Mie resonance in dielectric droplets with internal disorder

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We consider the influence of refractive index fluctuations (due to randomly distributed inclusions inside a dielectric spherical droplet) on the line shapes of scattering Mie resonances. The significant difference in the spatial distributions of the mode functions participating in the process does not allow one to employ the standard statistical ensembles used in random matrix theory. We propose to model the system by a simplified random ensemble which gives a very good agreement with the available experimental data, and we predict a type of line shape for narrow scattering resonances.

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I. INTRODUCTION

The scattering cross section of light by spherical homogeneous droplets depends on a single size parameter $2\pi a/\lambda$, given by the ratio of the droplet radius a and the light wavelength λ. At certain values of this parameter, one observes narrow scattering resonances, so-called Mie resonances [1], that correspond to long-living whispering gallery modes. At resonance with such a long-living mode, even for a low or moderate intensities of the incident light, the field strength inside the droplet becomes so large that highly nonlinear optical processes can occur with high rates. This property is of significant applied interest in the context of nonlinear quantum optics [2,3], laser physics [4], and nonlinear spectroscopy of weakly absorbing impurities [5,6].

In this paper we consider the effect of the refractive index disorder on the optical properties of such droplets. The inhomogeneity of the medium in the presence of randomly distributed inclusions increases the extinction inside the droplet and results in a decrease of the quality factors of the long-living modes, thus limiting the performances that are most interesting for applications.

Effects of large disorder have been widely studied in nuclear [7] and solid-state physics [8,9]. The description of universal phenomena in disordered media is usually done with the help of random matrix theory for Gaussian-distributed orthogonal, unitary, or simplectic ensembles [7,9]. However the regime of interest for the optics of droplets differs significantly due to the fact that the random perturbation is rather small, since the mean free path of a photon inside the droplet considerably exceeds the droplet size. In other words, the perturbation does not completely destroy the Mie resonances, and the properties of such an object are not universal—they depend not only on the disorder correlation properties, but also on particular parameters of the droplet such as its radius and mean refractive index and the frequency of the incident light. This situation has much in common with the description of metallic clusters [10], where the effects of the electron-phonon interaction have also been considered with the help of random matrix theory.

There is, however, an important difference between Mie scattering by dielectric droplets and the electronic properties of metallic clusters. For Mie scattering the random coupling of long-living resonances to other modes differs from the coupling of short-living modes among themselves, since the long- and short-living modes have very different and weakly overlapping mode functions. Therefore, they cannot be considered by standard random matrix theory with identical statistics of all the matrix elements of the perturbation. In other words, the perturbation matrix belongs neither to a Gaussian orthogonal nor a Gaussian unitary ensemble.

Here we develop an approach to the description of systems perturbed by a Gaussian random matrix, in which different matrix elements do not have the same dispersion. It turns out that perturbation by such a matrix results in an interesting type of line shape, that is different from the usual broadening of resonances known as Wigner semicircles [7].

In Sec. II, we recall some known relations for light scattering by spherical homogeneous droplets. In Sec. III, we present a standard general approach to randomly perturbed systems. In Sec. IV, we apply this to our particular case and introduce a model for the statistics of matrix elements. In Sec. V, we establish a relation between the mean square of the perturbation and the correlation function of the disorder. In Sec. VI, we discuss the results of the calculations and predict an interesting effect: a splitting of long-living Mie resonances into doublets. In Sec. VII we compare the results obtained for the quality factor with the experimental data of Ref. [11]. In Sec. VIII, we conclude by summarizing the main results obtained and a short discussion.

II. LIGHT SCATTERING OFF SPHERICAL DROPLETS

Light scattering by a sphere was considered in Refs. [12,13]. Here we present the main results of this theory. Let us consider the scalar wave equation

$$\nabla^2\psi+k^2m^2\psi=0, \tag{1}$$

where $k=\omega/c$ is the wave number in vacuum, m is the complex refractive index of the medium which depends on the frequency ω, and ψ is a scalar wave function.

In spherical coordinates, two linearly independent solutions of this equation read

$$\psi_{nl}=\frac{mk}{\pi}\sqrt{(2n+1)\frac{(n-l)!}{(n+l)!}}\begin{Bmatrix}\cos(l\varphi)\\ \sin(l\varphi)\end{Bmatrix}\times P_n^l(\cos(\theta))z_n(mkr), \tag{2}$$

where l and n are angular indices $0\leqslant l\leqslant n$, $P_n^l(\cos(\theta))$ are the associated Legendre polynomials, and $z_n(mkr)$ is either of the two kinds of linearly independent spherical Bessel functions. The general solution of Eq. (1) is a linear combination of these two solutions.

