Time-independent perturbation for leaking electromagnetic modes in open systems with application to resonances in microdroplets

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Laser cavities are open systems, in that energy can leak to the outside via output coupling. The "normal modes" are therefore quasinormal modes, with eigenvalues that are complex and eigenfunctions that extend outside the cavity, such that any normalization integral is dominated by the region outside; in short, such systems are non-Hermitian. This paper addresses the question: How is the complex eigenvalue (i.e., the mode frequency) changed when the cavity is perturbed by a small change of dielectric constant? The usual time-independent perturbation theory fails because of non-Hermiticity. By generalizing the work of Zeldovich [Sov. Phys.—JETP **12**, 542 (1961)] for scalar fields in one dimension, we express the change of frequency in terms of matrix elements involving the unperturbed eigenfunctions, so that the problem is reduced to quadrature. We then apply the formalism to shape perturbations of a dielectric microdroplet, and give analytic formulas for the frequency shifts of the morphology-dependent resonances. These results are, surprisingly, independent of the radial wave function, so that all integrals can be performed and explicit algebraic expressions are given for axially symmetric perturbations.

I. INTRODUCTION

A. Quasinormal modes in open systems

In this paper we consider the electromagnetic modes of an *open* system formed by mirrors and dielectrics. For our purpose, a system is said to be closed if the fields are confined to a finite region in space, for example in an enclosed microwave cavity, or more generally if the energy density is integrable. In contrast, it is said to be open if the fields are not strictly confined, but can leak to the whole universe. All optical cavities with some degree of output coupling belong to the latter category and we shall use the terms "open system" and "leaky cavity" interchangeably. Open systems are by far the more common, although when the leakage is small, concepts pertaining to closed systems can often be applied with a high degree of accuracy.

The modes of a leaky cavity have complex frequency eigenvalues ω, with $\gamma = -2 \operatorname{Im}\omega > 0$ describing the rate of leakage. Secondly, the eigenfunctions extend over the whole universe, so that any normalization integral is dominated by the region outside the cavity. In fact eigenfunctions are largest at infinity, because a spherical outgoing wave $\exp[-i\omega(t - r/c)]/r$ behaves as $\exp(+\gamma r/2c)/r$ when ω has an imaginary part. For these quasinormal modes, many familiar concepts derived for the normal modes of a Hermitian system do not apply.

In some circumstances, it is useful (indeed necessary) to consider not just the leaky cavity (size a), but the whole universe (size Λ, $\Lambda \rightarrow \infty$). By imposing suitable conditions at the boundary of the universe, the system as a whole becomes Hermitian. Then instead of the quasinormal modes of the cavity [for one dimension, spaced by $\sim c/a$ in frequency and normalized to $O(a^{-1/2})$] one discusses the modes of the universe [spaced by $\sim c/\Lambda$ in frequency and normalized to $O(\Lambda^{-1/2})$]. Each quasinormal mode of the cavity corresponds to a collection of modes of the universe and the ability to switch between these two points of view is often useful.[1,2] The latter description is often convenient because the standard techniques for Hermitian systems allow second quantization for dealing with emission, absorption, and lasing.[1-5] Nevertheless, for some applications, the concept of quasinormal modes is physically appealing and mathematically convenient, and we shall adopt the quasinormal mode point of view in this paper.

Quasinormal modes are relevant not only in optics, but whenever there is leakage or decay, for example in α decay and the associated quantum-mechanical tunneling problems,[6] in compound nucleus theory,[7] and even in the wave function of the universe.[8]

We are here concerned with the following question: Suppose the dielectric constant $\epsilon_0(\mathbf{r})$ is slightly perturbed by $\epsilon_1(\mathbf{r})$, what is the first-order change in the (complex) frequency eigenvalues? This mathematical problem is relevant to many physical situations: a small displacement of a dielectric lens, a temperature change affecting the dielectric constant, or nonlinear changes of the dielectric constant at high fields. We shall be particularly interested in the quasinormal modes of dielectric microdroplet, to be discussed in Secs. I B and III.

The usual formalism for Hermitian systems gives the first-order result

$$\frac{\Delta\omega}{\omega} = -\frac{1}{2} \frac{\int dV \, \epsilon_1(\mathbf{r}) |\mathbf{E}^{(0)}(\mathbf{r})|^2}{\int dV \, \epsilon_0(\mathbf{r}) |\mathbf{E}^{(0)}(\mathbf{r})|^2} \qquad (1.1)$$

where $\mathbf{E}^{(0)}$ is the electric field of the unperturbed mode. This formula does not make sense for an open system: the denominator is dominated by the region outside the leaky cavity, and diverges with the size of the universe Λ, while the numerator is independent of Λ since $\epsilon_1(\mathbf{r})$ is nonzero only in (or near) the cavity. The right-hand side, if naively adopted, would give zero in the limit $\Lambda \rightarrow \infty$. This is a ubiquitous problem with quasinormal modes, and the necessary reformulation of time-independent perturbation theory for open systems has been given by Zeldovich[9] for a scalar field in one dimension described by the Schrödinger equation. In Sec. II we extend the Zeldovich formalism to the Maxwell theory in three dimensions, including the case with degeneracy. The end result is a formula similar to (1.1), but where the denominator is expressed in terms of a volume plus a surface integral, such that its value is independent of the size of the universe, i.e., independent of the volume over which the integral is taken, provided it is sufficiently large. One interesting feature is that this formula involves the expression $\mathbf{E}^{(0)} \cdot \mathbf{E}^{(0)}$ and not $\mathbf{E}^{(0)*} \cdot \mathbf{E}^{(0)}$.

B. Quasinormal modes in microdroplets

We shall apply this formalism to the quasinormal modes in dielectric microdroplets, usually referred to as morphology-dependent resonances (MDR's), which are the subject of considerable recent interest and have a pronounced effect on the Mie scattering,[10] fluorescence,[11] Raman scattering,[12] Brillouin scattering,[13] lasing,[14] and chemical energy transfer[15] in these microdroplets, as well as on some aspects of rainbow formation.[16] We shall in particular consider distortions of the droplet shape, which may be induced by an external impulse, generated by intense acoustic waves excited through stimulated Brillouin scattering or simply caused by thermal fluctuations, and have been observed in several experiments.[17,18] The shape distortions, though strictly speaking time dependent, have typical frequencies only in the MHz range, and may be dealt with as static perturbations. The perturbation of these quasinormal modes has been investigated mathematically through a direct calculation of the elastic scattering cross section from a nonspherical scatterer by the T-matrix method[17,19] and one object of this paper is to solve this problem analytically by developing the perturbation theory for quasinormal modes. For readers interested in this application rather than in the formalism as such, we present below some of the results, with further details to be found in Sec. III.

A droplet of radius a (typically $5-50$ μm) is usually described by the size parameter $x = 2\pi a / \lambda$, where λ is the wavelength of light in vacuum. Consider a set of quasinormal modes with polarization μ ($=E$ or M for TE or TM modes), angular momentum l, and radial mode index ν giving the number of nodes of the wave function inside the droplet. These modes can be very narrow if $l \sim nx$,[20] where n is the refractive index; in geometric optics

language, the rays are trapped by total internal reflection around the rim at near-glancing angles. Now let the shape of the droplet be distorted to

$$r(\theta, \varphi) = a + \Delta\sqrt{4\pi} Y_{L,M}(\theta, \varphi) \qquad (1.2)$$

where $\Delta \ll a$, with the refractive index inside the droplet remaining unchanged. (The factor $\sqrt{4\pi}$ is introduced so that for $L = 0$ this describes a uniform expansion of the droplet by an amount Δ. Of course hydrostatic expansion is suppressed by the bulk modulus, and even to the extent that it is allowed, would cause a reduction in the density and hence the refractive index. Nevertheless the hypothetical uniform expansion will provide a useful way to understand some features of the final result.) For $(L, M) = (2, 0)$ and $\Delta / a \ll 1$, the distorted shape is a spheroid, with polar and equatorial radii r_p, r_e, and it is often convenient to express the amplitude of distortion via

$$e \equiv \frac{r_p - r_e}{a} = 3\sqrt{5}/4 \frac{\Delta}{a} . \qquad (1.3)$$

The $2l + 1$ modes can be labeled by an index $\alpha = -l, \dots, l$. For simplicity we assume that the perturbation maintains axial symmetry, i.e., $M = 0$, then α is identical with the azimuthal quantum number m; otherwise each mode α is a linear combination of different m's. The change in the mode frequency can be written as

$$\frac{\Delta\omega}{\omega} = -\frac{\Delta}{a} F(LM; \mu l \nu \alpha; n, x) \qquad (1.4)$$

where the right-hand side could in principle depend on the angular momentum L, M of the perturbation, the mode indices, the refractive index n, and the size parameter x. For a hydrostatic deformation ($L = 0$), $\Delta > 0$ means all eigenvalues are decreased, which is the reason for the sign convention in (1.4). The dependence on α means that the degeneracy is lifted. The object is to find F, especially for $L \ll l$, which is the case of physical interest. In this paper we consider only axially symmetric perturbations ($M = 0$); generalization is straightforward.

