylor expansion of the field $\vec{\mathbb{F}}_n(\mathbf{r})$ normal to the surface], then it is clear that the term is first order in the layer thickness h.

The elimination of the static mode contribution via the closure relation has been already explored in [26], in the context of the full matrix equation Eq. (A4)—this is referred to as ML2 in [26] and corresponds to one of the Mittag-Leffler representations of the dyadic Green's function (GF). However, as shown in [26] this leads to a slow convergence of the RSE,

as it turns out that the term $\sum_n \vec{\mathbb{F}}_n(\mathbf{r}) \otimes \vec{\mathbb{F}}_n(\mathbf{r}')$ can also contain a singularity. In the next subsection we will see how the result changes if the full singularity of the static-pole residue of the dyadic GF is taken into account.

4. Converting second-order terms to first order based on the static pole of the Green's dyadic

So far we have not used any symmetry of the basis system. Let us now consider a spherically-symmetric basis system, with isotropic radially dependent permittivity $\hat{\boldsymbol{\varepsilon}}(\mathbf{r}) = \hat{\mathbf{1}}\varepsilon(r)$ and permeability $\hat{\boldsymbol{\mu}}(\mathbf{r}) = \hat{\mathbf{1}}\mu(r)$, where $\hat{\mathbf{1}}$ is the 3×3 identity matrix. As derived in [26], the static pole residue of the dyadic GF of such a system has a δ-like singularity. Following [26], one can write the full dyadic GF in the basis of vector spherical harmonics (VSHs) as

$$\mathbb{G}(\mathbf{r}, \mathbf{r}') = \sum_{ij} \sum_{lm} [\hat{G}_{ij}(r, r')]_{l,m} \mathbf{Y}_{ilm}(\Omega) \otimes \mathbf{Y}_{jlm}(\Omega), \quad \text{(A16)}$$

where $\mathbf{Y}_{ilm}(\Omega)$ are the VSHs with $i = 1, 2, 3$, and l and m being, respectively, the orbital and magnetic quantum numbers. Here, $\hat{G}_{ij}$ are 2×2 matrices, each consisting of four elements: G_{ij}^{EE}, G_{ij}^{EH}, G_{ij}^{HE}, and G_{ij}^{HH} [compare with Eq. (A1)], the first and the last element being, respectively, the electric and magnetic part of the Green's dyadic. These two were shown [26] to have δ-like singularities, and their singular parts are given by

$$G_{33,\mathrm{sing}}^{EE}(r, r') = \frac{\delta(r - r')}{r^2 k \varepsilon(r)},$$
$$G_{33,\mathrm{sing}}^{HH}(r, r') = \frac{\delta(r - r')}{r^2 k \mu(r)}. \quad \text{(A17)}$$

Using the definition $\mathbf{Y}_{3lm}(\Omega) = \hat{\mathbf{r}} Y_{lm}(\Omega)$, where $Y_{lm}(\Omega)$ is a spherical harmonic and $\hat{\mathbf{r}}$ is a unit vector in the radial direction, the closure relation for spherical harmonics

$$\sum_{lm} Y_{lm}(\Omega) Y_{lm}(\Omega') = \frac{1}{\sin\theta} \delta(\theta - \theta') \delta(\phi - \phi'), \quad \text{(A18)}$$

and the spherical-coordinate representation of $\delta(\mathbf{r} - \mathbf{r}')$, the static-pole singularity of the Green's dyadic of a spherically-symmetric system takes the form

$$\mathbb{G}_{\mathrm{sing}}(\mathbf{r}, \mathbf{r}') = \begin{pmatrix} \hat{\mathrm{G}}_{\mathrm{sing}}^{EE}(\mathbf{r}, \mathbf{r}') & 0 \\ 0 & \hat{\mathrm{G}}_{\mathrm{sing}}^{HH}(\mathbf{r}, \mathbf{r}') \end{pmatrix}$$
$$= \frac{\delta(\mathbf{r} - \mathbf{r}')}{k} \mathbb{I}_{\hat{\boldsymbol{r}} \otimes \hat{\boldsymbol{r}}} \mathbb{P}^{-1}(\mathbf{r}), \quad \text{(A19)}$$

where

$$\mathbb{I}_{\hat{\boldsymbol{r}} \otimes \hat{\boldsymbol{r}}} = \begin{pmatrix} \hat{\mathbf{r}} \otimes \hat{\mathbf{r}} & 0 \\ 0 & \hat{\mathbf{r}} \otimes \hat{\mathbf{r}} \end{pmatrix} \quad \text{(A20)}$$

and

$$\mathbb{P}(\mathbf{r}) = \begin{pmatrix} \hat{\varepsilon}(\mathbf{r}) & 0 \\ 0 & \hat{\mu}(\mathbf{r}) \end{pmatrix} \quad \text{(A21)}$$

are 6×6 diagonal tensors.

Equation (A19) allows us to write the static-pole residue of the dyadic GF [see Eq. (A3)] in the form

$$\sum_{\lambda} \vec{\mathbb{F}}_\lambda(\mathbf{r}) \otimes \vec{\mathbb{F}}_\lambda(\mathbf{r}') = \mathbb{I}_{\hat{\boldsymbol{r}} \otimes \hat{\boldsymbol{r}}} \mathbb{P}^{-1}(\mathbf{r}) \delta(\mathbf{r} - \mathbf{r}') + \mathbb{R}(\mathbf{r}, \mathbf{r}'), \quad \text{(A22)}$$

where $\mathbb{R}(\mathbf{r}, \mathbf{r}')$ is a regular part of the Green's dyadic. Using Eq. (A22) in Eq. (A12) and integrating the term containing the δ function leads to

$$\sum_{\lambda} V_{n\lambda} V_{\lambda n} = \int \vec{\mathbb{F}}_n(\mathbf{r}) \cdot \Delta\mathbb{P}(\mathbf{r}) \mathbb{I}_{\hat{\boldsymbol{r}} \otimes \hat{\boldsymbol{r}}} \mathbb{P}^{-1}(\mathbf{r}) \Delta\mathbb{P}(\mathbf{r}) \vec{\mathbb{F}}_n(\mathbf{r}) d\mathbf{r}$$
$$+ \sum_n \iint \vec{\mathbb{F}}_n(\mathbf{r}) \cdot \Delta\mathbb{P}(\mathbf{r}) \mathbb{R}(\mathbf{r}, \mathbf{r}')$$
$$\times \Delta\mathbb{P}(\mathbf{r}') \vec{\mathbb{F}}_n(\mathbf{r}') d\mathbf{r} d\mathbf{r}'. \quad \text{(A23)}$$

The first term on the right-hand side of Eq. (A23) is a single-volume integral and is thus of first order in h, which needs to be included in the first-order perturbation theory result. The second term in Eq. (A23) contains a double-volume integral, not reducible to a single-volume integral, and is thus of second order, both in $\Delta\mathbb{P}$ and h. We note that $\mathbb{R}(\mathbf{r}, \mathbf{r}')$ may also show a singularity when the summation over all quantum numbers l and m is taken, similar to Eq. (A18); however, this does not influence the first-order results as modes with different l do not mix in first order.

5. Extracting the first-order correction from all higher-order terms

Based on the results for second order we can expect that even higher-order terms can have a contribution linear in h. Instead of attempting to derive corrections in the infinite perturbation series Eq. (A10) to arbitrary order and then extracting first-order corrections from them, we take the RSE matrix equation (A4) in a form, which does not explicitly depend on static modes [23,26,32],

$$(\varkappa - k_n) c_n = -\varkappa \sum_{n'} \tilde{V}_{nn'} c_{n'}, \quad \text{(A24)}$$

where

$$\tilde{V}_{nn'} = V_{nn'} - S_{nn'} \quad \text{(A25)}$$

and

$$S_{nn'} = \sum_{\lambda\lambda'} V_{n\lambda} W_{\lambda\lambda'} V_{\lambda' n'}, \quad \text{(A26)}$$

with W being the inverse of the matrix $\delta_{\lambda\lambda'} + V_{\lambda\lambda'}$. Note that regarding the RSs, Eq. (A24) is equivalent to Eq. (A4). We can express the matrix W as a Neumann series [33],

$$W = (I + V)^{-1} = I - V + V^2 - V^3 + V^4 - \ldots, \quad \text{(A27)}$$

where I is the identity matrix and V has matrix elements $V_{\lambda\lambda'}$, which are overlap integrals between static modes only. Inserting the expansion Eq. (A27) into Eq. (A26), we obtain for $n' = n$ the series

$$S_{nn} = \sum_{\lambda} V_{n\lambda} V_{\lambda n} - \sum_{\lambda\lambda_1} V_{n\lambda} V_{\lambda\lambda_1} V_{\lambda_1 n}$$
$$+ \sum_{\lambda\lambda_1\lambda_2} V_{n\lambda} V_{\lambda\lambda_1} V_{\lambda_1\lambda_2} V_{\lambda_2 n} - \ldots, \quad \text{(A28)}$$

which can also be understood as a part of the full perturbation series Eq. (4) relevant to the first-order correction we are looking for.

Now, using Eq. (A22) and neglecting any terms containing the regular part of the static-pole residue, we find approximately

$$S_{nn} \approx \int \vec{\mathbb{F}}_n \cdot [\Delta\mathbb{P}\mathbb{I}_{\hat{\mathbf{r}}\otimes\hat{\mathbf{r}}}\mathbb{P}^{-1} - (\Delta\mathbb{P}\mathbb{I}_{\hat{\mathbf{r}}\otimes\hat{\mathbf{r}}}\mathbb{P}^{-1})^2 + (\Delta\mathbb{P}\mathbb{I}_{\hat{\mathbf{r}}\otimes\hat{\mathbf{r}}}\mathbb{P}^{-1})^3 - \ldots]\Delta\mathbb{P}\vec{\mathbb{F}}_n d\mathbf{r}\,. \tag{A29}$$

The above infinite series in the square brackets can be summed up, again using the Neumann series Eq. (A27), which results in

$$S_{nn} \approx \int \vec{\mathbb{F}}_n \cdot [\mathbb{I} - (\mathbb{I} + \Delta\mathbb{P}\mathbb{I}_{\hat{\mathbf{r}}\otimes\hat{\mathbf{r}}}\mathbb{P}^{-1})^{-1}]\Delta\mathbb{P}\vec{\mathbb{F}}_n d\mathbf{r}, \tag{A30}$$

so that finally

$$\tilde{V}_{nn} = V_{nn} - S_{nn} \approx \int \vec{\mathbb{F}}_n \cdot (\mathbb{I} + \Delta\mathbb{P}\mathbb{I}_{\hat{\mathbf{r}}\otimes\hat{\mathbf{r}}}\mathbb{P}^{-1})^{-1}\Delta\mathbb{P}\vec{\mathbb{F}}_n d\mathbf{r}. \tag{A31}$$

Equation (A31) is a generalization of Eq. (11) of the main text, and is one of the main results of this work. It provides the exact first-order correction to the wave number via

$$\varkappa = k_n - k_n\tilde{V}_{nn}\,. \tag{A32}$$

The above result is valid for spherically-symmetric systems described by the generalized permittivity $\mathbb{P}(r)$, which are subject to an arbitrary perturbation $\Delta\mathbb{P}(\mathbf{r})$ without assumptions regarding its symmetry. Equation (A32) can also be seen as a first-order approximation to the diagonal version of Eq. (A24), similar to Eq. (A11) but using the modified matrix elements Eq. (A25). When $\Delta\mathbb{P}$ is small, for example in case of a small homogeneous perturbation across the whole sphere, the term $\Delta\mathbb{P}\mathbb{I}_{\hat{\mathbf{r}}\otimes\hat{\mathbf{r}}}\mathbb{P}^{-1}$ in Eq. (A31) provides a second-order contribution due to the static modes, and the original first-order approximation Eq. (A8) to the matrix equation Eq. (A4) is recovered. However, when $\Delta\mathbb{P}$ is not small, as in the case of a shape deformation of an interface hosting a discontinuity of $\mathbb{P}$, the term $\Delta\mathbb{P}\mathbb{I}_{\hat{\mathbf{r}}\otimes\hat{\mathbf{r}}}\mathbb{P}^{-1}$ provides a first-order contribution coming from the static modes, in this way modifying the first-order perturbation theory result.