The electric and magnetic fields are vectors satisfying the Maxwell equations

$$\vec{\nabla}\times\vec{E}=-ik\vec{H} \quad \text{and} \quad \vec{\nabla}\times\vec{H}=ikm^2\vec{E}, \tag{3}$$

and hence both $\vec{E}$ and $\vec{H}$ satisfy the vector wave equation

$$(\nabla^2+k^2m^2)\vec{\mathcal{V}}=\vec{0}, \tag{4}$$

which is equivalent to three scalar equations of the type of Eq. (1). The general gauge-invariant solution of Eq. (4) can be written as a linear combination of two solutions ϕ and ψ of Eq. (1) and their first and second derivatives. For the electric and magnetic fields, these solutions can be written in the form

$$\vec{E}=(\vec{M}_\psi+i\vec{N}_\phi) \quad \text{and} \quad \vec{H}=m(i\vec{N}_\psi-\vec{M}_\phi), \tag{5}$$

which implies that the electric and the magnetic fields are superposition of two modes: transverse electric (first term in parentheses) and transverse magnetic (second term). Here we concentrate only on the transverse electric mode [14] and take [15]

$$\vec{E}=\vec{M}_\psi \quad \text{and} \quad \vec{H}=im\vec{N}_\psi, \tag{6}$$

where the components of $\vec{M}$ and $\vec{N}$ in spherical coordinates read

$$M_\psi^r=0,\quad N_\psi^r=\frac{1}{mk}\frac{\partial^2(r\psi)}{\partial r^2}+mkr\psi$$

$$M_\psi^\theta=\frac{1}{r\sin(\theta)}\frac{\partial(r\psi)}{\partial\varphi},\quad N_\psi^\theta=\frac{1}{mkr}\frac{\partial^2(r\psi)}{\partial r\partial\theta}$$

$$M_\psi^\varphi=-\frac{1}{r}\frac{\partial(r\psi)}{\partial\theta},\quad N_\psi^\varphi=\frac{1}{mkr\sin(\theta)}\frac{\partial^2(r\psi)}{\partial r\partial\varphi}. \tag{7}$$

For droplets in vacuum we cast the incident ψ_{inc}, scattered ψ_{sca}, and internal ψ_{int} waves in term of spherical Bessel functions j_n and $h_n^{(1)}$, and associated Legendre polynomials P_n^l:

$$\psi_{inc}=e^{i\omega t}\sum_{n=0}^{\infty}(-i)^n\frac{2n+1}{n(n+1)}j_n(kr)\times P_n^1(\cos(\theta))\sin(\varphi),$$

$$\psi_{sca}=e^{i\omega t}\sum_{n=0}^{\infty}(-i)^n\frac{2n+1}{n(n+1)}b_n(k)h_n^{(1)}(kr)\times P_n^1(\cos(\theta))\sin(\varphi), \tag{8}$$

$$\psi_{int}=e^{i\omega t}\sum_{n=0}^{\infty}(-i)^n\frac{2n+1}{n(n+1)}a_n(k)j_n(mkr)\times P_n^1(\cos(\theta))\sin(\varphi).$$

To find the coefficients $a_n(k)$ and $b_n(k)$, we make use of boundary conditions at the droplet surface $r=a$ and arrive at

$$\begin{aligned} j_n(ka)+b_n(k)h_n^{(1)}(ka)&=a_n(k)j_n(mka),\\ j_n'(ka)+b_n(k)h_n^{(1)'}(ka)&=ma_n(k)j_n'(mka), \end{aligned} \tag{9}$$

where j_n' and $h_n^{(1)'}$ are j_n and $h_n^{(1)}$ first derivatives with the argument of the function, and m is the refractive index of the droplet. This yields

$$\begin{aligned} a_n(k)&=\frac{h_n^{(1)}(ka)j_n'(ka)-j_n(ka)h_n^{(1)'}(ka)}{mh_n^{(1)}(ka)j_n'(mka)-j_n(mka)h_n^{(1)'}(ka)},\\ b_n(k)&=\frac{j_n(mka)j_n'(ka)-mj_n(ka)j_n'(mka)}{mh_n^{(1)}(ka)j_n'(mka)-j_n(mka)h_n^{(1)'}(ka)}. \end{aligned} \tag{10}$$