We find some surprising simplifications for F, which are exact for all TE modes and valid up to corrections of $O(L^2/l^2)$ for TM modes: F is zero for odd L, while for even L, F turns out to be independent of n, x, μ, and ν; moreover, the dependence on α can be written as a polynomial in α:[21]

$$F = F(L, l, \alpha) = A(L, l) \sum_{k=0}^{L/2} f_k(L, l) \alpha^{2k} \qquad (1.5)$$

where by definition $f_0 = 1$. Moreover, these coefficients can be evaluated without reference to the radial wave functions, yielding extremely simple analytic results; for example, for axially symmetric quadrupole distortion ($L = 2$) and assuming $l \gg 1$,

$$A(L = 2, l) = \sqrt{5}/4 \quad \text{(independent of } l) ,$$
$$f_1 = -3/[l(l+1)] , \qquad (1.6)$$

i.e.,

$$\frac{\Delta\omega}{\omega} = -\frac{e}{6}\left[1 - \frac{3\alpha^2}{l(l+1)}\right] . \tag{1.7}$$

Complete expressions are given in Sec. III. All coefficients turn out to be always real.

The rest of this paper is organized as follows. Section II presents the general formalism and Sec. III gives the application to microdroplets, with technical details given in the Appendices. The perturbation is compared to numerical data for deformed dielectric spheres obtained by the T-matrix method. The results are discussed in Sec. IV.

II. FORMALISM

A. Perturbation in terms of E

For a system described by a spatially varying but time-independent complex dielectric constant $\epsilon(\mathbf{r})$, the modes with time dependence $\exp(-i\omega t)$ satisfy the time-independent Maxwell equation

$$\nabla\times(\nabla\times\mathbf{E}) - \omega^2\epsilon(\mathbf{r})\mathbf{E} = 0 . \tag{2.1}$$

We assume the dielectric is present only in a region of size $\sim a$, and that $\epsilon(\mathbf{r})\to 1$ at infinity. With the velocity of light in vacuum $=1$, the wave number k at infinity is identical to ω. Mirrors are ignored, though these can be readily incorporated through boundary conditions on (2.1). Quasinormal modes are solutions to (2.1) with outgoing wave boundary conditions, for which ω is complex. Because ω and $\epsilon(\mathbf{r})$ are complex, $\mathbf{E}^*$ does *not* satisfy (2.1).

The unperturbed system with dielectric constant $\epsilon_0(\mathbf{r})$ has a D-fold degenerate set of quasimodes $\mathbf{E}^{(0)\alpha}$, $\alpha = 1, \ldots, D$, satisfying

$$\nabla\times(\nabla\times\mathbf{E}^{(0)\alpha}) - \omega_0^2\epsilon_0(\mathbf{r})\mathbf{E}^{(0)\alpha} = 0 , \tag{2.2}$$

where the unperturbed frequency ω_0 is independent of α.

Now let the dielectric constant be perturbed to

$$\epsilon(\mathbf{r}) = \epsilon_0(\mathbf{r}) + \epsilon_1(\mathbf{r}) , \tag{2.3}$$

where the second term is small in some sense. Denote the electric field of the exact quasinormal modes as

$$\mathbf{F}^\lambda(\mathbf{r}) = \mathbf{F}^{(0)\lambda}(\mathbf{r}) + \mathbf{F}^{(1)\lambda}(\mathbf{r}) . \tag{2.4}$$

The first term is the field when $\epsilon_1\to 0$, and must be a linear combination

$$\mathbf{F}^{(0)\lambda}(\mathbf{r}) = \sum_\alpha a_\alpha^\lambda \mathbf{E}^{(0)\alpha}(\mathbf{r}) , \tag{2.5}$$

which satisfies (2.2) as well. On the other hand the exact fields satisfy

$$\nabla\times(\nabla\times\mathbf{F}^\lambda) - (\omega^\lambda)^2\epsilon(\mathbf{r})\mathbf{F}^\lambda = 0 , \tag{2.6}$$

in which the exact frequencies

$$\omega^\lambda = \omega_0 + \omega_1^\lambda \tag{2.7}$$

are in general dependent on λ, i.e., the degeneracy is lifted. The object is to calculate the frequency shifts ω_1^λ, $\lambda = 1, \ldots, D$ (both real and imaginary parts).

By subtracting (2.2) for $\mathbf{F}^{(0)\lambda}$ from (2.6),

$$\nabla\times(\nabla\times\mathbf{F}^{(1)\lambda}) - \omega_0^2\epsilon_0(\mathbf{r})\mathbf{F}^{(1)\lambda}$$
$$= \omega_0^2\epsilon_1(\mathbf{r})\mathbf{F}^\lambda + 2\omega_0\omega_1^\lambda\epsilon_0(\mathbf{r})\mathbf{F}^\lambda , \tag{2.8}$$

in which second-order terms proportional to ω_1^2 and $\omega_1\epsilon_1(\mathbf{r})$ have been discarded. It might appear that, to first order, $\mathbf{F}^\lambda$ on the right-hand side can be replaced by $\mathbf{F}^{(0)\lambda}$; however, as we shall see, this is not valid in the case of shape perturbations and we continue to write (2.8) in terms of $\mathbf{F}^\lambda$ rather than $\mathbf{F}^{(0)\lambda}$.

Now multiply by $\mathbf{E}^{(0)\alpha}$ (*not* its complex conjugate) and integrate over a sphere of radius R, $R \gg a$, $R \gg \lambda = 2\pi/\omega$. Thus

$$\int_R dV\, \mathbf{E}^{(0)\alpha}\cdot[\nabla\times(\nabla\times\mathbf{F}^{(1)\lambda}) - \omega_0^2\epsilon_0(\mathbf{r})\mathbf{F}^{(1)\lambda}] = \omega_0^2\int_R dV\, \epsilon_1(\mathbf{r})\mathbf{E}^{(0)\alpha}\cdot\mathbf{F}^\lambda + 2\omega_0\omega_1^\lambda\int_R dV\, \epsilon_0(\mathbf{r})\mathbf{E}^{(0)\alpha}\cdot\mathbf{F}^\lambda . \tag{2.9}$$

In fact, (2.9) shows that the perturbation can be effected after doing the integral. Thus the condition is that the discarded terms are small when so integrated, and not that they are small in a pointwise sense. Therefore changes in ϵ of order unity but extending over a small region can be accommodated. On the left-hand side of (2.9), we integrate by parts, obtaining the terms in (2.2), which add to zero. (This is why we must multiply by $\mathbf{E}^{(0)\alpha}$ and not its complex conjugate). However, because the system is non-Hermitian, there are nonzero surface terms, which are evaluated to be

$$-\int_R dS\,[E_i^{(0)\alpha}(\partial_r F_i^{(1)\lambda}) - (\partial_r E_i^{(0)\alpha})F_i^{(1)\lambda}] . \tag{2.10}$$

It is next necessary to eliminate $\mathbf{F}^{(1)\lambda}$ from this expression. In the far zone, the fields are spherical waves, and

$\partial_r(e^{ikr}/r) \sim ik(e^{ikr}/r)$, so

$$\partial_r(F_i^{(0)\lambda} + F_i^{(1)\lambda}) = ik^\lambda(F_i^{(0)\lambda} + F_i^{(1)\lambda}) , \tag{2.11a}$$

while for the zeroth-order field,

$$\partial_r F_i^{(0)\lambda} = ik_0 F_i^{(0)\lambda} . \tag{2.11b}$$

Taking the difference and neglecting a second-order term $k_1^\lambda F_i^{(1)\lambda}$, we find that $\mathbf{F}^{(1)\lambda}$ drops out completely and (2.10) can be expressed as

$$i\omega_1^\lambda\int_R dS\, \mathbf{E}^{(0)\alpha}\cdot\mathbf{F}^{(0)\lambda} . \tag{2.12}$$