In order to bring the above result to a more familiar form, let us consider unperturbed and perturbed nonmagnetic systems with isotropic permittivity. Equation (A31) then simplifies to

$$\tilde{V}_{nn} \approx \int \mathbf{E}_n(\mathbf{r}) \cdot \Delta\hat{P}(\mathbf{r})\mathbf{E}_n(\mathbf{r})d\mathbf{r}, \tag{A33}$$

where

$$\Delta\hat{P}(\mathbf{r}) = \begin{pmatrix} \Delta\varepsilon(\mathbf{r}) & 0 & 0 \\ 0 & \Delta\varepsilon(\mathbf{r}) & 0 \\ 0 & 0 & \frac{\varepsilon(r)\Delta\varepsilon(\mathbf{r})}{\varepsilon(r)+\Delta\varepsilon(\mathbf{r})} \end{pmatrix} \tag{A34}$$

with the bottom-right corner representing the $\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}$ component of the tensor. In terms of the electric field components parallel and normal to the surface of the basis system, Eq. (A33) can be written as

$$\tilde{V}_{nn} \approx \int \Big[\mathbf{E}_n^{\parallel}(\mathbf{r}) \cdot \Delta\varepsilon(\mathbf{r})\mathbf{E}_n^{\parallel}(\mathbf{r}) + \mathbf{E}_n^{\perp}(\mathbf{r}) \cdot \frac{\varepsilon(r)\Delta\varepsilon(\mathbf{r})}{\varepsilon(r)+\Delta\varepsilon(\mathbf{r})}\mathbf{E}_n^{\perp}(\mathbf{r}) \Big] d\mathbf{r}\,. \tag{A35}$$

This is consistent with the results in [26], where the same perturbation matrix was obtained for the general RSE from a modified spectral representation of the Green's dyadic, in which the singular part of the static pole was kept as a δ-like term, not expanded as a series. This has led, in particular, to a significantly faster convergence of the RSE. Interestingly, in [34], the same perturbation matrix as in Eq. (A34) was obtained, specifically for a shape perturbation. It was derived by using both the inside and outside fields in the overlap integral, and by taking the zeroth order of the Taylor expansion of the field, i.e., its value at the surface. A similar method was used later on in [7], again for shape perturbations. The approach of [7,34], however, masks the contribution of the static modes to the first-order correction and is limited to shape deformations only. Here we reproduce their results by fully considering the static pole of the Green's dyadic. In [35], which builds on the results of [12], the perturbation term $\frac{\varepsilon\Delta\varepsilon}{\varepsilon+\Delta\varepsilon}$ was also used, but for both components of the TM field. For small $\Delta\varepsilon$ this gives the correct results as it was shown in the paper; however, we expect that this fails for larger $\Delta\varepsilon$, which is the case of a shape perturbation. In [36], TE polarization was considered for a photonic-crystal slab at non-normal incidence, and accurate results could be obtained without using static modes for RSs of $ka \approx 0.2$, where $a = 100$ nm was the characteristic length of the structure (of the order of the thickness of the grating and its period). To the best of our knowledge, the role of static modes has not been yet investigated in photonic-crystal structures.

APPENDIX B: EXAMPLE: DIELECTRIC SPHERE IN VACUUM

In this section, we provide a few illustrations of the first-order results derived in Appendix A and their comparison with the exact solutions for a dielectric sphere in vacuum, perturbing either its refractive index (volume perturbation) or its size (boundary perturbation). The general formula correctly describing the wave number of a perturbed RS to first order is given by Eq. (A32),

$$\varkappa = k_n - k_n\tilde{V}_{nn} = k_n - k_n(V_{nn} - S_{nn}), \tag{B1}$$

here written also in terms of V_{nn} and S_{nn}, given by Eqs. (A5) and (A30), respectively. This highlights the importance of the static-mode contribution S_{nn}, which will be evaluated for the examples treated below, along with the diagonal matrix element V_{nn}. As basis system we choose in all examples a dielectric sphere of radius R and permittivity $\varepsilon = 4$, surrounded by vacuum.

1. Material perturbation

We apply the first-order formula Eq. (B1) to the homogeneous dielectric sphere in vacuum, perturbed uniformly, so that the permittivity of the sphere changes by $\Delta\varepsilon$. A particular

change of the permittivity profile is illustrated in Fig. 5(b). Here we focus on perturbation of the TM RSs with $l = 1$. For this value of the angular momentum, the spectrum of the basis system in the complex k plane, shown in Fig. 5(a), consists of only Fabry-Pérot modes, which for large $|k|$ are spaced with a period of $\pi/2\sqrt{\varepsilon}R$ in $\mathrm{Re}(k)$, and have the imaginary part converging to $\mathrm{Im}(k) = -\ln[(\sqrt{\varepsilon}-1)/(\sqrt{\varepsilon}+1)]/(2\sqrt{\varepsilon}R)$, similar to the modes of a homogeneous slab at normal incidence [6,27].

Applying a perturbation to the fundamental mode, shown in Fig. 5(c) up to $\Delta\varepsilon = 0.04$, reveals excellent agreement between the first-order result Eq. (B1) and the exact solution. For all the RSs shown in Fig. 5(a), we provide in (d) and (e) more details for $\Delta\varepsilon = 0.004$ and $\Delta\varepsilon = 0.04$, corresponding to, respectively, 0.1% and 1% change of the permittivity. V_{nn} and S_{nn} are given in Fig. 5(d). For the applied perturbations, $\Delta\varepsilon \ll 1$. Therefore, V_{nn}, which is of first order in $\Delta\varepsilon$ dominates, whereas S_{nn}, which is of second order in $\Delta\varepsilon$ is negligible. Accordingly, V_{nn} scales linearly and S_{nn} scales quadratically with $\Delta\varepsilon$. This is consistent with examples in the literature, where for small $\Delta\varepsilon$ static mode contributions were not included in the RSE [37]. The magnitude of S_{nn} is reducing with increasing $|k|$. This is intuitively expected as we move away from the influence of the $k = 0$ pole in the k plane. Figure 5(e) shows the relative error with respect to the exact solution of the first-order Eq. (B1), with or without the static-mode contribution, which is given by S_{nn}. One can see that the error scales quadratically in $\Delta\varepsilon$ for both, confirming that the first-order correction to k is accurate for all the RSs. The relative errors calculated with or without static modes are similar, confirming that the static pole of the Green's dyadic has a weak contribution when $\Delta\varepsilon$ is small.

2. Size perturbation

We now apply to the same homogeneous dielectric sphere as in the previous section a perturbation that changes its size by h. This corresponds to a perturbation $\Delta\varepsilon = 1 - \varepsilon$, applied to a layer of thickness $|h|$ on the inner side of the sphere surface. The permittivity change is illustrated in Fig. 6(a), which corresponds to the shift of an interface hosting a discontinuity of ε. We apply a perturbation of $h/R = -0.01$ and $h/R = -0.001$, corresponding to, respectively, 1% and 0.1% change of the initial size, with the minus sign noting a decrease of the sphere radius. One can see from Fig. 6(b) that in the complex k plane, the first-order result for the fundamental mode, calculated via Eq. (B1), is in good agreement with the exact solution.

Figure 6(c) shows the values of V_{nn} and S_{nn} for a large number of the RSs. For small k both V_{nn} and S_{nn} are of similar magnitude, and for all k both change linearly with h. This confirms that both terms contribute in first order under size perturbation. The magnitude of S_{nn} decreases for large k as discussed above. We can see that in this region $|V_{nn}|$ fluctuates with k, which we would not expect if the first-order approximation was correct. To understand this, we recall that the secular equation determining the modes use the dimensionless quantity kR, so