Coefficients of Eq. (10) together with Eqs. (6)—(8) determine fields $\vec{E}$ and $\vec{B}$ for a uniform dielectric droplet in transverse electric mode. Therefore, with the allowance for Eq. (10) the functions z_n of Eq. (2) take the form

$$z_n=\begin{cases} a_n(k)j_n(mkr), & r<a\\ j_n(kr)+b_n(k)h_n^{(1)}(kr), & r>a. \end{cases} \tag{11}$$

The imaginary part of scattering coefficient $b(k)=\Sigma_n b_n(k)$, responsible for extinction in an ensemble of droplets, is shown in Fig. 1. It consists of a succession of narrow and broad resonances. Note that the dependence of the coefficent $a(k)$ has broad and narrow peaks in the same resonances position as $b(k)$. In what follows we study the deformation of this profile with the increase of random perturbation of the refractive index caused by inclusions.

III. LIGHT SCATTERING PERTURBED BY INCLUSIONS

In order to consider the effect of inclusions, we assume that they are randomly distributed inside the droplet. This type of assumption is widely employed in different branches of physics, and yields a general result valid for any types of randomly perturbed physical systems, which can be formulated in terms of Green's functions and their renormalization.

Green's functions are solutions at point $\vec{r}$ of a linear differential equation containing a source term at point $\vec{r}_0$. If a linear perturbation $\hat{V}$ is added to the differential operator, then Green's function $\hat{G}$ can be expanded in an all-orders power series over $\hat{V}$,

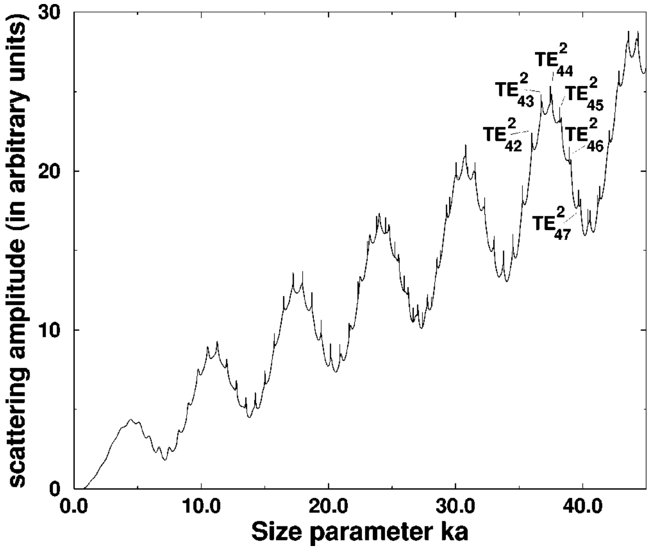

FIG. 1. Scattering coefficient Im$[b(k)]$ responsible for extinction in an ensemble of identical droplets. It is composed of a succession of narrow and broad resonances. The notation TE_n^m represents the transverse electric resonance of mode number n and of mode order m; see Ref. [11]. At a low size parameter, one sees resonances with a low mode number, whereas higher mode numbers correspond to larger-size parameters.

$$\hat{G}=\hat{G}_0+\hat{G}_0\hat{V}\hat{G}_0+\hat{G}_0\hat{V}\hat{G}_0\hat{V}\hat{G}_0+\hat{G}_0\hat{V}\hat{G}_0\hat{V}\hat{G}_0\hat{V}\hat{G}_0+\cdots, \tag{12}$$

where $\hat{G}_0$ is the unperturbed Green's function.

For the case of a random perturbation with zero mean, one finds a general relation for the averaged Green's function $\langle\hat{G}\rangle$, where only even orders of $\hat{V}$ are important. We assume [16] that the averages of the different powers of the perturbation operator $\hat{V}$ are given by the sums of the different combinations of binary correlations, which for the fourth correlation read, e.g.,

$$\begin{aligned}\langle V(r_1)V(r_2)V(r_3)V(r_4)\rangle &=\langle V(r_1)V(r_2)\rangle\langle V(r_3)V(r_4)\rangle+\langle V(r_1)V(r_4)\rangle\\ &\quad\times\langle V(r_2)V(r_3)\rangle+\langle V(r_1)V(r_3)\rangle\langle V(r_2)V(r_4)\rangle.\end{aligned} \tag{13}$$