Putting this into (2.9) gives

$$\omega_1^\lambda \left[2\omega_0 \int_R dV\, \epsilon_0(\mathbf{r}) \mathbf{E}^{(0)\alpha} \cdot \mathbf{F}^\lambda + i \int_R dS\, \mathbf{E}^{(0)\alpha} \cdot \mathbf{F}^{(0)\lambda} \right]$$

$$= -\omega_0^2 \int_R dV\, \epsilon_1(\mathbf{r}) \mathbf{E}^{(0)\alpha} \cdot \mathbf{F}^\lambda \ . \qquad (2.13)$$

Since ω_1^λ is a small quantity, we may replace $\mathbf{F}^\lambda$ on the left-hand side by $\mathbf{F}^{(0)\lambda}$. For the moment also assume ϵ_1 is also everywhere small, then $\mathbf{F}^\lambda$ on the right-hand side can also be replaced by $\mathbf{F}^{(0)\lambda}$. Recalling the definition (2.5), we get the eigenvalue equation

$$\sum_\beta V^{\alpha\beta} a_\beta^\lambda = - \left[\frac{2\omega_1^\lambda}{\omega_0} \right] \sum_\beta G^{\alpha\beta} a_\beta^\lambda \qquad (2.14)$$

for the λth eigenvalue $-2\omega_1^\lambda/\omega_0$ and the λth eigenvector $\mathbf{a}^\lambda = (a_1^\lambda, a_2^\lambda, \ldots, a_D^\lambda)$. The $D \times D$ matrices are

$$V^{\alpha\beta} = \int_R dV\, \epsilon_1(\mathbf{r}) \mathbf{E}^{(0)\alpha} \cdot \mathbf{E}^{(0)\beta} \ , \qquad (2.15)$$

$$G^{\alpha\beta} = \int_R dV\, \epsilon_0(\mathbf{r}) \mathbf{E}^{(0)\alpha} \cdot \mathbf{E}^{(0)\beta} + \frac{i}{2\omega_0} \int_R dS\, \mathbf{E}^{(0)\alpha} \cdot \mathbf{E}^{(0)\beta}$$

$$(2.16)$$

depending only on the unperturbed eigenfunctions and eigenvalue. This is then the formal solution of the time-independent perturbation. In the nondegenerate case, $V^{\alpha\beta}$ and $G^{\alpha\beta}$ are the analogs of the numerator and the denominators in (1.1), respectively.

Since R is an arbitrary large distances, we must verify that $V^{\alpha\beta}$ and $G^{\alpha\beta}$ are independent of R; this is nontrivial since the fields extend to infinity. The case of $V^{\alpha\beta}$ is simple, because $\epsilon_1(\mathbf{r})$ is nonzero only in (or near) the cavity of size $\sim a$, so any $R \gg a$ will give the same $V^{\alpha\beta}$. For $G^{\alpha\beta}$, we note that

$$\frac{dG^{\alpha\beta}}{dR} = \int d\Omega \left[1 + \frac{i}{2\omega_0} \frac{d}{dR} \right] g^{\alpha\beta}(R) \ , \qquad (2.17)$$

where we have used $\epsilon_0(R) = 1$ for $R \gg a$ and

$$g^{\alpha\beta}(R) = R^2 \mathbf{E}^{(0)\alpha}(R, \theta, \varphi) \cdot \mathbf{E}^{(0)\beta}(R, \theta, \varphi) \ . \qquad (2.18)$$

But at large distance the fields are outgoing waves:

$$R\, \mathbf{E}^{(0)\alpha}(R, \theta, \varphi) = \mathbf{a}^\alpha(\theta, \varphi) e^{ik_0 R} \ , \qquad (2.19)$$

so that d/dR acting on each $\mathbf{E}^{(0)}$ factor gives ik_0. Hence $dG/dR = 0$ and thus (2.14) has an unambiguous meaning independent of R, thus solving the problem associated with (1.1) as discussed in the Introduction. The independence of R will be verified explicitly in Appendix A for the example of microdroplets with shape perturbation. This important property relies on two features of $G^{\alpha\beta}$: (a) $\mathbf{E}^{(0)} \cdot \mathbf{E}^{(0)}$ is involved rather than $\mathbf{E}^{(0)*} \cdot \mathbf{E}^{(0)}$, and (b) there is a surface term.

The present formalism can be applied to closed systems with real $\epsilon(\mathbf{r})$ as well. In that case, the wave function vanishes at infinity, so the second term in (2.16) is zero for large R, while the first term converges. The field $\mathbf{E}^{(0)}$ can be chosen to be real, and $\mathbf{E}^{(0)} \cdot \mathbf{E}^{(0)}$ need not be distinguished from $\mathbf{E}^{(0)*} \cdot \mathbf{E}^{(0)}$. In that case (2.14) would be just a trivial degenerate generalization of (1.1).

The corrections to the first-order perturbation result

will be discussed in the context of microdroplets in Sec. III. These corrections are such that the formal result (a) is accurate for the shifts in the real parts, but (b) gives only an order of magnitude estimate for the changes in the imaginary parts (widths).

B. Perturbation in terms of B

In some cases, it may be more convenient to express $G^{\alpha\beta}$ in terms of the magnetic field $\mathbf{B}^{(0)\alpha}$, corresponding to the electric field $\mathbf{E}^{(0)\alpha}$. In the volume integral in (2.16), we use the Maxwell equation

$$\epsilon_0(\mathbf{r}) \mathbf{E}^{(0)\alpha} = \frac{i}{\omega_0} \nabla \times \mathbf{B}^{(0)\alpha} \ , \qquad (2.20)$$

integrate by parts, and convert the resultant $\nabla \times \mathbf{E}^{(0)\beta}$ into

$$\nabla \times \mathbf{E}^{(0)\beta} = i\omega_0 \mathbf{B}^{(0)\beta} \ . \qquad (2.21)$$

Again because the system is not Hermitian, there is in addition a surface term:

$$\int_R dV\, \epsilon_0(\mathbf{r}) \mathbf{E}^{(0)\alpha} \cdot \mathbf{E}^{(0)\beta}$$

$$= -\int_R dV\, \mathbf{B}^{(0)\alpha} \cdot \mathbf{B}^{(0)\beta} - \frac{i}{\omega_0} \int_R dS\, \hat{\mathbf{n}} \cdot (\mathbf{E}^{(0)\beta} \times \mathbf{B}^{(0)\alpha}) \ .$$

$$(2.22)$$

This resembles the familiar statement that, except for surface terms, the electric and magnetic energies are equal—except that there is now a relative minus sign in the volume integrals. The reason is that in the "usual" case one of the field factors is complex conjugated, so that one of the factors of ω_0 in (2.20) and (2.21) would acquire a minus sign.

Since all fields at infinity are spherical outgoing waves, we further have

$$\hat{\mathbf{n}} \cdot (\mathbf{E}^{(0)\beta} \times \mathbf{B}^{(0)\alpha}) = \mathbf{E}^{(0)\beta} \cdot \mathbf{E}^{(0)\alpha} = \mathbf{B}^{(0)\beta} \cdot \mathbf{B}^{(0)\alpha} \qquad (2.23)$$

and we may write $G^{\alpha\beta}$ as

$$G^{\alpha\beta} = -\int_R dV\, \mathbf{B}^{(0)\alpha} \cdot \mathbf{B}^{(0)\beta} - \frac{i}{2\omega_0} \int_R dS\, \mathbf{B}^{(0)\alpha} \cdot \mathbf{B}^{(0)\beta} \ .$$

$$(2.24)$$

C. Linear transformation and adjoint field

It will be convenient to take linear combinations of (2.14) and write

$$\sum_\beta \overline{V}^{\alpha\beta} a_\beta^\lambda = - \left[\frac{2\omega_1^\lambda}{\omega_0} \right] \sum_\beta \overline{G}^{\alpha\beta} a_\beta^\lambda \qquad (2.25)$$

where

$$\overline{V}^{\alpha\beta} = \sum_\gamma c_\gamma^\alpha V^{\gamma\beta} \ ,$$

$$\overline{G}^{\alpha\beta} = \sum_\gamma c_\gamma^\alpha G^{\gamma\beta} \ , \qquad (2.26)$$

and the matrix c_γ^α is left unspecified at the moment. This transformation can be equivalently expressed if we define *adjoint* fields

$$\mathbf{E}^{(0)\alpha\dagger} = \sum_\gamma c_\gamma^\alpha \mathbf{E}^{(0)\gamma} \tag{2.27}$$