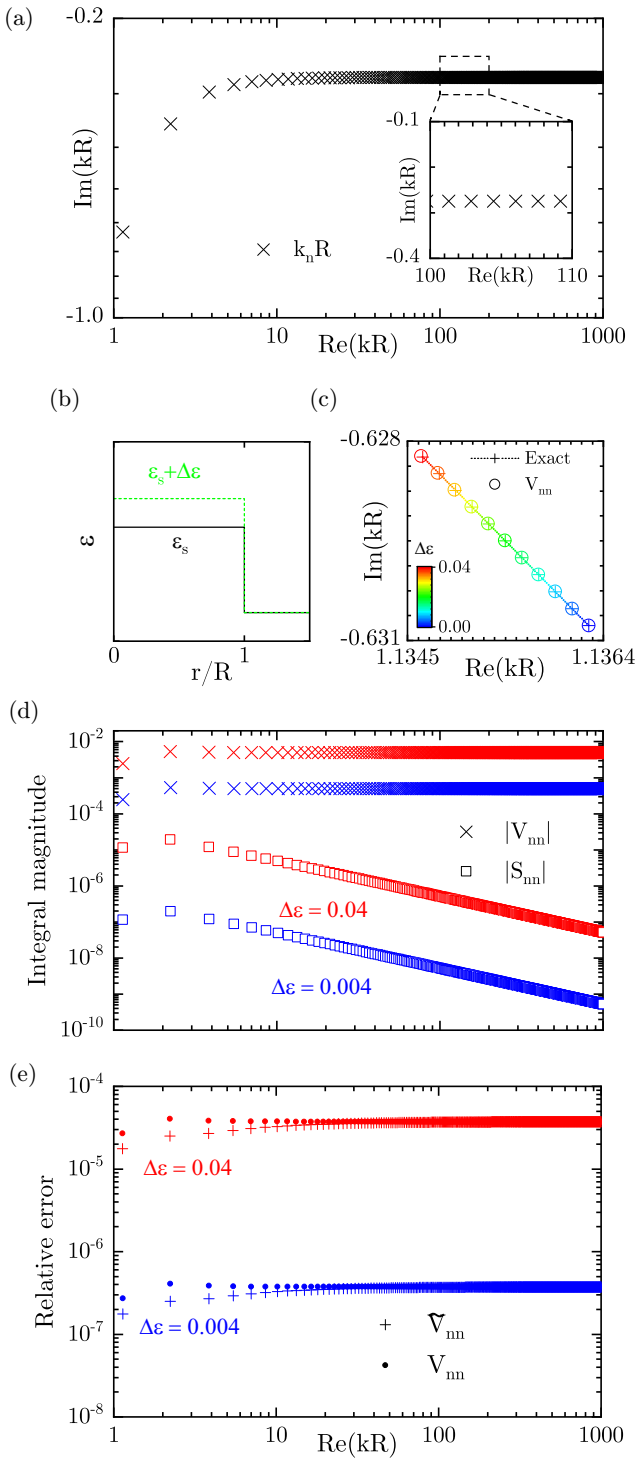

FIG. 5. (a) Wave numbers of TM modes with angular momentum $l = 1$ of a dielectric sphere with permittivity $\varepsilon = 4$ and radius R, surrounded by vacuum. (b) Illustration of the homogeneous permittivity perturbation across the sphere. (c) Perturbed wave number of the fundamental RS with the permittivity change given by the color code. (d) Magnitude of the matrix elements $|V_{nn}|$ and $|S_{nn}|$ of the perturbation. (e) Relative error of the perturbed RS wave numbers calculated with (crosses) and without (dots) inclusion of static modes.

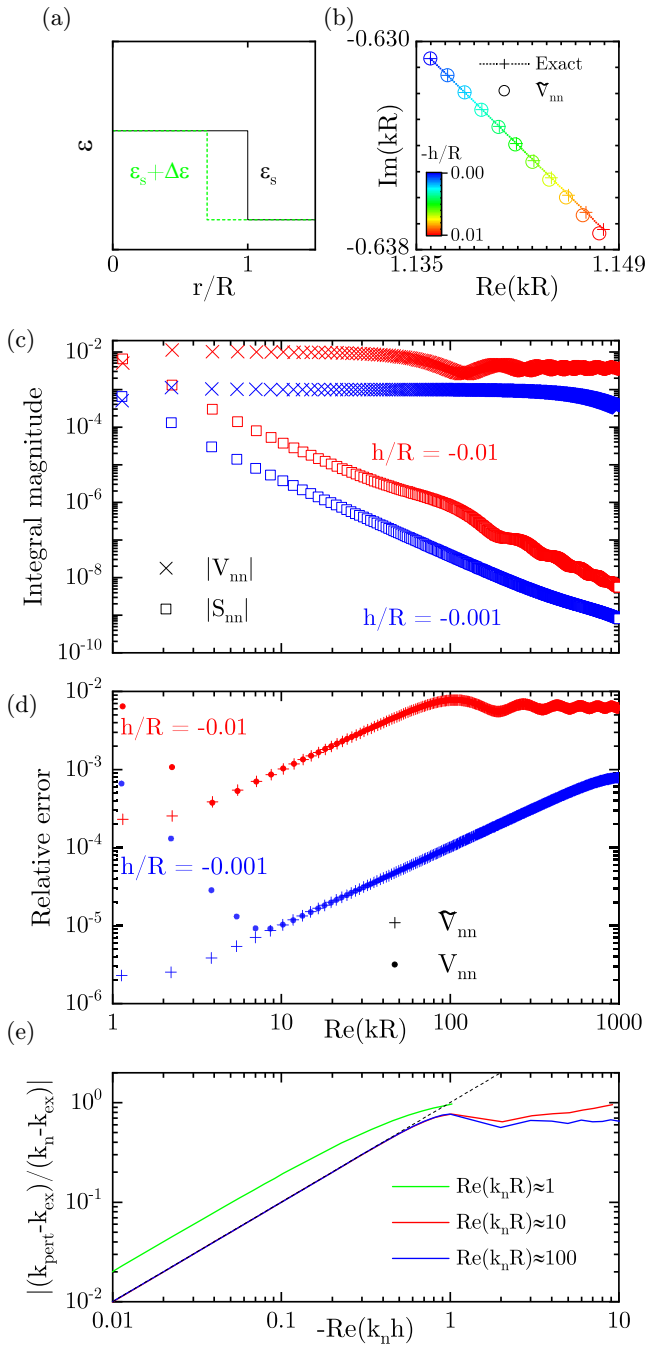

FIG. 6. (a) Illustration of the permittivity perturbation corresponding to a radius reduction of the sphere by $-h$. (b) Perturbed wave number of the fundamental RS with the sphere radius change given by the color code. (c) Magnitude of the matrix elements $|V_{nn}|$ and $|S_{nn}|$ of the perturbation. (d) Relative error of the perturbed RS wave numbers calculated with (crosses) and without (dots) inclusion of static modes. (e) Absolute value of the error of the perturbed RS wave numbers relative to the wave number change due to the perturbation, as functions of $\mathrm{Re}(k_nh)$, for three selected modes, and k_{ex} denote the exact perturbed wavenumber. The dashed line indicates a linear scale.

that k and R are inversely proportional to each other for a given mode under size perturbation. Therefore, the perturbed wave number can be claculated as $\varkappa = k_nR/(R+h) \approx k_n/(1+\tilde{V}_{nn})$, or alternatively, $\varkappa \approx k_n(1-\tilde{V}_{nn}) \approx k_n(1-h/R)$, implying that $\tilde{V}_{nn}$ should be constant with respect to k_n that is, it should be state-independent, in first order.

One can see from Fig. 6(d) that the error for RSs of low $|k|$ can increase by several orders of magnitude when the static modes are not included, exemplifying their importance. The relative error scales quadratically with h for $|kh| \ll 1$. The error increases with k, and at $|kh| > 1$ (i.e. at $\mathrm{Re}(kR) = 1000$ in (d)) we can observe that the error only reduces proportionally to h. This is further exemplified in Fig. 6(e) by showing the absolute value of the error relative to the change of the wave number. The former should change quadratically, while the latter linearly, therefore the ratio should scale linearly if the first order correction is dominant. However, in practice it is only scaling linearly when $|kh| < 1$. This suggests that the first-order approximation requires $|kh| \ll 1$. This, together with the oscillations in the magnitude of the matrix elements, also suggests that for size perturbations, contributions from other neighboring RSs are important for Fabry-Pérot modes when $|kh| \gtrsim 1$, meaning that higher-order corrections need to be taken into account in the perturbation series representation Eq. (A10), or alternatively, in the context of the full matrix equation Eq. (A4), the off-diagonal elements cannot be neglected.

Moving to an angular momentum number $l = 50$, the results of the size perturbation are shown in Fig. 7. For larger values of l, high-quality whispering-gallery modes form in spectra, and the fundamental mode position shifts to higher k. Also, a sharp peak associated with the Brewster angle is formed in the spectrum of the RSs [27].

We find that for this high l the role of the static pole is lessened, but still relevant, reducing the error by up to an order of magnitude for the whispering-gallery modes. The contribution from both matrix elements scales about the same way as for the $l = 1$ case, i.e., linearly in h, but here the magnitude of the static-mode contribution is significantly smaller than the RS contribution. This is related to the large detuning $|kR| \gg 1$ from the static pole. Considering the field contribution to the overlap integrals, for low values of the angular momentum the tangential and radial component of the field are similar in magnitude, whereas for high l the radial part, which contributes to the static-pole integral, is much weaker.

Considering the relative error, we observe the same features as for $l = 1$, namely, small error for the modes with $|kh| \ll 1$, which then grows up to the point $|kh| \approx 1$. If we compare relative errors with and without the static pole included, we can see that with their correct inclusion we still gain an order of magnitude accuracy for the $h/R = -0.001$ case, and about a factor of 2 for $h/R = -0.01$. In Fig. 7(d) we can see that for larger perturbations, the fundamental whispering-gallery mode shows higher errors relative to the change of the wave number when compared to Fabry-Pérot modes. This is attributed to the localization of the whispering-gallery mode close to the surface of the sphere.

APPENDIX C: SPHERICALLY-SYMMETRIC DIELECTRIC SYSTEMS

In this section, we focus on nondispersive dielectric spherically-symmetric systems, in order to provide explicit and easy-to-follow derivations of the first-order correction to