This assumption, being quite natural for noninteracting inclusions, allows one to find the sum of the perturbation series [Eq. (12)] to all orders. One can depict binary correlations by linking the different $\hat{V}$ with horizontal braces in the perturbation series:

$$\begin{aligned}\langle\hat{G}\rangle &= \hat{G}_0 + \langle\hat{G}_0\overbrace{\hat{V}\hat{G}_0\hat{V}}\hat{G}_0\rangle+\langle\hat{G}_0\overbrace{\hat{V}\hat{G}_0\underbrace{\hat{V}\hat{G}_0\hat{V}}\hat{G}_0\hat{V}}\hat{G}_0\rangle\\ &+\langle\hat{G}_0\overbrace{\hat{V}\hat{G}_0\hat{V}}\hat{G}_0\overbrace{\hat{V}\hat{G}_0\hat{V}}\hat{G}_0\rangle+\langle\hat{G}_0\overbrace{\hat{V}\hat{G}_0\hat{V}\hat{G}_0\hat{V}}\hat{G}_0\hat{V}\hat{G}_0\rangle\\ &+\ldots\end{aligned} \tag{14}$$

The main contribution is known to come from the coupling of ''bubble'' topology, or, in other words, only terms with no crossing of coupling lines are important. In particular the third and fourth terms on the right-hand side of Eq. (14) correspond to the first and second terms in Eq. (13), and the last shown term vanishes.

Terms of this series have a physical meaning. In our particular problem, $\hat{V}$ allows for inclusions into the droplet, and $\langle\hat{G}\rangle$ is the Green's function averaged over all possible distributions of these inclusions. The first term $\hat{G}_0$ represents the case where light encounters no inclusion by crossing the droplet. The second term corresponds to the case where light originally in a given field mode encounters an inclusion, is scattered to another mode, and then scatters back before leaving the droplet. It is sketched by Fig. 2(a).

The third and fourth terms correspond in two different ways to binary correlations of the perturbation in the same order of the expansion. For the third term light is scattered successively at three inclusions and comes back, as shown in Fig. 2(b). For the fourth term it comes back to the first inclusion before going to the third one Fig. 2(c). The next terms are represented in Figs. 2(d), 2(e), 2(f) and 2(g). The expansion can be regrouped in a different way: terms (a), (b), (d), (e), all of which have the same shape at the origin, can be summarized in Fig. 2(h) where the circle around the inclusion means that from this inclusion, light undergoes a certain number of events before coming back. Thus term (h) factorizes as

$$\begin{aligned}(h) &= \langle\hat{G}_0\overbrace{\hat{V}(\hat{G}_0+\hat{G}_0\underbrace{\hat{V}\hat{G}_0\hat{V}}\hat{G}_0+\ldots)\hat{V}}\hat{G}_0\rangle\\ &= \hat{G}_0\underbrace{\hat{V}\langle\hat{G}\rangle\hat{V}}\hat{G}_0\end{aligned} \tag{15}$$

By continuing this reasoning, we can write the Green's function averaged over all inclusions distributions in the following form:

$$\begin{aligned}\langle\hat{G}\rangle &= \hat{G}_0+\hat{G}_0\underbrace{\hat{V}\langle\hat{G}\rangle\hat{V}}\hat{G}_0\\ &+\hat{G}_0\underbrace{\hat{V}\langle\hat{G}\rangle\hat{V}}\hat{G}_0\underbrace{\hat{V}\langle\hat{G}\rangle\hat{V}}\hat{G}_0+\ldots\end{aligned} \tag{16}$$

After factorizing $\hat{G}_0$ out, we can recognize a geometrical series. Therefore $\langle\hat{G}\rangle$ reads

$$\langle\hat{G}\rangle = \frac{1}{\hat{G}_0^{-1}-\underbrace{\hat{V}\langle\hat{G}\rangle\hat{V}}} \tag{17}$$

We now write the general formula [Eq. (17)] for our particular problem, that is for the Green's functions of the Maxwell equations, under the assumption that $\langle V(\vec{r})V(\vec{r}\,')\rangle=V^2\delta(\vec{r}-\vec{r}\,')$. In the k representation, $\hat{G}_0(k,\omega)=(\omega^2-c^2k^2)^{-1}$ has two singularities at $k=\omega/c$ and $k=-\omega/c$ and then $\hat{G}_0(k,\omega)=(1/2\omega)([1/(\omega-ck)]+[1/(\omega+ck)])$. In the case $ak\gg1$, by making use of the resonance approximation, we keep only the resonant term and then $\hat{G}_0^{-1}(k,\omega)=2\omega(\omega-ck)$. The kernel of the operator $\underbrace{\hat{V}\langle\hat{G}\rangle\hat{V}}$ in Eq. (17) can also be found in the k representation:

$$(\underbrace{\hat{V}\langle\hat{G}\rangle\hat{V}})_{nl}^{n'l'}(\vec{k},\vec{k}',\omega)=\int\psi_{nl}^{\star}(\vec{k}\vec{r})(\underbrace{\hat{V}\langle\hat{G}\rangle\hat{V}})(\vec{r},\vec{r}',\omega)\,\psi_{n'l'}(\vec{k}'\vec{r}')d^3r\,d^3r' \tag{18}$$

where

$$(\underbrace{\hat{V}\langle\hat{G}\rangle\hat{V}})(\vec{r},\vec{r'},\omega)=V^2\delta(r-r')\sum_{n''n'''}\sum_{l''l'''}\int\psi_{n''l''}(\vec{k}''\vec{r})\langle G_{n''n'''}(\vec{k}'',\vec{k}''',\omega)\rangle\psi^*_{n'''l'''}(\vec{k}'''\vec{r'})d^3k''d^3k''' \tag{19}$$

and ψ_{nl} are given by Eq. (2), with the corresponding Bessel functions replacing function z_n. Here we have also taken into account that, due to the spherical symmetry, ‘‘on average’’ the Green’s function $\langle\hat{G}\rangle$ does not depend on the ‘‘magnetic’’ quantum number l. The main contribution comes from the diagonal matrix elements with $\vec{r}=\vec{r}'$, $\vec{k}=\vec{k}'$, $n=n'$, $l=l'$, $\vec{k}''=\vec{k}'''$, $n''=n'''$, and $l''=l'''$, and we arrive at

$$(\underbrace{\hat{V}\langle\hat{G}\rangle\hat{V}})^{nl}_{nl}(\vec{k},\omega)=\sum_{n'l'}\int\langle G_{n'n'}(\vec{k}',\vec{k}',\omega)\rangle W^{n'l'}_{nl}(\vec{k},\vec{k}')d^3k', \tag{20}$$

where

$$W^{n'l'}_{nl}(\vec{k},\vec{k}')=V^2\int|\psi_{nl}(\vec{k}\vec{r})|^2|\psi_{n'l'}(\vec{k}'\vec{r})|^2d^3r. \tag{21}$$

We denote $\langle G_{nn}(\vec{k},\vec{k},\omega)\rangle=\langle G_n(\vec{k},\omega)\rangle$, and rewrite Eq. (17) in the compact form [17]

$$G_n(\vec{k},\omega)=\left(\omega-ck-\sum_{n'l'}\int G_{n'}(\vec{k}',\omega)W^{n'l'}_{nl}(\vec{k},\vec{k}')d^3k'\right)^{-1}, \tag{22}$$

which determines $G_n(\vec{k},\omega)$ implicitly. The kernel $W^{n'l'}_{nl}(\vec{k},\vec{k}')$ has a physical meaning. It describes the average squared coupling between modes $(\vec{k},n,l)$ and $(\vec{k}',n',l')$. It is not a constant but depends on the wave number k and on the indices n and l. This is the main difference from a system perturbed by a random matrix with a constant mean square [18]. The spherical symmetry on average allows one to perform a further simplification of Eq. (22) by performing a summation of $W^{n'l'}_{nl}(\vec{k},\vec{k}')$ over l', which results in an l-independent kernel $W^{n'}_{n}(\vec{k},\vec{k}')=\Sigma_{l'}W^{n'l'}_{nl}(\vec{k},\vec{k}')$, and yields

$$G_n(\vec{k},\omega)=\left(\omega-ck-\sum_{n'}\int G_{n'}(\vec{k}',\omega)W^{n'}_{n}(\vec{k},\vec{k}')d^3k'\right)^{-1}. \tag{23}$$

IV. COUPLING OF MODES

In order to find an explicit expression for the kernel $W^{n'}_n(k,k')$, we make use of expressions (2) and (11) for the mode functions, and substitute them into Eq. (19). This yields an expression which contains the factors $|a_n(k)|^2$ and the overlap integrals of the squares of the Bessel functions and of the Legendre polynomials responsible for the radial and angular parts of the mode function, respectively. Coefficients $a_n(k)$ change rapidly with k: they have sharp resonance structures at k corresponding to the whispering gallery modes. Conversely, the integral of Bessel functions is a smooth function of k that we replace by its value at $k=k'=\omega/c$. Moreover, it turns out that the overlap matrix $\hat{W}$ given by the integrals of Legendre polynomials and Bessel functions has just one dominating eigenvalue, which gives more than 80% of the overall contribution to the Schmidt expansion. This corresponds to an eigenvector almost independent of n and n'. Therefore, in the dependence of $W^{n'}_n$ on k, k', n, and n' we keep only the coefficients $|a_n(k)|^2|a_{n'}(k')|^2$, and replace the rest by a constant. For simplicity we denote this by $\tilde{V}^2$, which yields