(and similarly for **B** if necessary), which are defined *without* complex conjugation. Then

$$\overline{V}^{\alpha\beta} = \int_R dV \, \epsilon_1(\mathbf{r}) \mathbf{E}^{(0)\alpha\dagger} \cdot \mathbf{E}^{(0)\beta} \, , \tag{2.28}$$

$$\overline{G}^{\alpha\beta} = \int_R dV \, \epsilon_0(\mathbf{r}) \mathbf{E}^{(0)\alpha\dagger} \cdot \mathbf{E}^{(0)\beta}$$
$$+ \frac{i}{2\omega_0} \int_R dS \, \mathbf{E}^{(0)\alpha\dagger} \cdot \mathbf{E}^{(0)\beta} \, . \tag{2.29}$$

The adjoint fields will be convenient for the following reason. In the D-dimensional vector space spanned by $\{\mathbf{E}^{(0)\alpha}\}$, there will be a natural inner product, which, in general, will *not* be integrals involving $\mathbf{E}^{(0)\alpha} \cdot \mathbf{E}^{(0)\beta}$. By choosing c_γ^α, we can often make the inner product coincide with integrals involving $\mathbf{E}^{(0)\alpha\dagger} \cdot \mathbf{E}^{(0)\beta}$, which will make (2.25) more natural in terms of the vector space structure. The ideal choice of c_γ^α will obviously be proportional to the inverse of $G^{\alpha\beta}$, so that $\overline{G}^{\alpha\beta}$ is proportional to the identity. These remarks will become clear in the context of the examples in Sec. III.

D. Shape perturbation

The formalism requires a slight refinement in the case of shape perturbations. To be specific, consider a dielectric body B with refractive index n placed in vacuum. Use a curvilinear coordinate system (u,ξ,η) such that the surface of B is described by $u(\xi,\eta)=a$, where a is a constant, and the interior (exterior) is given by $u < a$ $(u > a)$. For a spherical body, (u,ξ,η) is identified with polar coordinates (r,θ,φ). The dielectric constant is

$$\epsilon_0(\mathbf{r}) = 1 + (n^2 - 1)\Theta(a - u) \tag{2.30}$$

where Θ is the unit step function. Now let the boundary be perturbed to

$$u(\xi,\eta) = a + \Delta\sqrt{4\pi}Y(\xi,\eta) \tag{2.31}$$

where Y is some function of $O(1)$ and the $\sqrt{4\pi}$ factor follows the convention in (1.2). Then the change in dielectric constant can be written to first order as

$$\epsilon_1(\mathbf{r}) = (n^2 - 1)\Delta\delta(u - a)\sqrt{4\pi}Y(\xi,\eta) \, . \tag{2.32}$$

In this case ϵ_1 is *not* everywhere small and in fact the function $\Delta\delta(u - a)$ represents a unit dielectric fluctuation in a thin layer of thickness Δ. Now if we put (2.32) into (2.15), the integral is ambiguous because the δ function in ϵ_1 multiplies the *discontinuous* function $\mathbf{E}^{(0)\alpha\dagger} \cdot \mathbf{E}^{(0)\beta}$. The problem can be traced back to (2.13), where, on the right-hand side, we replaced $\mathbf{F}^\lambda$ by $\mathbf{F}^{(0)\lambda}$. This is justified only if $F^{(1)\lambda}$ is small.

In the present case, let $\mathbf{r}$ be a point just outside the surface, such that under the shape perturbation, it is just inside the surface. To lowest order in Δ, the fields at $\mathbf{r}$ will simply change from the "outside" value to the "inside"

value. This change is small for the *tangential* (t) component of **E** and for the *normal* (n) component of $\mathbf{D} = \epsilon\mathbf{E}$. Write $\mathbf{F}^\lambda$ as $\mathbf{E}^\lambda$ and express the integrand on the right-hand side of (2.13) as

$$\epsilon_1 \mathbf{E}_t^{(0)\alpha} \cdot \mathbf{E}_t^\lambda + (\epsilon - \epsilon_0) \frac{D_n^{(0)\alpha}}{\epsilon_0} \frac{D_n^\lambda}{\epsilon} \, . \tag{2.33}$$

Now $\mathbf{E}_t^\lambda$ and D_n^λ, because of their continuity across the surface, can be replaced by their zeroth-order values, and we get

$$\epsilon_1 \mathbf{E}_t^{(0)\alpha} \cdot \mathbf{E}_t^{(0)\lambda} + \left[\frac{1}{\epsilon_0} - \frac{1}{\epsilon} \right] D_n^{(0)\alpha} D_n^{(0)\lambda} \, . \tag{2.34}$$

Furthermore, in analogy to (2.32)

$$\frac{1}{\epsilon_0} - \frac{1}{\epsilon} = \left[1 - \frac{1}{n^2} \right] \Delta\delta(r - a)\sqrt{4\pi}Y(\xi,\eta)$$
$$= \frac{1}{n^2}\epsilon_1(\mathbf{r}) \, , \tag{2.35}$$

while

$$D_n^{(0)\alpha} = n^2 E_n^{(0)\alpha}(\text{in}) \, , \tag{2.36}$$

where the right-hand side indicates that the field is to be evaluated on the inside surface. Thus

$$V^{\alpha\beta} = (n^2 - 1)\Delta\sqrt{4\pi} \int dS \, Y(\xi,\eta)$$
$$\times (\mathbf{E}_t^{(0)\alpha} \cdot \mathbf{E}_t^{(0)\beta} + n^2 E_n^{(0)\alpha} E_n^{(0)\beta}) \, , \tag{2.37}$$

the integral being taken on the *inside* of the surface. The particular combination, with the normal component enhanced by a factor n^2, is always found in these circumstances.[22] The same applies to the transformed matrix $\overline{V}^{\alpha\beta}$. The integrals in $G^{\alpha\beta}$ are not affected.

For a closed cavity whose walls have a finite conductivity, which is a non-Hermitian system, a rather similar formulation has been given.[23]

III. MORPHOLOGY-DEPENDENT RESONANCES IN MICRODROPLETS

The formalism in the preceding section permits the frequency shifts ω_1^λ to be evaluated in terms of the unperturbed mode functions. In practice however, the modes of ordinary laser cavities are extremely difficult to calculate accurately,[24] even in the absence of perturbations. Microdroplets, quite apart from their experimental interest,[10–16,23] offer an example where the unperturbed mode functions are readily calculable because of the spherical symmetry, so that the frequency shifts can be evaluated explicitly. Our notations are defined in Sec. I B and the refractive index n is taken to be real, although complex values can be readily handled. In this paper we shall deal with axially symmetric perturbations $(M = 0)$. It is not difficult to generalize to other cases.

A. TE modes

The TE modes are given by

$$\mathbf{E}^{(0)\alpha}(\mathbf{r}) = \phi(r)\mathbf{X}_{l,\alpha}(\theta,\varphi)$$ (3.1)

where

$$\mathbf{X}_{l,\alpha} = \frac{1}{\sqrt{l(l+1)}}\hat{\mathbf{L}}Y_{l,\alpha}, \quad \hat{\mathbf{L}} = \mathbf{r}\times(-i\nabla)$$ (3.2)

are the vector spherical harmonics, and the radial function depends on l, n, and the size parameter $x = k_0 a$. Define the adjoint fields as

$$\mathbf{E}^{(0)\alpha\dagger}(\mathbf{r}) = \phi(r)\mathbf{X}_{l,\alpha}^*(\theta,\varphi) ,$$ (3.3a)

i.e.,

$$c_\gamma^\alpha = (-1)^{\alpha+1}\delta_{\gamma,-\alpha} .$$ (3.3b)

Thus the adjoint operator simply means that the angular wave function but not the radial wave function is conjugated. Then $\bar{G}^{\alpha\beta}$ is proportional to the indentity

$$\bar{G}^{\alpha\beta} = \delta^{\alpha\beta}\left[\int_0^R dr\, r^2\epsilon_0(r)\phi(r)^2 + \frac{i}{2\omega_0}R^2\phi(R)^2\right]$$

$$\equiv \delta^{\alpha\beta}G .$$ (3.4)

The fact that this is independent of R is shown explicitly in Appendix A.