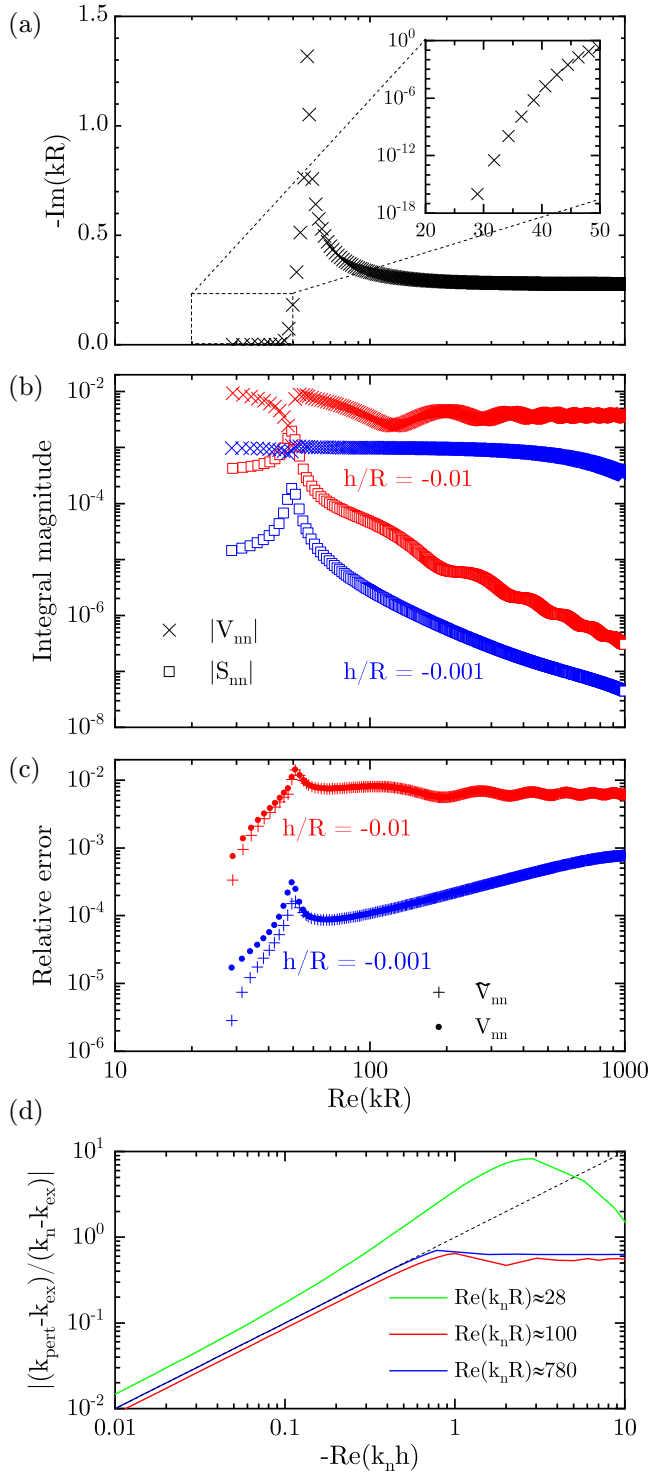

FIG. 7. (a) Wave numbers of TM modes with angular momentum $l = 50$ of a dielectric sphere with radius R and permittivity $\varepsilon = 4$, surrounded by vacuum. Inset shows a zoom of the high-quality whispering-gallery modes in a logarithmic scale. (b) Magnitude of the matrix elements $|V_{nn}|$ and $|S_{nn}|$ of the perturbation. (c) Relative error of the perturbed RS wave numbers calculated with (crosses) and without (dots) inclusion of static modes. (d) Absolute error of the perturbed RS wave numbers relative to the wave number change due to the perturbation, as functions of $\mathrm{Re}(k_n h)$, for three selected modes. Green line is the fundamental whispering-gallery mode.

the RS wave numbers. We start with the RSs in TE polarization where the correct first-order result follows directly from the diagonal approximation within the standard RSE approach. We also provide an alternative derivation of the first-order result, without using the RSE. We then focus on TM polarization where different versions of the RSE lead to different results for the first-order correction to the RS wave number and discuss the source of their inconsistency.

1. TE polarization

In TE polarization, Maxwell's wave equation for the RSs is reduced to a scalar wave equation for the electric field [26,27],

$$\left[\frac{d^2}{dr^2} - \frac{\alpha^2}{r^2} + \varepsilon(r)k_n^2\right]E_n(r) = 0, \tag{C1}$$

where r is the radial coordinate, $\varepsilon(r)$ is the permittivity of the unperturbed spherically-symmetric system, $\alpha^2 = l(l+1)$, l is the orbital quantum number, k_n is the RS wave number, and $E_n(r)/r$ is the radially-dependent part of the RS electric field.

A perturbed RS in TE polarization, with the wave number k and electric field $E(r)/r$, satisfies the perturbed wave equation

$$\left[\frac{d^2}{dr^2} - \frac{\alpha^2}{r^2} + \varepsilon(r)k^2 + \Delta\varepsilon(r)k^2\right]E(r) = 0, \tag{C2}$$

where $\Delta\varepsilon(r)$ is a perturbation of the permittivity.

a. RSE-based approach

Using the solution of the unperturbed problem, Eq. (C2) can be solved via a Lippmann-Schwinger equation

$$E(r) = -k\int_0^\infty G_k(r,r')\Delta\varepsilon(r')E(r')dr' \tag{C3}$$

where $G_k(r,r')$ is the unperturbed GF satisfying the equation

$$\left[\frac{d^2}{dr^2} - \frac{\alpha^2}{r^2} + \varepsilon(r)k^2\right]G_k(r,r') = k\delta(r-r'). \tag{C4}$$

The GF has the following Mittag-Leffler expansions in terms of the RSs only [26]:

$$G_k(r,r') = \sum_n \frac{E_n(r)E_n(r')}{k-k_n} = \sum_n \frac{k}{k_n}\frac{E_n(r)E_n(r')}{k-k_n}, \tag{C5}$$

provided that the RS fields are normalized [6,26] according to

$$2\int_0^R \varepsilon(r)E_n^2(r)dr + \frac{1}{k_n^2}\left[E_n(r)E_n'(r) + rE_n(r)E_n''(r) - rE_n'^2(r)\right]_{R_+} = 1, \tag{C6}$$

where R is the radius of the system (including all the inhomogeneities of the permittivity) and $R_+ = R + 0_+$ with 0_+ being a positive infinitesimal.

Since the Mittag-Leffler expansions Eq. (C5) are valid only within the unperturbed system volume (i.e., for $r \leqslant R$), in order to be able to use them for the RSE, we require that

$\Delta\varepsilon(r) = 0$ outside the basis system. Substituting the first series from Eq. (C5) into Eq. (C3), we obtain

$$E(r) = -k \sum_n \frac{E_n(r)}{k - k_n} \int_0^R E_n(r')\Delta\varepsilon(r')E(r')dr' \quad \text{(C7)}$$

$$= \sum_n c_n E_n(r), \quad \text{(C8)}$$

introducing in the second line the expansion coefficients

$$c_n = -\frac{k}{k - k_n} \sum_n \int_0^R E_n(r)\Delta\varepsilon(r)E(r)dr\,. \quad \text{(C9)}$$

Finally, substituting the expansion Eq. (C8) into the integral in Eq. (C9) results in the RSE matrix equation,

$$(k - k_n)c_n = -k \sum_m c_m \int_0^R E_n(r)\Delta\varepsilon(r)E_m(r)dr, \quad \text{(C10)}$$

which is linear in the perturbed RS wave number k. Note that using instead the second series from Eq. (C5) would lead to a nonlinear (quadratic) eigenvalue problem.

It is now straightforward to obtain from Eq. (C10) the first-order approximation for the RS wave number. To do this, we first keep only the diagonal terms $n = m$ in the matrix equation and then neglect any higher-order corrections,

$$k - k_n \approx -k_n \int_0^R \Delta\varepsilon(r)E_n^2(r)dr\,. \quad \text{(C11)}$$

b. Direct evaluation of the first-order correction to the wave number

The same first-order result Eq. (C11) can be obtained in a more straightforward manner directly from the wave equations, without introducing the GF and any expansions. Multiplying Eq. (C1) with $E(r)$ and Eq. (C2) with $E_n(r)$, taking their difference and integrating the result between 0 and R_+, we find

$$\int_0^{R_+} (EE_n'' - E_nE'')dr + \int_0^R \left(\varepsilon k_n^2 - \varepsilon k^2 - \Delta\varepsilon k^2\right)EE_n dr = 0, \quad \text{(C12)}$$

omitting the dependence on r everywhere for brevity. Integrating by parts and using the fact that the fields vanish at the origin, $E(0) = E_n(0) = 0$, transforms the first integral in Eq. (C12) to

$$[EE_n' - E_nE']_{R_+}, \quad \text{(C13)}$$

which can be further evaluated by using the outgoing wave boundary conditions for the RSs and the analytic form of the solution outside the system,

$$\begin{aligned} E_n(r) &= C_n H(k_n r), \\ E(r) &= C(k)H(kr), \end{aligned} \quad \text{(C14)}$$

where C_n and $C(k)$ are some constants, and $H(z) = zh_l^{(1)}(z)$, with $h_l^{(1)}(z)$ being the spherical Hankel function of first kind. The expression Eq. (C13) can then be written as

$$\begin{aligned} [EE_n' - E_nE']_{R_+} = C_nC(k)[&H(kR)H'(k_nR)k_n \\ &- H(k_nR)H'(kR)k] \end{aligned} \quad \text{(C15)}$$

and can be further evaluated to first order in $k - k_n$ by using the Taylor expansion

$$H(z) \approx H(z_0) + H'(z)(z - z_0), \quad \text{(C16)}$$

$$H'(z) \approx H'(z_0) + H''(z)(z - z_0)\,. \quad \text{(C17)}$$

Then we find

$$\begin{aligned} [EE_n' - E_nE']_{R_+} &\approx C_nC(k)[\{H(k_nR) + H'(k_nR)(k - k_n)R\}H'(k_nR)k_n - H(k_nR)\{H'(k_nR) + H''(k_nR)(k - k_n)R\}k] \\ &= C_nC(k)(k - k_n)[-H(k_nR)H'(k_nR) + k_nR\{H'(k_nR)\}^2 - kRH(k_nR)H''(k_nR)] \\ &\approx -\frac{k - k_n}{k_n}\left[E_n(r)E_n'(r) + rE_n(r)E_n''(r) - rE_n'^2(r)\right]_{R_+}, \end{aligned} \quad \text{(C18)}$$

in the last line keeping only terms linear in $k - k_n$ and using the facts that outside the system, $E_n'(r) = C_nH'(k_nr)k_n$ and $E_n''(r) = C_nH''(k_nr)k_n$, and also that $C(k) \approx C_n + C'(k_n)(k - k_n)$, in which the last term can be dropped.

Now, evaluating the second integral in Eq. (C12) to first order,

$$\int_0^R \left(\varepsilon k_n^2 - \varepsilon k^2 - \Delta\varepsilon k^2\right)EE_n dr \approx 2k_n(k_n - k)\int_0^R \varepsilon E_n^2 dr - k_n^2 \int_0^R \Delta\varepsilon E_n^2 dr, \quad \text{(C19)}$$

we obtain the first-order correction to the RS wave number,

$$k - k_n \approx \frac{-k_n \int_0^R \Delta\varepsilon(r)E_n^2(r)dr}{2\int_0^R \varepsilon(r)E_n^2(r)dr + k_n^{-2}\left[E_n(r)E_n'(r) + rE_n(r)E_n''(r) - rE_n'^2(r)\right]_{R_+}}\,. \quad \text{(C20)}$$

This result is equivalent to Eq. (C11). In fact, if the RSs are normalized according to Eq. (C6), the denominator in Eq. (C20) disappears, making Eqs. (C11) and (C20) identical.

2. TM polarization

Now we apply the approaches used in Appendix C 1 to the RSs in TM polarization. In this polarization, Maxwell's wave equation is also reducible to a scalar wave equation, but for the magnetic field [26,27],

$$\left[\frac{d}{dr}\frac{1}{\varepsilon(r)}\frac{d}{dr}-\frac{\alpha^2}{r^2\varepsilon(r)}+k_n^2\right]H_n(r)=0, \tag{C21}$$

where $-iH_n(r)/r$ is the radially-dependent part of the RS magnetic field. The wave equation for a perturbed RS with the wave number k and magnetic field $-iH(r)/r$ is given by

$$\left[\frac{d}{dr}\frac{1}{\varepsilon_p(r)}\frac{d}{dr}-\frac{\alpha^2}{r^2\varepsilon_p(r)}+k^2\right]H(r)=0, \tag{C22}$$

where

$$\varepsilon_p(r)=\varepsilon(r)+\Delta\varepsilon(r) \tag{C23}$$

and $\Delta\varepsilon(r)$ is a perturbation of the permittivity, as before.