$$W^{n'}_n(k,k')=\tilde{V}^2|a_n(k)|^2|a_{n'}(k')|^2. \tag{24}$$

Then Eq. (23) takes the form

$$G_n(k,\omega)=\left(\omega-ck-\tilde{V}^2|a_n(k)|^2\times\sum_{n'}\int|a_{n'}(k')|^2G_{n'}(k',\omega)k'^2dk'\right)^{-1}. \tag{25}$$

Equation (25) differs considerably from the case of a random matrix with a constant dispersion of the matrix elements, by the fact that the matrix elements have now different statistics, since the mean square of an element $W_{nn'}(k,k')$ depends on the product $|a_n(k)|^2|a_{n'}(k')|^2$. Note that this is not a general type of dependence, but a particular one: its factorized form is the key assumption of our model.

This form allows one to reduce the operator equation (25) to an algebraic one by introducing the function

$$M(\omega)=\sum_n\int|a_n(k)|^2G_n(k,\omega)k^2dk. \tag{26}$$

After the substitution of $G_n(k,\omega)$ we arrive at

$$M(\omega)=\sum_n\int\frac{|a_n(k)|^2k^2dk}{\omega-ck-\tilde{V}^2|a_n(k)|^2M(\omega)}, \tag{27}$$

which is an algebraic equation with respect to M. We find $M(\omega)$ by an iterative numerical solution of Eq. (27).

We determine the scattering coefficient $b(\omega)$ from the Kramers-Kronig relation where the standard $1/(\omega-ck)$ kernel is replaced by the transformed Green’s functions [Eq. (25)]

$$\mathrm{Re}(b(\omega))=\frac{1}{\pi}\sum_n\int\mathrm{Im}(b_n(k))\times\mathrm{Re}\left(\frac{1}{\omega-ck-\tilde{V}^2|a_n(k)|^2M(\omega)}\right)dk, \tag{28}$$

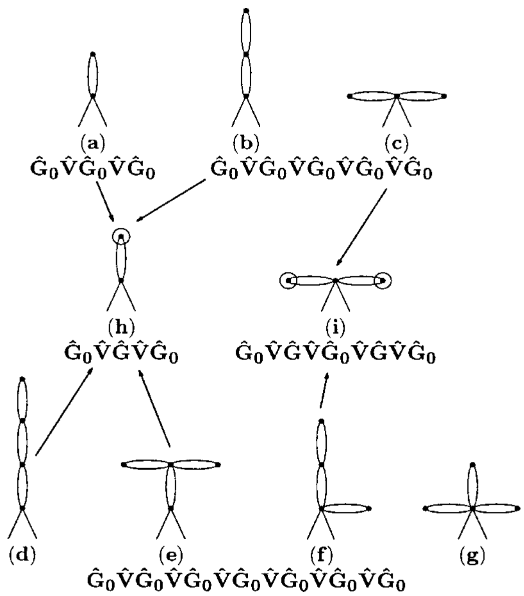

FIG. 2. Representation of the first terms of the Green's function expansion, and their factorization.

$$\mathrm{Im}(b(\omega))=\frac{1}{\pi}\sum_n \int \mathrm{Im}(b_n(k)) \times \mathrm{Im}\left(\frac{1}{\omega-ck-\tilde{V}^2|a_n(k)|^2M(\omega)}\right)dk \quad (29)$$

that we calculate numerically for a given $M(\omega)$.

V. RELATIONSHIP BETWEEN PERTURBATION AND SIZE OF INCLUSIONS

The refractive index m entering Eq. (4) is related to the dielectric constant $\varepsilon=m^2$. In the droplet, ε is not constant because of the presence of randomly distributed inclusions. Therefore, Eq. (4) can be written in the form

$$(\nabla^2+k^2m^2+k^2\delta\varepsilon(r))\vec{E}=0, \quad (30)$$

where $\delta\varepsilon(\vec{r})=\varepsilon(\vec{r})-m^2$. Considering the term $k^2\delta\varepsilon(r)$ as a random perturbation, we can make use of the results of Sec. III. Equation (19) takes the form

$$\begin{aligned}(\hat{V}\langle\hat{G}\rangle\hat{V})(\vec{r},\vec{r}\,',\omega) &= k^4\langle\delta\varepsilon(\vec{r})\delta\varepsilon(\vec{r}\,')\rangle \\ &\times\sum_{n''n'''}\sum_{l''l'''}\int \psi_{n''l''}(\vec{k}''\vec{r}) \\ &\times\langle G_{n''n'''}(\vec{k}'',\vec{k}''',\omega)\rangle\psi^{\star}_{n'''l'''}(\vec{k}'''\vec{r}\,')d^3k''d^3k'''.\end{aligned} \quad (31)$$