Because the perturbation has $M = 0$, α remains a good quantum number, and $\bar{V}^{\alpha\beta}$ is diagonal. Thus the eigenvalue equation (2.26) is readily solved to give

$$\frac{\omega_1^\alpha}{\omega_0} = -\frac{1}{2}\frac{\bar{V}^{\alpha\alpha}}{G} = -\frac{\Delta}{a}I_R F$$ (3.5)

where on the right-hand side we have extracted a factor Δ/a and separated into radial and angular integrals

$$I_R(n,x,l,v) = (2G)^{-1}\int dr\, r^2[(n^2-1)a\delta(r-a)]\phi(r)^2$$

$$= (2G)^{-1}(n^2-1)a^3\phi(a)^2 ,$$ (3.6)

G being given by (3.4), and

$$F(L,l,\alpha) = \sqrt{4\pi}\int d\Omega\, \mathbf{X}_{l,\alpha}^*\cdot Y_{L,0}\mathbf{X}_{l,\alpha} .$$ (3.7)

We have explicitly indicated the dependence of I_R and F on the various parameters; in particular I_R is independent of L.

Now specialize to a hydrostatic perturbation with $L = 0$; then from (3.7) $F = 1$. Such a perturbation maintains spherical symmetry and for a sphere the resonance occurs at a given value of $x = \omega a$, so

$$\frac{\omega_1^\alpha}{\omega_0} = \frac{\Delta\omega}{\omega} = -\frac{\Delta}{a} .$$

Hence we conclude that $I_R = 1$. But I_R is independent of L, so this result derived for $L = 0$ must be universally valid. There is then never any need to evaluate radial functions and the right-hand side of (3.5) is independent of n, x, and v. Since this result is somewhat unexpected, we provide an explicit demonstration in Appendix B. Of course, $I_R = 1$ only for those combinations of (n,x,l,v) which define a resonance.

Thus the frequency shift is given by (1.4) and it remains to do the angular integral F, which is zero unless L is even and can in general be evaluated through Clebsch-Gordan coefficients:[25,26]

$$F(L,l,\alpha) = A(L,l)f(\alpha)$$ (3.8)

where

$$A(L,l) = \frac{2l+1}{\sqrt{2L+1}}[C(llL;000)]^2\left[1 - \frac{L(L+1)}{2l(l+1)}\right]$$ (3.9)

and the Clebsch-Gordan coefficients are given by

$$C(llL;000) = (-1)^{l-L/2}\left[\frac{2L+1}{2l+L+1}\right]^{1/2}$$

$$\times \frac{\tau(2l+L)}{\tau(2l-L)}\frac{1}{\tau(L)^2} ,$$ (3.10)

$$\tau(x) = (x/2)!/\sqrt{x!} .$$ (3.11)

The α dependence enters through

$$f(\alpha) = (-1)^\alpha C(llL;\alpha,-\alpha,0)/C(llL;000)$$ (3.12)

whose dependence on L and l is suppressed and which may be obtained through the recursion relations

$$f(0) = 1, f(1) = 1 - \frac{L(L+1)}{2l(l+1)},$$

$$f(\alpha+1) = \{[2l(l+1)-2\alpha^2-L(L+1)]f(\alpha)-(l-\alpha+1)(l+\alpha)f(\alpha-1)\}/[(l+\alpha+1)(l-\alpha)] .$$ (3.13)

While this gives a complete characterization, a convenient alternative form for small L is

$$f(\alpha) = \sum_{k=0}^{L/2} f_k\alpha^{2k} .$$ (3.14)

[Since $f(\alpha)$ is defined only for $2l+1$ discrete values of α,

it can always be represented by a polynomial of degree up to $2l$. The above formula is nontrivial in that only terms up to α^L appear. In practice, one can use (3.13) to find $f(0), f(1), \ldots, f(L)$, from which $f_0, f_1, \ldots, f_L$ can be determined, and hence all $f(\alpha)$ can be simply evaluated from (3.14).] Explicitly for $L = 2$

$$A(L=2,l)=4\sqrt{5}\frac{[l(l+1)]^2}{(2l-1)2l(2l+2)(2l+3)}$$

$$\times\left[1-\frac{3}{l(l+1)}\right] \qquad (3.15)$$

$$f_0=1, \quad f_1=-3/[l(l+1)] .$$

Details of these calculation are given in Appendix C.

B. TM modes

In this case the magnetic fields are given by

$$\mathbf{B}^{(0)\alpha}(\mathbf{r})=\phi(r)\mathbf{X}_{l,\alpha}(\theta,\varphi) \qquad (3.16)$$

so using the form (2.25)

$$\bar{G}^{\alpha\beta}=\delta^{\alpha\beta}\left[-\int_0^R dr\, r^2\phi(r)^2-\frac{i}{2\omega_0}R^2\phi(R)^2\right]$$

$$\equiv\delta^{\alpha\beta}G' . \qquad (3.17)$$

Now the electric field is

$$\mathbf{E}^{(0)\alpha}(\mathbf{r})=\frac{i}{\omega_0\epsilon_0(\mathbf{r})}\nabla\times\mathbf{B}^{(0)\alpha} \qquad (3.18)$$

and we only need $\mathbf{E}$ just inside the surface

$$\mathbf{E}^{(0)\alpha}(r=a^-,\theta,\varphi)=\frac{i}{\omega_0 n^2}\left[i\frac{\phi(a)}{a}\sqrt{l(l+1)}Y_{l,\alpha}\hat{\mathbf{n}}\right.$$

$$\left.+\frac{1}{r}\frac{\partial(r\phi)}{\partial r}\bigg|_a \hat{\mathbf{n}}\times\mathbf{X}_{l,\alpha}\right] .$$

$$(3.19)$$

Thus putting this into (2.37)

$$\bar{V}^{\alpha\alpha}=-\frac{(n^2-1)\Delta}{\omega_0^2 n^4}\left[n^2[\phi(a)]^2 l(l+1)F_1'\right.$$

$$\left.+\left[\frac{\partial(r\phi)}{\partial r}\right]_a^2 F_2'\right] . \qquad (3.20)$$

Note that under the adjoint operation, $\mathbf{X}_{l,\alpha}$ goes into $\mathbf{X}_{l,\alpha}^*$, but $Y_{l,\alpha}$ goes into $-Y_{l,\alpha}^*$. The angular integrals are

$$F_1'=\sqrt{4\pi}\int d\Omega\, Y_{l,\alpha}^* Y_{L,0}Y_{l,\alpha} ,$$

$$F_2'=\sqrt{4\pi}\int d\Omega(\hat{\mathbf{n}}\times\mathbf{X}_{l,\alpha})^*\cdot Y_{L,0}(\hat{\mathbf{n}}\times\mathbf{X}_{l,\alpha}) . \qquad (3.21)$$

It is known that

$$F_1'=F/\cos\theta, \quad F_2'=F \qquad (3.22)$$

where θ is the angle between $\mathbf{l}_1$ and $\mathbf{l}_2$ in the classical vector addition $\mathbf{l}_1+\mathbf{L}=\mathbf{l}_2$, with $|\mathbf{l}_1|=|\mathbf{l}_2|=l$:

$$\cos\theta=1-\frac{L(L+1)}{2l(l+1)} . \qquad (3.23)$$

We shall assume for simplicity that $L\ll l$, so all the angular integrals are equal, and we can write the frequency shifts as in (3.5), where

$$I_R(n,x,l,\nu)=-(2G')^{-1}\frac{(n^2-1)a^3}{x^2n^4}$$

$$\times\left[n^2\phi(a)^2 l(l+1)+\left[\frac{\partial(r\phi)}{\partial r}\right]_a^2\right] \qquad (3.24$$

and G' is now given by (3.17), while F is the same as (3.7). By exactly the same argument as before, we see that $I_R=1$ independent of the radial function; this is again verified explicitly in Appendix A. Thus all the results for TE modes carry over to TM modes as well. The $O(L^2/l^2)$ corrections in the TM case, which we have neglected, are purely real up to $O(\mathrm{Im}k_0/\mathrm{Re}k_0)=O(Q^{-1})$.

C. Comparison with numerical calculation and error estimates

The problem of the scattering of electromagnetic waves from a dielectric body can be formulated via the T-matrix method;[17,19] the spherical wave basis is of course particularly convenient for slightly deformed droplets. We have calculated the scattering in this manner for a droplet with real refractive index $n=2$ and size parameter $x\sim6.7$; several cases have been considered and three typical ones are given in Table I. Because the shape perturbation maintains axial symmetry ($M=0$), each sector with a definite m can be treated separately. The shifted position of the resonance is found by stepping through the size parameter.