RSE-based approaches

Again, using the solution of the unperturbed problem, Eq. (C22) can be solved via a Lippmann-Schwinger equation

$$H(r)=-\frac{1}{k}\int_0^R G_k(r,r')\tilde{\Delta}(r')H(r')dr' \tag{C24}$$

where $G_k(r,r')$ is the unperturbed magnetic GF, satisfying the equation

$$\left[\frac{d}{dr}\frac{1}{\varepsilon(r)}\frac{d}{dr}-\frac{\alpha^2}{r^2\varepsilon(r)}+k^2\right]G_k(r,r')=k\delta(r-r'), \tag{C25}$$

and

$$\tilde{\Delta}(r)=\tilde{\Delta}_1(r)+\tilde{\Delta}_2(r) \tag{C26}$$

is a perturbation, with

$$\tilde{\Delta}_1(r)=\frac{d}{dr}\frac{1}{\varepsilon_p(r)}\frac{d}{dr}-\frac{d}{dr}\frac{1}{\varepsilon(r)}\frac{d}{dr}, \tag{C27}$$

$$\tilde{\Delta}_2(r)=-\frac{\alpha^2}{r^2\varepsilon_p(r)}+\frac{\alpha^2}{r^2\varepsilon(r)}. \tag{C28}$$

The GF has the following Mittag-Leffler expansions within the system ($r\leqslant R$), again in terms of the RSs *only*,

$$G_k(r,r')=\sum_n\frac{H_n(r)H_n(r')}{k-k_n}=\sum_n\frac{k}{k_n}\frac{H_n(r)H_n(r')}{k-k_n}, \tag{C29}$$

provided that the RS fields are properly normalized [26].

Substituting the second series from Eq. (C29) into Eq. (C24), we obtain

$$H(r)=\sum_n c_nH_n(r), \tag{C30}$$

where

$$c_nk_n(k-k_n)=-\int_0^R H_n(r)\tilde{\Delta}(r)H(r)dr. \tag{C31}$$

Below we evaluate the effect of the two parts of the perturbation, Eqs. (C27) and (C28), separately, omitting the dependence on r for brevity of notations. Integrating by parts and using the facts that $H_n(0)=0$ and $\Delta\varepsilon(R)=0$, the first part of the perturbation integral takes the form

$$\begin{aligned}\int_0^R H_n\tilde{\Delta}_1Hdr&=\int_0^R H_n\left(\frac{d}{dr}\frac{1}{\varepsilon_p}-\frac{d}{dr}\frac{1}{\varepsilon}\right)H'dr\\&=\left[H_n\left(\frac{1}{\varepsilon_p}-\frac{1}{\varepsilon}\right)H'\right]_0^R-\int_0^R H_n'\left(\frac{1}{\varepsilon_p}-\frac{1}{\varepsilon}\right)H'dr\\&=\int_0^R H_n'\frac{\Delta\varepsilon}{\varepsilon\varepsilon_p}H'dr,\end{aligned} \tag{C32}$$

while the second part can be written as

$$\begin{aligned}\int_0^R H_n\tilde{\Delta}_2Hdr&=\int_0^R H_n\left(-\frac{\alpha^2}{r^2\varepsilon_p}+\frac{\alpha^2}{r^2\varepsilon}\right)Hdr\\&=\int_0^R H_n\frac{\alpha^2}{r^2}\frac{\Delta\varepsilon}{\varepsilon\varepsilon_p}Hdr.\end{aligned} \tag{C33}$$

As in the TE case, one can use in Eqs. (C32) and (C33) the expansion Eq. (C30), in order to obtain an RSE matrix equation. However, Eq. (C32) involves the first derivatives of the fields. A naive way to proceed would be to simply differentiate the series Eq. (C30),

$$H'(r)=\sum_n c_nH_n'(r), \tag{C34}$$

and substitute it into Eq. (C32). Recalling the expressions for $E_n^{\parallel}$ and $E_n^{\perp}$, the tangential and normal components (with respect to the sphere surface) of the RS electric field in TM polarization [26],

$$\begin{aligned}E_n^{\parallel}(r)&=-\frac{1}{k_n\varepsilon(r)}H_n'(r),\\E_n^{\perp}(r)&=-\frac{\alpha}{k_nr\varepsilon(r)}H_n(r),\end{aligned} \tag{C35}$$

this results in a linear matrix eigenvalue problem in terms of the RSs *only*,

$$c_n(k-k_n)=-\sum_m c_mk_m\int_0^R\mathbf{E}_n\cdot\frac{\Delta\varepsilon}{\varepsilon_p}\varepsilon\mathbf{E}_mdr, \tag{C36}$$

where $\mathbf{E}_n$ is a vector field with components $E_n^{\parallel}$ and $E_n^{\perp}$ [with $\mathbf{E}_n(r)/r$ being the radial part of the actual vector of the RS electric field]. Applying the diagonal approximation to Eq. (C36) yields the following first-order formula:

$$k-k_n\approx-k_n\int_0^R\mathbf{E}_n\cdot\frac{\Delta\varepsilon}{\varepsilon_p}\varepsilon\mathbf{E}_ndr, \tag{C37}$$

which is generally incorrect for boundary perturbations, as it is shown below. Note that Eq. (C36) is still rigorous. However, it was shown in [26] to be a slowly converging version of the RSE for boundary perturbations, such as a change of the size of a homogeneous dielectric sphere. The approximation Eq. (C37), in turn, has some missing terms in first order and can be rectified by including a proper contribution of all other RSs, similar to the effect of static modes discussed in the main text and also in Appendix A above.

The incorrect first-order result Eq. (C37) can also be fixed by taking into account Maxwell's boundary conditions in the field expansion. Since the tangential components of the electric field, such as $E_n^{\parallel}$, are continuous, it is better to expand a continuous function in terms of continuous ones, so that instead of Eq. (C34), one can use an expansion

$$\frac{H'(r)}{\varepsilon_p(r)} = \sum_n c_n \frac{H_n'(r)}{\varepsilon(r)} \quad + \quad \text{static modes}, \tag{C38}$$

in accordance with Eq. (C35) for the tangential component of the electric field. Using the expansions Eqs. (C38) and (C30), respectively, in the integrals Eqs. (C32) and (C33) results in a different linear matrix eigenvalue problem,

$$c_n(k-k_n) = -\sum_m c_m k_m (V_{nm}^{\parallel} + V_{nm}^{\perp}) + \text{static modes}, \tag{C39}$$

where the matrix elements are given by

$$V_{nm}^{\parallel} = \int_0^R E_n^{\parallel} \Delta\varepsilon E_m^{\parallel} dr,$$
$$V_{nm}^{\perp} = \int_0^R E_n^{\perp} \frac{\Delta\varepsilon}{\varepsilon_p} \varepsilon E_m^{\perp} dr. \tag{C40}$$

Extracting the diagonal approximation from Eq. (C39) results in the correct first order for the RS wave number,

$$k - k_n \approx -k_n (V_{nn}^{\parallel} + V_{nn}^{\perp}), \tag{C41}$$

which is equivalent to Eq. (A35) for spherically-symmetric perturbations $\Delta\varepsilon(r)$ and can be derived also by a direct evaluation of the first-order correction, as done in Appendix C 1 b for TE polarization (see also Appendix E below for a general derivation).

Note that replacing the expansion Eq. (C34) with Eq. (C38) and using the latter together with Eq. (C30) implies that instead of expanding a scalar function $H(r)$ into a complete set of scalar functions $H_n(r)$ we expand a vector field $\{H(r), H'(r)/\varepsilon_p(r)\}$ into vector functions $\{H_n(r), H_n'(r)/\varepsilon(r)\}$. The latter may, however, be incomplete, so one has to use for completeness additional vector functions—static modes, which are added to Eqs. (C38) and (C39), and their components have to be added also to the expansion Eq. (C30). It has been shown in [23] that such functions are indeed required for the RSE.

APPENDIX D: OTHER FIELDS OF PHYSICS

In this section, we consider examples from other fields of physics describing wave phenomena and demonstrate that for systems with interfaces, the same effect of higher-order terms of the perturbation series contributing to the first-order correction (in the deformation depth) to the eigenmode wave numbers takes place.

1. Condensed matter physics: Effective-mass approximation for semiconductor heterostructures

In condensed matter physics, an effective Schrödinger wave equation describing the motion of a carrier (an electron or a hole) has the following form in the effective-mass approximation:

$$\left[-\frac{\hbar^2}{2}\nabla\cdot\hat{\boldsymbol{\mu}}^{-1}(\mathbf{r})\nabla + V(\mathbf{r})\right]\Psi(\mathbf{r}) = E\Psi(\mathbf{r}), \tag{D1}$$

where $\hat{\boldsymbol{\mu}}(\mathbf{r})$ is a spatially-dependent tensor of the effective mass, which depends on the material and can change abruptly at material interfaces. While typical semiconductor heterostructures have planar interfaces separating different materials, we concentrate below on spherically-symmetric systems, in order to ease a comparison with TM modes in optical systems considered in detail in Appendix C 2.

For a spherically-symmetric isotropic effective mass $\mu(r)$ and spherically-symmetric potential $V(r)$, Eq. (D1) reduces to

$$\left[\frac{d}{dr}\frac{1}{\mu(r)}\frac{d}{dr} - \frac{\alpha^2}{r^2} - V(r) + k_n^2\right]\psi_n(r) = 0, \tag{D2}$$

where $\psi_n(r)/r$ is the radial part of the full wave function $\Psi(\mathbf{r})$, $k_n = \sqrt{E_n}$ is a wave number corresponding to the state energy E_n, and index n is used to label quantum RSs. Here units with $\hbar^2 = 2$ are used for brevity of notations.