Let the variation of the dielectric constant be a superposition of contributions of N statistically independent small spherical inclusions, each with a radius r_0. This corresponds to the perturbation

$$\delta\varepsilon(\vec{r})=\frac{4}{3}\pi r_0^3\delta\varepsilon\sum_{i=1}^{N}\delta(\vec{r}-\vec{r}_i), \quad (32)$$

where the constant shift of the refractive index, which makes $\langle\delta\varepsilon\rangle=0$, and can be included in m, is omitted. Substitution of Eqs. (31) and (32) into Eq. (18) yields Eq. (21) in the form

$$W_{nl}^{n'l'}(\vec{k},\vec{k}\,')=k^4\left(\frac{4}{3}\pi r_0^3\right)^2\delta\varepsilon^2\sum_{i=1}^{N}|\psi_{nl}(\vec{k}\vec{r}_i)|^2|\psi_{n'l'}(\vec{k}\,'\vec{r}_i)|^2. \quad (33)$$

Here $\delta\varepsilon$ is given by the difference of squares of the refractive indices of the droplet medium and the material of inclusions ($m^2_{droplet}-m^2_{inclusions}$), and ψ_{nl} is given by Eq. (8). The sum in Eq. (33) can be interpreted as a contribution of the inclusions that are found at a position where both mode functions are nonvanishing. In other words, it depends on the overlap of the energy distribution in the mode functions. The short-living mode functions of small quality factors occupy, more or less uniformly, all the volume of the droplet, whereas the whispering gallery modes are localized in a narrow ($\sim\lambda$) shell near the droplet surface, and the width of the shell depends on the radial number of the mode. For TE^2_{47} this amounts to 2λ, and Eq. (33) yields

$$W_{nn'}(k,k')=k^4\left(\frac{4}{3}\pi r_0^3\right)^2\delta\varepsilon^2N \times\frac{4\pi a^2(2\lambda)}{\frac{4}{3}\pi a^3}|a_n(k)|^2|a_{n'}(k')|^2. \quad (34)$$

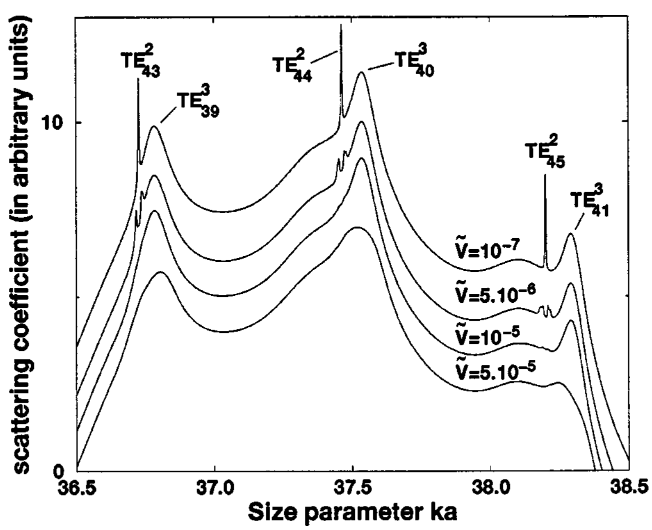

FIG. 3. Imaginary part of the scattering coefficient responsible for the extinction of four different values of the perturbation $\tilde{V}$. Narrow resonances (TE^2_{43}, TE^2_{44}, and TE^2_{45}) broaden, split, and disappear; however wide ones (TE^3_{39}, TE^3_{40}, and TE^3_{41}) are less affected, and only broaden.

We make use of this expression in Sec. VI for comparison of the experimental and theoretical results.

VI. RESULTS

Transformation of the scattering cross section in the presence of a random perturbation, found with the help of Eq. (29), is shown in Fig. 3. One sees that all narrow resonances experience similar shape transformations and split into doublets, whereas broad resonances do not manifest similar behaviors, and their shape remain of a ''bell'' type.