The calculated fractional shifts $\Delta\omega/\omega$ are plotted against m^2 in Fig. 1 (points) and compared with the first-order perturbation result presented in this paper (lines); here ω and $\Delta\omega$ refer to the real parts only. By (3.14), the theoretical curve is a polynomial of order $L/2$ in m^2. The agreement shown in Fig. 1 is excellent.

Since F in (1.4) is real, the imaginary part of ω should satisfy the same relation, namely,

$$\frac{\Delta\gamma}{\gamma}=\frac{\gamma}{\gamma_0}-1=-\frac{\Delta}{a}F \qquad (3.25)$$

where γ_0 and γ are the widths of the resonance in the unperturbed and the perturbed spheres. In the example of

TABLE I. The parameters of the resonant mode and of the shape perturbation for the three cases considered.

	(a)	(b)	(c)
Resonant mode			
Polarization	TE	TM	TE
l	10	9	10
x (for sphere)	6.8263	6.6728	6.8263
Perturbation			
L	2	2	4
M	0	0	0
Δ/a ($\times 10^{-3}$)	-8.94	-8.94	3.66
e	-0.03	-0.03	

case (a), the right-hand side of (3.25) ranges from $\sim 6 \times 10^{-3}$ ($m = 0$) to $\sim -8 \times 10^{-3}$ ($m = l$) [the line in Fig. 1(a)], but numerical results show that $\Delta \gamma / \gamma$ ranges from ~ 0.16 ($m = 0$) to ~ 0.02 ($m = l$); thus there is a substantial fractional disagreement. This is not so surprising since we are calculating the change in the *small* quantity γ (down by a factor of Q from ω). However, most experiments would be sensitive to γ rather than $\Delta \gamma$, and the error is in practice not important.

This comparison leads naturally to a discussion of the sources of error in our result (1.4) for $\Delta \omega / \omega$. First of all, there will be second-order corrections, say $(\Delta / a)^2 H_1 e^{i\beta_1}$, where $H_1 = O(1)$ and in general $\beta_1 \neq 0$. Secondly, the
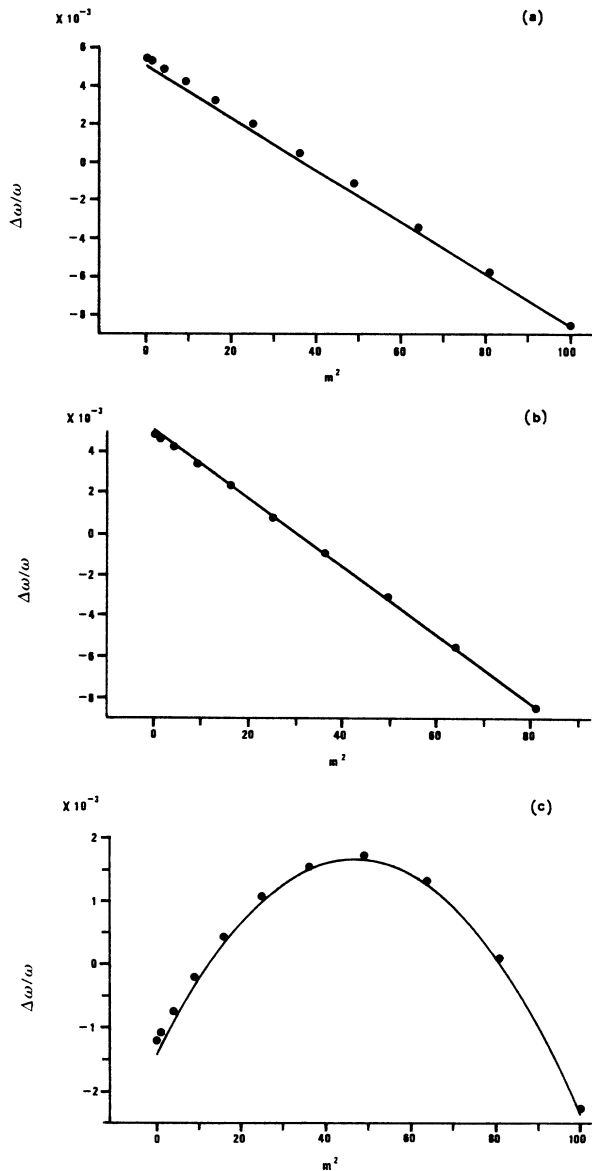

FIG. 1. The fractional frequency shift $\Delta \omega / \omega$ vs m^2, where m is the azimuthal quantum number of the quasinormal mode. Points are numerical results using T-matrix methods, for a droplet with refractive index $n = 2$. Lines are the first-order perturbation results given in this paper. The parameters for the three cases are as in Table I.

spherical wave approximation (2.19) contains relative errors in E of the form

$$\frac{\lambda}{R} e^{ik_0 R} . \tag{3.26}$$

Recalling that k_0 has an imaginary part, we see that R should be chosen to be

$$R \ll \frac{1}{\mathrm{Im}k_0} \sim \frac{Q}{\mathrm{Re}k_0}$$

so that the correction in (3.26) is of order

$$\frac{\lambda}{R} \sim \frac{\lambda(\mathrm{Re}k_0)}{Q} \sim \frac{1}{Q} .$$

Incorporating this correction in F,

$$F \rightarrow F + \frac{1}{Q} H_2 e^{i\beta_2} \tag{3.27}$$

where $H_2 = O(1)$ and in general $\beta_2 \neq 0$.
Putting these together

$$\frac{\Delta \omega}{\omega} = -\frac{\Delta}{a} \left[F + \frac{1}{Q} H_2 e^{i\beta_2} \right] + \left[\frac{\Delta}{a} \right]^2 H_1 e^{i\beta_1} . \tag{3.28}$$

The conclusion is that the *first-order result* $-(\Delta / a)F$ *contains relative errors of order* Δ / a *and* $1/Q$, in contrast to Hermitian systems, where the only corrections are of relative order Δ / a. (Of course, the Hermitian case may be regarded as the $Q \rightarrow \infty$ limit of the general non-Hermitian case.) In the examples given here, both $1/Q$ and Δ / a are at the $10^{-3} - 10^{-2}$ level, which accounts for the small remaining discrepancy in the real parts in Fig. 1.

The form of (3.28) implies that the error in the first-order calculation

$$D \equiv \left[\frac{\Delta \omega}{\omega} \right]_{\text{true}} - \left[\frac{\Delta \omega}{\omega} \right]_{\text{theory}} \tag{3.29}$$

where ω and $\Delta \omega$ refer only to the real parts, takes the form

$$D = d_1 \left[\frac{\Delta}{a} \right] + d_2 \left[\frac{\Delta}{a} \right]^2 \tag{3.30}$$

where

$$d_1 = -\frac{1}{Q} H_2 \cos\beta_2 ,$$

$$d_2 = H_1 \cos\beta_1$$

in general depend on m. The peculiar feature is the existence of $d_1 \neq 0$. To verify this, we have taken case (a) in Table I and varied Δ / a to calculate $(\Delta \omega / \omega)_{\text{true}}$ numerically. Figure 2 shows the calculated D versus Δ / a for the $m = 0$ mode (points) and a fit to a quadratic function as in (3.30) (line). The fit gives $d_1 = 0.015$; for all m's, we find that $d_1 \neq 0$ in general. This comparison thus verifies the existence of a correction of the $(\Delta / a)(1/Q)$ type.