We consider also a perturbed Schrödinger wave equation

$$\left[\frac{d}{dr}\frac{1}{\mu_p(r)}\frac{d}{dr} - \frac{\alpha^2}{r^2} - V_p(r) + k^2\right]\psi(r) = 0 \tag{D3}$$

for perturbed wave number k and wave function $\psi(r)$, and

$$\mu_p(r) = \mu(r) + \Delta\mu(r), \quad V_p(r) = V(r) + \Delta V(r), \tag{D4}$$

where $\Delta\mu(r)$ and $\Delta V(r)$ are perturbations of the effective mass and potential, respectively. As in Appendix C 2, Eq. (D3) can be solved with the help of the GF $G_k(r, r')$ of the unperturbed problem, which satisfies the equation

$$\left[\frac{d}{dr}\frac{1}{\mu(r)}\frac{d}{dr} - \frac{\alpha^2}{r^2} - V(r) + k^2\right]G_k(r, r') = k\delta(r - r') \tag{D5}$$

and has the following Mittag-Leffler expansions in terms of the unperturbed RSs:

$$G_k(r, r') = \sum_n \frac{\psi_n(r)\psi_n(r')}{2(k-k_n)} = \sum_n \frac{k}{k_n}\frac{\psi_n(r)\psi_n(r')}{2(k-k_n)}, \tag{D6}$$

provided that the RS fields are properly normalized [38–40]. The Lippmann-Schwinger equation has the form

$$\psi(r) = -\frac{1}{k}\int_0^R G_k(r, r')\tilde{\Delta}(r')\psi(r')dr', \tag{D7}$$

where

$$\tilde{\Delta}(r) = \frac{d}{dr}\frac{1}{\mu_p(r)}\frac{d}{dr} - \frac{d}{dr}\frac{1}{\mu(r)}\frac{d}{dr} - \Delta V(r). \tag{D8}$$

Substituting the second series from Eq. (D6) into Eq. (D7) and integrating by parts, we obtain

$$\psi(r) = \sum_n c_n \psi_n(r), \tag{D9}$$

where

$$2k_n(k-k_n)c_n = -\int_0^R \psi_n' \frac{\Delta\mu}{\mu\mu_p}\psi' dr + \int_0^R \psi_n \Delta V \psi dr. \tag{D10}$$

Then, substituting the expansion Eq. (D9) into the integrals in Eq. (D10) results in a linear matrix eigenvalue problem

$$c_n(k-k_n)=\sum_m c_m\frac{-U_{nm}+V_{nm}}{2k_n}, \tag{D11}$$

where

$$U_{nm}=\int_0^R \psi_n'\frac{\Delta\mu}{\mu\mu_p}\psi_m' dr,$$
$$V_{nm}=\int_0^R \psi_n\Delta V\psi_m dr \tag{D12}$$

are the matrix elements of the perturbation. The diagonal approximation to Eq. (D11) results in a first-order formula for the wave number,

$$k-k_n\approx -\frac{U_{nn}}{2k_n}+\frac{V_{nn}}{2k_n}, \tag{D13}$$

in which the first term is incorrect for boundary perturbations. In terms of the rigorous matrix equation (D11), the first term in Eq. (D13) can be corrected by taking into account the contribution of all other RSs in all perturbation orders, which is similar to the cumulative effect of static modes in optical systems, considered in the main text. Alternatively, it can be corrected also by using an expansion for the derivatives,

$$\frac{\psi'(r)}{\mu_p(r)}=\sum_n c_n\frac{\psi_n'(r)}{\mu(r)}+\text{additional modes}, \tag{D14}$$

expanding a continuous function into a set of continuous functions. One should be mindful, however, that some "additional modes" have to be included for completeness in the expansion in this case, similar to static modes in optical problems. In this formulation, however, they do not influence the correct first-order result, which can be obtained from the diagonal approximation after using Eq. (D14) in the first integral in Eq. (D10),

$$k-k_n\approx -\frac{\tilde{U}_{nn}}{2k_n}+\frac{V_{nn}}{2k_n}, \tag{D15}$$

where

$$\tilde{U}_{nn}=\int_0^R \Delta\mu\left(\frac{\psi_n'}{\mu}\right)^2 dr \tag{D16}$$

is a corrected diagonal matrix element due to the effective mass perturbation.

2. Acoustics

The acoustic wave equation for the total pressure $P=p+\delta p$ in inhomogeneous quiescent (static) media, with constant ambient pressure p, spatially variable ambient density ρ and speed of sound c, with gravity neglected, can be written, assuming harmonic time dependence, as [41]

$$\rho\nabla\cdot\left(\frac{1}{\rho}\nabla P\right)+k^2P=0, \tag{D17}$$

which, in case of a spherically symmetric inhomogeneity creating an acoustic cavity can be written as

$$\left(\rho(r)\frac{d}{dr}\frac{1}{\rho(r)}\frac{d}{dr}-\frac{\alpha^2}{r^2}\right)P(r)+k^2P(r)=0\,. \tag{D18}$$

The acoustic boundary conditions at an interface mean that the pressure is continuous, although not necessarily smooth, but the medium velocity normal to the surface is continuous. "The requirement of pressure continuity assumes no mass transport across the interface and neglects surface tension; under such circumstances it is the fluid-dynamic counterpart of Newton's third law" [41]. The requirement for the velocity component approximately means that the normal component of the displacement to the surface element is continuous. The second-order operator in Eq. (D17) has the form similar to that in Eq. (D1), with a discontinuous density $\rho(\boldsymbol{r})$ in the case of a boundary between two media playing the same role as the discontinuous effective mass tensor $\hat{\mu}(\boldsymbol{r})$, and an equation similar to Eq. (D15) would determine the eigenmode shift if the boundary of the cavity is deformed. Note that here it is assumed that the densities of both media separated by an interface are finite, allowing propagation of waves on either side of the interface. This is different from the boundary perturbation for eigenmodes in cavities where a soft or hard (rigid) boundary is assumed. The former assumption leads to zero pressure at the boundary, whereas the latter leads to zero propagation velocity along the surface normal. Both cases allow application of the perturbative methods from [11] and have been treated in the literature [42,43].

APPENDIX E: FULL DERIVATION OF EQ. (18)

In this section, we provide a full derivation of Eq. (18) of the main text.

Maxwell's equations for the unperturbed system can be written in the form [25]

$$[k_n\hat{\mathbb{P}}_0(k_n,\mathbf{r})-\hat{\mathbb{D}}(\mathbf{r})]\vec{\mathbb{F}}_n(\mathbf{r})=0, \tag{E1}$$

where

$$\hat{\mathbb{P}}_0(k,\mathbf{r})=\begin{pmatrix}\hat{\boldsymbol{\epsilon}}(k,\mathbf{r}) & -i\hat{\boldsymbol{\xi}}(k,\mathbf{r})\\ i\hat{\boldsymbol{\zeta}}(k,\mathbf{r}) & \hat{\boldsymbol{\mu}}(k,\mathbf{r})\end{pmatrix},$$
$$\hat{\mathbb{D}}(\mathbf{r})=\begin{pmatrix}\hat{\mathbf{0}} & \nabla\times\\ \nabla\times & \hat{\mathbf{0}}\end{pmatrix}, \tag{E2}$$
$$\vec{\mathbb{F}}_n(\mathbf{r})=\begin{pmatrix}\mathbf{E}_n(\mathbf{r})\\ i\mathbf{H}_n(\mathbf{r})\end{pmatrix},$$

as in Eqs. (12) and (13) of the main text. The electromagnetic field $\vec{\mathbb{F}}(\mathbf{r})$ and the wave number k of a perturbed RS are, in turn, solutions of the perturbed Maxwell equations

$$[k\hat{\mathbb{P}}(k,\mathbf{r})-\hat{\mathbb{D}}(\mathbf{r})]\vec{\mathbb{F}}(\mathbf{r})=0, \tag{E3}$$

where $\hat{\mathbb{P}}(k,\mathbf{r})=\hat{\mathbb{P}}_0(k,\mathbf{r})+\Delta\hat{\mathbb{P}}(k,\mathbf{r})$,

$$\vec{\mathbb{F}}(\mathbf{r})=\begin{pmatrix}\mathbf{E}(\mathbf{r})\\ i\mathbf{H}(\mathbf{r})\end{pmatrix}, \tag{E4}$$

and

$$\Delta\hat{\mathbb{P}}(k,\mathbf{r})=\begin{pmatrix}\Delta\hat{\boldsymbol{\epsilon}}(k,\mathbf{r}) & -i\Delta\hat{\boldsymbol{\xi}}(k,\mathbf{r})\\ i\Delta\hat{\boldsymbol{\zeta}}(k,\mathbf{r}) & \Delta\hat{\boldsymbol{\mu}}(k,\mathbf{r})\end{pmatrix} \tag{E5}$$

is a perturbation. As in the main text, we assume below isotropic and reciprocal materials. Multiplying Eq. (E1) with

$\vec{\mathbb{F}}$ and Eq. (E3) with $\vec{\mathbb{F}}_n$, integrating both equations over a volume V_1 containing the original system volume and the perturbation $\Delta\hat{\mathbb{P}}$, and taking the difference between the results, we obtain

$$\int_{V_1}[\vec{\mathbb{F}}(\mathbf{r})\cdot k_n\hat{\mathbb{P}}_0(k_n,\mathbf{r})\vec{\mathbb{F}}_n(\mathbf{r})-\vec{\mathbb{F}}_n(\mathbf{r})\cdot k\hat{\mathbb{P}}(k,\mathbf{r})\vec{\mathbb{F}}(\mathbf{r})]d\mathbf{r} - \int_{V_1}[\vec{\mathbb{F}}(\mathbf{r})\cdot\hat{\mathbb{D}}(\mathbf{r})\vec{\mathbb{F}}_n(\mathbf{r})-\vec{\mathbb{F}}_n(\mathbf{r})\cdot\hat{\mathbb{D}}(\mathbf{r})\vec{\mathbb{F}}(\mathbf{r})]d\mathbf{r}=0. \tag{E6}$$

The second line in Eq. (E6) can be transformed, using the divergence theorem, to the surface integral [25]

$$-i\oint_{S_1}[\mathbf{E}_n(\mathbf{r})\times\mathbf{H}(\mathbf{r})-\mathbf{E}(\mathbf{r})\times\mathbf{H}_n(\mathbf{r})]\cdot d\mathbf{S}, \tag{E7}$$

where S_1 is the boundary of V_1. Note that (i) for internal perturbations, $V_1=V_0$ and $S_1=S_0$, where V_0 is the unperturbed system volume and S_0 is its boundary; (ii) for finite external perturbations, including deformations outwards, $V_1=V$ and $S_1=S$, where V is the perturbed system volume and S is its boundary; and (iii) for both internal and external perturbations, $V_1=V_0\cup V$ so that S_1 is neither S_0 nor S but includes both surfaces.