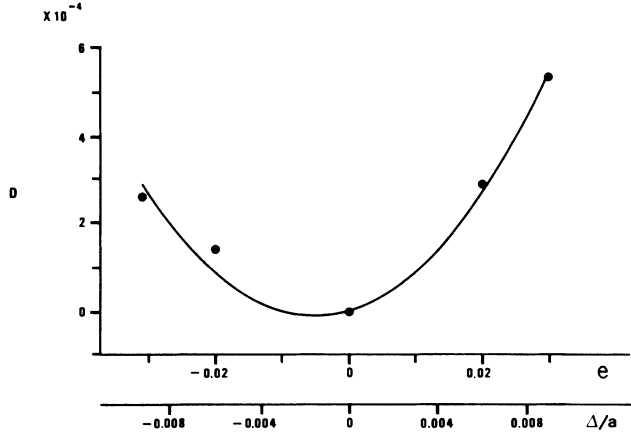On account of the $(\Delta / a)(1/Q)$ correction, the imaginary part is more subtle, and we begin by examining the

FIG. 2. The remaining error D in $\Delta\omega/\omega$ as defined in (3.29) for the $m = 0$ mode of case (a) (but with different values of Δ/a), vs Δ/a or e (points). The line is a fit to a quadratic function (3.30). The small portion of the graph below the horizontal axis indicates a relatively small correction going like Δ/a.

phases β_1, β_2. As $Q \to \infty$, the system becomes Hermitian and the frequency shifts must be real. This shows $\beta_1 \to 0$, i.e., $\beta_1 = O(1/Q)$; however, no conclusion can be drawn for β_2, so we must assume $\beta_2 = O(1)$. Now if we extract the imaginary part from (3.28),

$$\frac{\gamma}{\gamma_0} - 1 = -\frac{\Delta}{a}\left[F + \frac{1}{Q}H_2\cos\beta_2\right] + \left[\frac{\Delta}{a}\right]^2 H_1\cos\beta_1$$

$$-\frac{2\omega_0}{\gamma_0}\left[-\frac{\Delta}{a}\frac{1}{Q}H_2\sin\beta_2 + \left[\frac{\Delta}{a}\right]^2 H_1\sin\beta_1\right].$$

$$(3.31)$$

The last two terms, containing the factor $2\omega_0/\gamma_0$, reflects the mixing of the real and imaginary parts due to the perturbation. The last term creates no problems, since $\sin\beta_1 \sim \beta_1 \sim 1/Q$ cancels $2\omega_0/\gamma_0$, but since $\beta_2 = O(1)$, the other term is of order $(\Delta/a)H_2$ *without any factor of $1/Q$*. Absorbing some constants into H_2, we write (3.31) schematically as

$$\frac{\gamma}{\gamma_0} - 1 = -\frac{\Delta}{a}(F + H_2) + O((\Delta/a)(1/Q)) + O((\Delta/a)^2).$$

$$(3.32)$$

Thus we expect significant percentage errors in $\gamma/\gamma_0 - 1$, as is in fact found to be the case. However, it remains true that γ/γ_0 is altered only by order Δ/a, which is often adequate for applications.

D. Physical interpretation for quadrupole distortions

Some aspects of the results for microdroplets have a simple interpretation in geometric optics. For simplicity we consider a quadrupole distortion, $(L, M) = (2, 0)$. Resonance should occur when the path length s and the wavelength λ/n are in a definite ratio, or $s/\lambda = X$ where X is a fixed number. Hence

$$\frac{\Delta\omega}{\omega} = -\frac{\Delta s}{s}. \qquad (3.33)$$

To the extent that the photon can be regarded as traveling round the rim, s would be equal to the circumference.[27]

Now consider the mode with $m = l$. Since l_z is maximum, the photon path is around the equator, so

$$s = 2\pi r_e = 2\pi a(1 - e/3), \quad \Delta s/s = -e/3$$

giving $\Delta\omega/\omega = e/3$, in agreement with (1.7) for $l \gg 1$. Next suppose $m = 0$; since $l_z = 0$, the photon path is along a meridian, so

$$s = 2\pi(r_p r_e)^{1/2} = 2\pi a(1 + e/6), \quad \Delta s/s = e/6,$$

giving $\Delta\omega/\omega = -e/6$, again in agreement with (1.7).

This interpretation then provides yet another check on the formalism, and "explains" why the results are independent of the details of the radial functions, and of polarization.

IV. DISCUSSION

We have developed a convenient formalism for dealing with the first-order perturbation of the frequencies (real and imaginary parts) of quasinormal modes in open systems. As with all perturbation theory, it can be applied whenever the unperturbed wave functions are known. Shape perturbations of dielectric microspheres provide one such example, and, surprisingly, the results are independent of the details of the wave functions and can be expressed explicitly in terms of the angular momentum values. This simplification occurs because the dependence on the mode indices and on the angular momentum L of the shape perturbation factorizes, and the $L = 0$ case is trivial.

These results open the way to addressing a number of issues involving MDR's in dielectric microspheres.

(1) It has been observed that the effective Q values of the MDR's as determined from photon storage time are smaller than expected, limited to about 10^6.[28] One possible explanation is that thermal fluctuations generate shape distortions, of order $\Delta/a \sim 10^{-6}$, thus splitting the $2l + 1$ modes of a multiplet by $\Delta\omega/\omega$ of the same magnitude [since F in (1.3) is of order unity]. If the frequency spread can be interpreted as the inverse of the confinement time in the usual way, that would then place a limit on Q of the right order of magnitude. It would be interesting to investigate this mechanism in detail.[29]

(2) One can calculate the elastic scattering cross section of electromagnetic waves by slightly deformed spheres in the following approximate way. Select only one resonant multiplet; for all other modes, assume the scattering is completely unchanged. For the $D = 2l + 1$ resonant modes, go to the basis defined by the eigenvectors of (2.25), which would be the usual states labeled by azimuthal quantum number m if the perturbation preserves axial symmetry. In this basis, the scattering matrix T for the unperturbed system is schematically

$$T^{\alpha\beta} = B\delta^{\alpha\beta} + R(\omega_0)\delta^{\alpha\beta} \qquad (4.1)$$

where B denotes background and $R(\omega_0)$ denotes a resonance described by a pole at the complex frequency ω_0. Now simply let the perturbed T matrix be

$$T^{\alpha\beta} = B\delta^{\alpha\beta} + R(\omega_0 + \omega_1^{\alpha})\delta^{\alpha\beta} \qquad (4.2)$$

where ω_1^{α} are the frequency shifts calculated in Sec. III.

Of course we can calculate the change in $T^{\alpha\beta}$ perturbatively, but (4.2) is equivalent to summing an infinite set of diagrams in T. We expect (4.2) to be accurate whereas a low-order truncation of the perturbation series for T to be inaccurate, because T is a very sharp function of frequency (or size parameter x); a frequency shift is essentially a "horizontal shift" of the resonance profile, which is equivalent to a very large "vertical shift" in T, i.e.,

$$\frac{\Delta T}{T} \sim \left| \frac{\omega_0}{T} \frac{\partial T}{\partial \omega_0} \right| \frac{\omega_1}{\omega_0} \sim Q\frac{\omega_1}{\omega_0} \gg \frac{\omega_1}{\omega_0} . \qquad (4.3)$$

Work on these issues is in progress, and planned to be reported elsewhere.

ACKNOWLEDGMENTS

APPENDIX A

We shall evaluate $G^{\alpha\beta}$ explicitly for the case of spherical dielectric droplets and demonstrate that it is independent of R. First of all, consider the electric field $\mathbf{E}$ of TE quasimodes, which are given by (3.1) with

$$\phi(r) = \begin{cases} Aj_l(nkr), & r \le a \\ h_l^{(1)}(kr), & r > a \end{cases} \qquad (A1)$$

where j_l is the spherical Bessel function and $h_l^{(1)}$ the outgoing spherical Hankel function. This appendix will deal only with the unperturbed functions, so we write the wave number k_0 simply as k, which is nevertheless complex. Boundary conditions at $r = a$ give

$$A = h_l^{(1)}(x)/j_l(nx) \qquad (A2)$$

and

$$\frac{j_l(nx)}{nj_l'(nx)} = \frac{h_l^{(1)}(x)}{h_l^{(1)'}(x)} \qquad (A3)$$

where $x = ka$ is a complex number (but with only a small imaginary part) and the prime denotes differentiation with respect to the argument. It is then straightforward to prove that

$$G = n^2 \int_0^a dr\, r^2 j_l(nkr)^2 + \int_0^R dr\, r^2 [h_l^{(1)}(kr)]^2$$
$$+ \frac{i}{2\omega}[Rh_l^{(1)}(kR)]^2 . \qquad (A4)$$

The first two terms come from the volume integrals while the last term is the surface integral. The integrals in the expression of G can be performed using the standard formula

$$\int_0^x dx\, x^2 [Z_l(\alpha x)]^2$$
$$= \frac{x^3}{2}\{[Z_l(\alpha x)]^2 - Z_{l-1}(\alpha x)Z_{l+1}(\alpha x)\} ,$$

where $Z_l(\alpha x)$ is any spherical Bessel function, and G is simplified into a sum of three terms:

$$G = (n^2 - 1)\frac{a^3}{2}[h_l^{(1)}(x)]^2 + \frac{a^3}{2}G_1(x) + \frac{1}{2k^3}G_2(\xi) \qquad (A5)$$

with

$$G_1(x) = h_{l-1}(x)h_{l+1}(x)$$
$$- n^2 \left[\frac{h_l^{(1)}(x)}{j_l(nx)} \right]^2 j_{l-1}(nx)j_{l+1}(nx) ,$$

$$G_2(\xi) = \xi^3\{[h_l^{(1)}(\xi)]^2 - h_{l-1}^{(1)}(\xi)h_{l+1}^{(1)}(\xi)\} + i\xi^2[h_l^{(1)}(\xi)]^2 ,$$

and $\xi = kR$. It is not difficult to show that $G_1(x) = 0$ using (A3) and the well-known properties of Bessel functions. Moreover, as $|\xi| \to \infty$, we can apply the asymptotic expansion of Hankel function to simplify $G_2(\xi)$, which gives

$$G_2(\xi) \sim (e^{2i\xi}/\xi)[1 + O(\xi^{-1})] .$$

If $|\text{Re}(k)/\text{Im}(k)| \gg 1$ or, in other words, the quality factor of the quasinormal mode is very large, there exists a range of values of R such that $|\xi| \gg 1$ while $|\text{Im}\,\xi| \ll 1$ and hence $G_2(\xi)$ is negligible. G is then proved to be given by

$$G = (n^2 - 1)\frac{a^3}{2}[h_l^{(1)}(x)]^2 \qquad (A6)$$

independent of R for sufficiently large values of R.

Secondly, consider the TM case whose magnetic fields are given by (3.16) with

$$\phi(r) = \begin{cases} -in^2 k A' j_l(nkr), & r \le a \\ -ikh_l^{(1)}(kr), & r > a \end{cases} \qquad (A7)$$

while the electric fields are given by (3.18). From boundary conditions one can show that

$$A' = \frac{h_l^{(1)}(x)}{n^2 j_l(nx)} \qquad (A8)$$

and

$$\frac{[xj_l(nx)]'}{n^2 j_l(nx)} = \frac{[xh_l^{(1)}(x)]'}{h_l^{(1)}(x)} \equiv B . \qquad (A9)$$

Using the same trick as in the TE mode case, it is not difficult to prove that

$$G = \frac{n^2 - 1}{n^2} \frac{x}{2} h_l^{(1)}(x)^2 [n^2 B^2 + l(l+1)] , \qquad (A10)$$

again independent of R.

From (A6) and (A1), G for TE modes is given by

$$G = (n^2 - 1)\frac{a^3}{2}[\phi(a)]^2 \tag{A11}$$

and it is obvious that $I_R = 1$ by substituting (A11) into (3.6).

For TM modes,

$$I_R = (2G)^{-1}\frac{(1-n^2)a^3}{x^2 n^4}\left[n^2 l(l+1)[\phi(a)]^2 + \left|\frac{\partial(r\phi)}{\partial r}\right|_a^2\right]$$

$$= (2G)^{-1}\frac{(n^2-1)x}{kn^4}\{n^2 l(l+1)[h_l^{(1)}(x)]^2$$

$$+ A'^2[xj_l(nx)]'^2\} \tag{A12}$$

while from (A10) and (A9)

$$G = \frac{x}{2k}\frac{n^2-1}{n^4}\{n^2 l(l+1)[h_l^{(1)}(x)]^2 + A'^2[xj_l(nx)]'^2\} ; \tag{A13}$$

thus I_R is guaranteed to be unity. This fact may be re-

garded as a verification of the perturbation formalism for the case $L = 0$.

APPENDIX B

First of all consider the integral

$$I = \int d\Omega \, Y_{l,\alpha}^* Y_{l,\alpha}(\hat{L}^2 Y_{L,M})$$

$$= L(L+1)\int d\Omega \, Y_{l,\alpha}^* Y_{l,\alpha} Y_{L,M}$$

$$= (-1)^\alpha L(L+1)\frac{2l+1}{\sqrt{4\pi(2L+1)}}$$

$$\times C(llL;000)C(llL;-\alpha,\alpha,M), \tag{B1}$$

and the last line of (B1) follows directly from Eq. (4.6.3) in Ref. 26. However, from the Hermiticity of $\hat{L}^2$ regarded as an operator on the unit sphere, it is easy to show that

$$I = \int d\Omega[\hat{L}^2(Y_{l,\alpha}^* Y_{l,\alpha})]^* Y_{L,M} \tag{B2}$$

and

$$[\hat{L}^2(Y_{l,\alpha}^* Y_{l,\alpha})]^* = [(\hat{L}^2 Y_{l,\alpha}^*)Y_{l,\alpha} + Y_{l,\alpha}^*(\hat{L}^2 Y_{l,\alpha}) + 2(\mathbf{L}Y_{l,\alpha}^*)\cdot(\mathbf{L}Y_{l,\alpha})]^*$$

$$= 2l(l+1)(|Y_{l,\alpha}|^2 - |\mathbf{X}_{l,\alpha}|^2) . \tag{B3}$$

Therefore

$$I = L(L+1)\int d\Omega |Y_{l,\alpha}|^2 Y_{L,M} = 2l(l+1)\left[\int d\Omega |Y_{l,\alpha}|^2 Y_{L,M} - \int d\Omega |\mathbf{X}_{l,\alpha}|^2 Y_{L,M}\right]$$

and

$$\int d\Omega |\mathbf{X}_{l,\alpha}|^2 Y_{L,M} = \left[1 - \frac{L(L+1)}{2l(l+1)}\right]\int d\Omega |Y_{l,\alpha}|^2 Y_{L,M}$$

$$= (-1)^\alpha \frac{2l+1}{\sqrt{4\pi(2L+1)}}C(llL;000)C(llL;-\alpha,\alpha,M)\left[1 - \frac{L(L+1)}{2l(l+1)}\right] . \tag{B5}$$

Substitute the result of (B5) into (3.7) and it yields

$$F(L,l,\alpha) = \frac{2l+1}{\sqrt{2L+1}}[C(llL;000)]^2\left[1 - \frac{L(L+1)}{2l(l+1)}\right]f(\alpha) \tag{B6}$$

where $f(\alpha)$ is given by (3.12). From the recursion relation of the Clebsch-Gordan coefficient[26]

$$[J(J+1) - j_1(j_1+1) - j_2(j_2+1) - 2m(M-m)]C(j_1 j_2 J;m,M-m,M)$$

$$= [(j_1-m+1)(j_1+m)(j_2+M-m+1)(j_2-M+m)]^{1/2}C(j_1 j_2 J;m-1,M-m+1,M)$$

$$+ [(j_1+m+1)(j_1-m)(j_2-M+m+1)(j_2+M-m)]^{1/2}C(j_1 j_2 J;m+1,M-m-1,M) ,$$

(3.13) can be established without too much difficulty.

The α dependence can be expressed more simply as follows:

$$f(\alpha) \propto \int d\Omega\, Y_{l,\alpha}^*(\theta,\varphi) P_L(\cos\theta) Y_{l,\alpha}(\theta,\varphi) , \qquad (B7)$$

which can be written as a linear combination of

$$\int d\Omega\, |(\cos\theta)^k Y_{l,\alpha}(\theta,\varphi)|^2 \qquad (B8)$$

with $k = 0, \ldots, L/2$. We use the identity

$$(\cos\theta) Y_{l,\alpha} = b_{l,\alpha} Y_{l-1,\alpha} + b_{l+1,\alpha} Y_{l+1,\alpha} , \qquad (B9)$$

$$b_{l,\alpha} = \left[\frac{(l+\alpha)(l-\alpha)}{(2l-1)(2l+1)} \right]^{1/2} \qquad (B10)$$

repeatedly to eliminate $(\cos\theta)^k$. The integrand in (B8) can then be expressed as a sum of terms of the form

$$\left[\prod_i b_{l_i,\alpha} \right] Y_{l',\alpha}^* Y_{l,\alpha} . \qquad (B11)$$

It is readily verified that if $l = l'$, then each $b_{l_i,\alpha}$ in the product occurs an even number of times, giving an α dependence

$$b_{l_i,\alpha}^2 \propto (l_i + \alpha)(l_i - \alpha) = l_i^2 - \alpha^2 . \qquad (B12)$$

Since there are at most L factors of b, (B7) can be expressed as a polynomial in α up to degree α^L.

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