To proceed with the first line in Eq. (E6), we use the real, normalized vector field $\hat{\mathbf{n}}(\mathbf{r})$, see Fig. 2 in the main text, which is normal to both surfaces S_0 and S of the unperturbed and perturbed systems, respectively. We then define the projections of perturbed and unperturbed RS fields,

$$\vec{\mathbb{F}}^{\perp}(\mathbf{r})=\begin{pmatrix}\mathbf{E}^{\perp}(\mathbf{r})\\ i\mathbf{H}^{\perp}(\mathbf{r})\end{pmatrix}=\begin{pmatrix}\hat{\mathbf{n}}(\mathbf{r})[\hat{\mathbf{n}}(\mathbf{r})\cdot\mathbf{E}(\mathbf{r})]\\ \hat{\mathbf{n}}(\mathbf{r})[\hat{\mathbf{n}}(\mathbf{r})\cdot i\mathbf{H}(\mathbf{r})]\end{pmatrix},$$
$$\vec{\mathbb{F}}_n^{\perp}(\mathbf{r})=\begin{pmatrix}\mathbf{E}_n^{\perp}(\mathbf{r})\\ i\mathbf{H}_n^{\perp}(\mathbf{r})\end{pmatrix}=\begin{pmatrix}\hat{\mathbf{n}}(\mathbf{r})[\hat{\mathbf{n}}(\mathbf{r})\cdot\mathbf{E}_n(\mathbf{r})]\\ \hat{\mathbf{n}}(\mathbf{r})[\hat{\mathbf{n}}(\mathbf{r})\cdot i\mathbf{H}_n(\mathbf{r})]\end{pmatrix}, \tag{E8}$$

which are normal to both S_0 and S, and

$$\vec{\mathbb{F}}^{\parallel}(\mathbf{r})=\vec{\mathbb{F}}(\mathbf{r})-\vec{\mathbb{F}}^{\perp}(\mathbf{r}),$$
$$\vec{\mathbb{F}}_n^{\parallel}(\mathbf{r})=\vec{\mathbb{F}}_n(\mathbf{r})-\vec{\mathbb{F}}_n^{\perp}(\mathbf{r}), \tag{E9}$$

which are parallel (tangential) to both surfaces. Note that the field is normal or parallel to a surface only when its argument $\mathbf{r}$ describes a point at that surface. Now, according to Maxwell's boundary conditions, fields $\vec{\mathbb{F}}^{\parallel}(\mathbf{r})$ and $\hat{\mathbb{P}}(k,\mathbf{r})\vec{\mathbb{F}}^{\perp}(\mathbf{r})$ are continuous both at S_0 and S. Similarly, the unperturbed fields $\vec{\mathbb{F}}_n^{\parallel}(\mathbf{r})$ and $\hat{\mathbb{P}}_0(k_n,\mathbf{r})\vec{\mathbb{F}}_n^{\perp}(\mathbf{r})$, introduced in the same manner, are also continuous at S_0 and S. To derive the first-order correction to the wave number, we approximate the perturbed RS field in zeroth order as

$$\vec{\mathbb{F}}^{\parallel}(\mathbf{r})\approx\vec{\mathbb{F}}_n^{\parallel}(\mathbf{r}),$$
$$\hat{\mathbb{P}}(k,\mathbf{r})\vec{\mathbb{F}}^{\perp}(\mathbf{r})\approx\hat{\mathbb{P}}_0(k_n,\mathbf{r})\vec{\mathbb{F}}_n^{\perp}(\mathbf{r}), \tag{E10}$$

in this way respecting Maxwell's boundary conditions on both surfaces. This results in

$$\vec{\mathbb{F}}=\vec{\mathbb{F}}_n^{\parallel}+\hat{\mathbb{P}}^{-1}(k)\hat{\mathbb{P}}_0(k_n)\vec{\mathbb{F}}_n^{\perp}=\vec{\mathbb{F}}_n+[\hat{\mathbb{P}}^{-1}(k)\hat{\mathbb{P}}_0(k_n)-1]\vec{\mathbb{F}}_n^{\perp}, \tag{E11}$$

where $\hat{\mathbb{P}}^{-1}(k)$ is the inverse of $\hat{\mathbb{P}}(k)$. Note that here and below, we omit for brevity the spatial arguments but keep the wave number dependence. Now, using Eq. (E11), the reciprocity of the optical systems, and a Taylor expansion to first order in $k-k_n$ of terms containing the generalized permittivity, such as $\hat{\mathbb{P}}(k,\mathbf{r})\approx\hat{\mathbb{P}}(k_n,\mathbf{r})+[\hat{\mathbb{P}}(k,\mathbf{r})]'_{k_n}(k-k_n)$, we find for the integrand of the first line in Eq. (E6)

$$\begin{aligned}[k_n\hat{\mathbb{P}}_0(k_n)-k\hat{\mathbb{P}}(k)]\vec{\mathbb{F}} &\approx [k_n\hat{\mathbb{P}}_0(k_n)-k\hat{\mathbb{P}}_0(k)]\vec{\mathbb{F}}_n-k\Delta\hat{\mathbb{P}}(k)\vec{\mathbb{F}}_n+k_n\hat{\mathbb{P}}_0(k_n)[\hat{\mathbb{P}}^{-1}(k)\hat{\mathbb{P}}_0(k_n)-1]\vec{\mathbb{F}}_n^{\perp}-k[\hat{\mathbb{P}}_0(k_n)-\hat{\mathbb{P}}(k)]\vec{\mathbb{F}}_n^{\perp}\\ &\approx -(k-k_n)[k\hat{\mathbb{P}}_0(k)]'_{k_n}\vec{\mathbb{F}}_n-k\Delta\hat{\mathbb{P}}(k)\vec{\mathbb{F}}_n^{\parallel}-k_n\hat{\mathbb{P}}_0(k_n)\hat{\mathbb{P}}^{-1}(k)[\hat{\mathbb{P}}(k)-\hat{\mathbb{P}}_0(k_n)]\vec{\mathbb{F}}_n^{\perp}-k[\hat{\mathbb{P}}_0(k_n)-\hat{\mathbb{P}}_0(k)]\vec{\mathbb{F}}_n^{\perp}\\ &\approx -(k-k_n)[k\hat{\mathbb{P}}_0(k)]'_{k_n}\vec{\mathbb{F}}_n-k\Delta\hat{\mathbb{P}}(k)\vec{\mathbb{F}}_n^{\parallel}-k_n\hat{\mathbb{P}}_0(k_n)\hat{\mathbb{P}}^{-1}(k)\Delta\hat{\mathbb{P}}(k)\vec{\mathbb{F}}_n^{\perp},\end{aligned} \tag{E12}$$

in the last line neglecting higher-order terms, which are

$$k_n(k-k_n)[\hat{\mathbb{P}}(k)-\hat{\mathbb{P}}_0(k_n)]\hat{\mathbb{P}}^{-1}(k)[\hat{\mathbb{P}}_0(k)]'_{k_n}\vec{\mathbb{F}}_n^{\perp}+(k-k_n)^2[\hat{\mathbb{P}}_0(k)]'_{k_n}\vec{\mathbb{F}}_n^{\perp}.$$

Finally, using an analytic continuation $\vec{\mathbb{Q}}(k,\mathbf{r})$ of the RS fields outside V_1, such that $\vec{\mathbb{Q}}(k,\mathbf{r})=\vec{\mathbb{F}}(\mathbf{r})$ and $\vec{\mathbb{Q}}(k_n,\mathbf{r})=\vec{\mathbb{F}}_n(\mathbf{r})$, and also Taylor expanding it to first order,

$$\vec{\mathbb{Q}}(k,\mathbf{r})\approx\vec{\mathbb{F}}_n(\mathbf{r})+(k-k_n)\vec{\mathbb{F}}'_n(\mathbf{r}), \tag{E13}$$

the surface term Eq. (E7) transforms to

$$-i(k-k_n)\oint_{S_1}\left[\mathbf{E}_n(\mathbf{r})\times\mathbf{H}'_n(\mathbf{r})-\mathbf{E}'_n(\mathbf{r})\times\mathbf{H}_n(\mathbf{r})\right]\cdot d\mathbf{S}. \tag{E14}$$

Substituting the last line of Eq. (E12) and the surface integral Eq. (E14) into Eq. (E6) and taking the limit $k\to k_n$, we find the first-order correction to the wave number,

$$k-k_n\approx-\frac{k_n\int_{V_1}[\vec{\mathbb{F}}_n^{\parallel}\cdot\Delta\hat{\mathbb{P}}\vec{\mathbb{F}}_n^{\parallel}+\vec{\mathbb{F}}_n^{\perp}\cdot\hat{\mathbb{P}}_0\hat{\mathbb{P}}^{-1}\Delta\hat{\mathbb{P}}\vec{\mathbb{F}}_n^{\perp}]d\mathbf{r}}{\int_{V_1}\vec{\mathbb{F}}_n\cdot[k\hat{\mathbb{P}}_0]'_{k_n}\vec{\mathbb{F}}_n d\mathbf{r}+i\oint_{S_1}(\mathbf{E}_n\times\mathbf{H}'_n-\mathbf{E}'_n\times\mathbf{H}_n)\cdot d\mathbf{S}}. \tag{E15}$$

Using the analytic normalization [25] of the RSs,

$$\int_{V_1} \vec{\mathbb{F}}_n \cdot [k\hat{\mathbb{P}}_0]'_{k_n} \vec{\mathbb{F}}_n d\mathbf{r} + i \oint_{S_1} (\mathbf{E}_n \times \mathbf{H}'_n - \mathbf{E}'_n \times \mathbf{H}_n) \cdot d\mathbf{S} = \int_{V_0} \vec{\mathbb{F}}_n \cdot [k\hat{\mathbb{P}}_0]'_{k_n} \vec{\mathbb{F}}_n d\mathbf{r} + i \oint_{S_0} (\mathbf{E}_n \times \mathbf{H}'_n - \mathbf{E}'_n \times \mathbf{H}_n) \cdot d\mathbf{S} = 1, \quad \text{(E16)}$$

which is independent of the volume of integration, provided that it contains all the inhomogeneities of the system, we arrive at Eq. (18) of the main text. Note that the integration in the numerator of Eq. (E15) can be extended to the full space, as done in Eq. (18) of the main text, because $\Delta\hat{\mathbb{P}} = 0$ outside V_1. Using the second line of Eq. (E16), the final result Eq. (E15) simplifies to

$$k - k_n \approx -k_n \int [\vec{\mathbb{F}}_n^{\parallel} \cdot \Delta\hat{\mathbb{P}} \vec{\mathbb{F}}_n^{\parallel} + \vec{\mathbb{F}}_n^{\perp} \cdot \hat{\mathbb{P}}_0 \hat{\mathbb{P}}^{-1} \Delta\hat{\mathbb{P}} \vec{\mathbb{F}}_n^{\perp}] d\mathbf{r}\,. \quad \text{(E17)}$$

APPENDIX F: TREATING ANISOTROPIC PERMITTIVITIES

The first-order perturbation theory approach presented in the main text and detailed in Appendix E is based on the separation of the electromagnetic field into the normal and tangential components and approximating continuous components of the perturbed field with continuous unperturbed field components, see Eq. (E11). For the electric field only, such continuous components are the tangential electric field $\mathbf{E}^{\parallel}$ and the normal displacement field $\mathbf{D}^{\perp}$. The latter is, however, not proportional to the normal component of the electric field $\mathbf{E}^{\perp}$ if the permittivity tensor is anisotropic. One can instead have a more general relation

$$\boldsymbol{E} = \hat{A}\mathbf{E}^{\parallel} + \hat{B}\mathbf{D}^{\perp} \quad \text{(F1)}$$

with $\hat{A}$ and $\hat{B}$ being some tensors. Below we show how these tensors can be found, focusing on a two-dimensional case for clarity of presentation.

Let us first define the basis vectors $\hat{e}^{\perp}(\boldsymbol{r})$ and $\hat{e}^{\parallel}(\boldsymbol{r})$, where $\hat{e}^{\perp}$ is normal to the surface and $\hat{e}^{\parallel}$ is parallel (tangential) to the surface of the resonator at point $\mathbf{r}$. We write the electric field on the surface of the resonator in terms of these basis vectors as

$$\boldsymbol{E} = E_{\perp}\hat{e}^{\perp} + E_{\parallel}\hat{e}^{\parallel}, \quad \text{(F2)}$$

and the permittivity tensor in the new basis is given by

$$\hat{\boldsymbol{\varepsilon}} = \begin{pmatrix} \varepsilon_{\perp\perp} & \varepsilon_{\perp\parallel} \\ \varepsilon_{\parallel\perp} & \varepsilon_{\parallel\parallel} \end{pmatrix}, \quad \text{(F3)}$$

where for brevity we have dropped the dependence on the spatial coordinate $\boldsymbol{r}$ and the wave number k. The electric displacement field can be written as $\boldsymbol{D} = \hat{\boldsymbol{\varepsilon}}\boldsymbol{E}$, which takes the following matrix form:

$$\begin{pmatrix} D_{\perp} \\ D_{\parallel} \end{pmatrix} = \begin{pmatrix} \varepsilon_{\perp\perp} & \varepsilon_{\perp\parallel} \\ \varepsilon_{\parallel\perp} & \varepsilon_{\parallel\parallel} \end{pmatrix} \begin{pmatrix} E_{\perp} \\ E_{\parallel} \end{pmatrix} = \begin{pmatrix} \varepsilon_{\perp\perp}E_{\perp} + \varepsilon_{\perp\parallel}E_{\parallel} \\ \varepsilon_{\parallel\perp}E_{\perp} + \varepsilon_{\parallel\parallel}E_{\parallel} \end{pmatrix}. \quad \text{(F4)}$$

Clearly, the normal component of the displacement, $D_{\perp} = \varepsilon_{\perp\perp}E_{\perp} + \varepsilon_{\perp\parallel}E_{\parallel}$, which is continuous across the surface, contains a mixture of the normal and parallel components of the electric field. Then Eq. (F1) can be written in matrix form as

$$\begin{pmatrix} E_{\perp} \\ E_{\parallel} \end{pmatrix} = \begin{pmatrix} A_{\perp\perp} & A_{\perp\parallel} \\ A_{\parallel\perp} & A_{\parallel\parallel} \end{pmatrix} \begin{pmatrix} 0 \\ E_{\parallel} \end{pmatrix} + \begin{pmatrix} B_{\perp\perp} & B_{\perp\parallel} \\ B_{\parallel\perp} & B_{\parallel\parallel} \end{pmatrix} \begin{pmatrix} \varepsilon_{\perp\perp}E_{\perp} + \varepsilon_{\perp\parallel}E_{\parallel} \\ 0 \end{pmatrix}. \quad \text{(F5)}$$

The above matrix equation provides four simultaneous equations

$$\begin{aligned} 1 &= B_{\perp\perp}\varepsilon_{\perp\perp}, \\ 0 &= B_{\perp\perp}\varepsilon_{\perp\parallel} + A_{\perp\parallel}, \\ 0 &= B_{\parallel\perp}\varepsilon_{\perp\perp}, \\ 1 &= B_{\parallel\perp}\varepsilon_{\perp\parallel} + A_{\parallel\parallel}, \end{aligned} \quad \text{(F6)}$$

determining the following four components of matrices $\hat{A}$ and $\hat{B}$:

$$\begin{aligned} A_{\perp\parallel} &= -\varepsilon_{\perp\parallel}/\varepsilon_{\perp\perp}, \\ A_{\parallel\parallel} &= 1, \\ B_{\perp\perp} &= 1/\varepsilon_{\perp\perp}, \\ B_{\parallel\perp} &= 0\,. \end{aligned} \quad \text{(F7)}$$

All other components of $\hat{A}$ and $\hat{B}$ are not required as they do not contribute to Eq. (F1) and therefore can be set to zero for simplicity.

We note that a similar approach was used in [44] for closed nondispersive systems in three dimensions.

APPENDIX G: EVALUATION OF THE MATRIX ELEMENTS FOR ELLIPTICAL PERTURBATIONS

For TM polarization, the electric field of a nonmagnetic sphere is defined in spherical coordinates as

$$\mathbf{E}(\mathbf{r}) = E_{\phi}(\mathbf{r})\hat{\boldsymbol{\phi}} + E_{\theta}(\mathbf{r})\hat{\boldsymbol{\theta}} + E_r(\mathbf{r})\hat{\boldsymbol{r}}, \quad \text{(G1)}$$

or in terms of the notation of Eq. (A35) the field is given by

$$\mathbf{E}^{\parallel} = E_{\phi}(\mathbf{r})\hat{\boldsymbol{\phi}} + E_{\theta}(\mathbf{r})\hat{\boldsymbol{\theta}}, \quad \text{(G2)}$$

$$\mathbf{E}^{\perp} = E_r(\mathbf{r})\hat{\boldsymbol{r}}. \quad \text{(G3)}$$

The electric-field components of the RS with the wave number k_n take the form [13,45]

$$E_{\phi}(\mathbf{r}) = -A\frac{J'(x)}{n_r r}\frac{1}{\alpha}\frac{1}{\sin\theta}\frac{\partial}{\partial\phi}Y_{lm}(\theta,\phi), \quad \text{(G4)}$$

$$E_{\theta}(\mathbf{r}) = -A\frac{J'(x)}{n_r r}\frac{1}{\alpha}\frac{\partial}{\partial\theta}Y_{lm}(\theta,\phi), \quad \text{(G5)}$$

$$E_r(\mathbf{r}) = -A\frac{j_l(x)}{n_r r}\alpha Y_{lm}(\theta,\phi), \quad \text{(G6)}$$

where $J(x) = x j_l(x)$ is the Ricatti-Bessel function, $j_l(x)$ is the spherical Bessel function, and the prime means the derivative with respect to the argument. In Eqs. (G4)–(G6) we use $x =$

$n_r k_n r$, where $n_r = \sqrt{\varepsilon(k_n)}$ is the refractive index of the sphere. The normalization constant A is defined as [45,46]

$$A = \frac{1}{J(x_0)} \frac{1}{\sqrt{R\left(n_r^2 - 1\right)D}}, \tag{G7}$$

$$D = \left(\frac{j_{l-1}(x_0)}{j_l(x_0)} - \frac{l}{x_0}\right)^2 \frac{1}{n_r^2} + \frac{\alpha^2}{x_0^2} + \eta C, \tag{G8}$$

$$\left(n_r^2 - 1\right)C = -\frac{2l}{x_0^2} + \left(\frac{j_{l-1}(x_0)}{j_l(x_0)}\right)^2 - \frac{j_{l-2}(x_0)}{j_l(x_0)}, \tag{G9}$$

$$\eta = \frac{k}{2\varepsilon(k)} \frac{\partial \varepsilon(k)}{\partial k}\bigg|_{k=k_n}, \tag{G10}$$

where $x_0 = n_r k_n R$, with R being the radius of the sphere. $Y_{lm}(\theta, \phi)$ are real-valued spherical harmonics in the form

$$Y_{lm}(\theta, \phi) = c_l^{|m|} P_l^{|m|}(\cos\theta)\chi_m(\phi), \tag{G11}$$

$$c_l^{|m|} = \sqrt{\frac{2l+1}{2} \frac{(l-|m|)!}{(l+|m|)!}}, \tag{G12}$$

$$\chi_m(\phi) = \begin{cases} \pi^{-1/2}\sin(m\phi) & m < 0 \\ (2\pi)^{-1/2} & m = 0 \\ \pi^{-1/2}\cos(m\phi) & m > 0, \end{cases} \tag{G13}$$

and $P_l^{|m|}$ are the associated Legendre polynomials.

Generally, the integral in Eq. (A35) needs to be evaluated in 3D, but in case of a cylindrically-symmetric perturbation the integration along ϕ can be done analytically, and the remaining integrals along θ and r can be done numerically. In case of the sphere deformed to a cylindrically-symmetric ellipsoid, the lower limit of the radial integral is given by the equation of the corresponding ellipse and takes the form

$$r_<(\theta) = \frac{ab}{\sqrt{b\cos^2\theta + a\sin^2\theta}} \tag{G14}$$

where a and b are, respectively, the semiminor and semimajor axes of the ellipse. This makes the integrals along θ and r nonseparable. As the integral along r cannot be done analytically, we evaluate the double integral numerically in 2D. We note that to align the ellipsoid as in [7], the ellipse given by Eq. (G14) is rotated by 90° degrees, so θ in Eq. (G14) is replaced by $\pi/2 - \theta$. This results in the semiaxis a aligned with the polar axis. If we do not make this substitution then the semiaxis a would be aligned normal to the polar axis and the modes $m = 0$ and $|m| = 1$ would be swapped. Here we consider perturbations changing the length a. Combining the above we can derive the following form of the diagonal matrix element:

$$V_{nn} = \left[\frac{c_l^{|m|}A}{n_r}\right]^2 \int_0^\pi \sin\theta d\theta \int_{r_<(\theta)}^R dr \Bigg\{ \left[P_l^{|m|}(\cos\theta)\right]^2 [\alpha j_l(n_r kr)]^2 \frac{\varepsilon\Delta\varepsilon}{\varepsilon + \Delta\varepsilon} + \frac{1}{\alpha^2}\left(\frac{m^2}{\sin^2\theta}\left[P_l^{|m|}(\cos\theta)\right]^2 + \sin^2\theta\left[\frac{P_l^{|m|}(\cos\theta)}{\cos\theta}\right]^2\right)[J'(n_r kr)]^2\Delta\varepsilon \Bigg\}, \tag{G15}$$

with $\Delta\varepsilon = 1 - \varepsilon(k)$ for the shape or size perturbation. In case of homogeneous size perturbations, which are spherically symmetric, the integral along θ can also be done analytically, leaving only the radial integral to be evaluated, which then takes the form

$$V_{nn} = \frac{A^2}{n_r^2} \int_{r_<}^{r_>} dr \left\{ \alpha^2 j_l^2(n_r kr) \frac{\varepsilon\Delta\varepsilon}{\varepsilon + \Delta\varepsilon} + [J'(n_r kr)]^2 \Delta\varepsilon \right\}, \tag{G16}$$

where $r_<$ and $r_>$ denote, respectively, the upper and lower limits of r.

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