To be specific, here we concentrate on the resonance TE^2_{47} for which experimental data of Ref. [11] are available. In Fig. 4 we show in detail the variation of the resonance contours with the increase of the random perturbation. One sees a gradual broadening of the resonance with an increase of $\tilde{V}^2$. Moreover, the line shape changes, acquiring a double-hump form, which is different from the semicircular shape typical of a resonance broadened by a random perturbation [7,18].

The origin of such a line shape can be traced by considering a model expression that takes into account the variation of the damping rate of a resonance as a function of frequency. Let us consider a profile

$$f(\omega)=\frac{1}{\omega-i\,\dfrac{\gamma}{\omega^2+\gamma^2}-i\gamma'},\tag{35}$$

with the width $\Gamma=[\gamma/(\omega^2+\gamma^2)]+\gamma'$ depending on the frequency ω. Here γ and γ' are two parameters. For large γ, the imaginary part of $f(\omega)$ exhibits only one peak. When γ decreases, we can observe that the function splits into two peaks (Fig. 5).

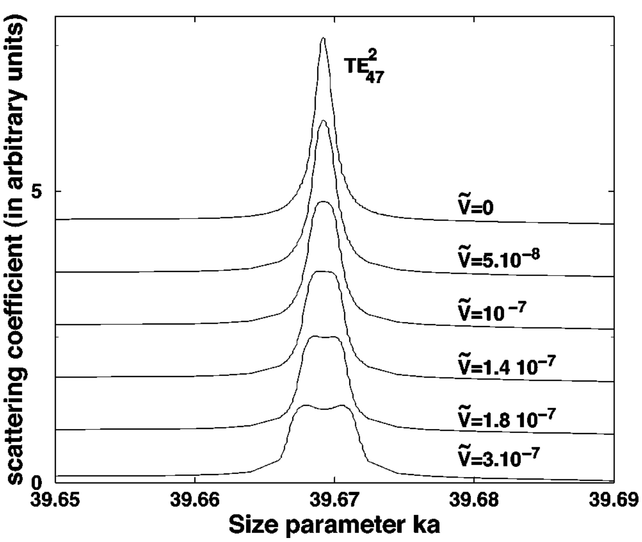

FIG. 4. Broadening and splitting of the transverse electric resonance TE^2_{47} for six different values of perturbation $\tilde{V}$. With increasing $\tilde{V}$, the resonance broadens and its amplitude decreases, its summit becomes flat ($\tilde{V}=1.4\times10^{-7}$), and its center grows hollow ($\tilde{V}=1.8\times10^{-7}$), giving birth to two peaks ($\tilde{V}=3\times10^{-7}$).

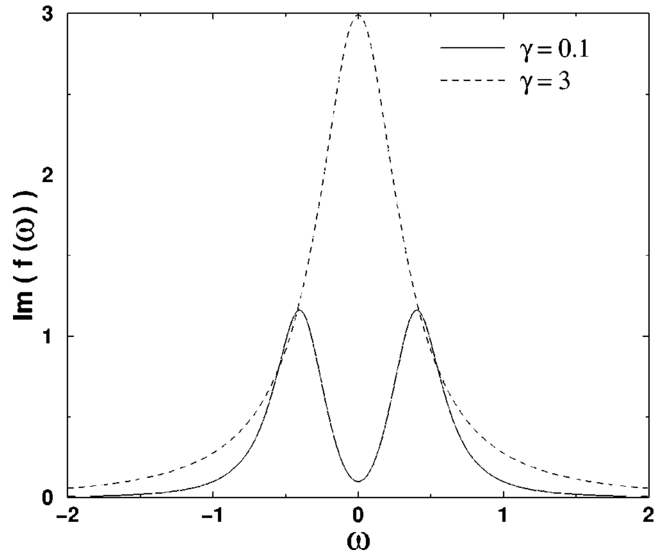

FIG. 5. Two regimes of the imaginary part of the function $f(\omega)$, for $\gamma'=10^{-3}$.

This means that the ratio of the frequency-dependent and -independent dampings determines the shape of the profile. For different Mie resonances this ratio, found in a self-consistent way with the help of Eq. (27), is apparently different. This explains the different behaviors of the narrow and broad resonances.

VII. COMPARISON WITH EXPERIMENT

As we already mentioned at the beginning of this paper, resonances in the intensity of scattered light are functions of a single variable, the size parameter $x=ka$, where a is the radius of the droplet and k is the wave number of the incident light. In the experiment of Ref. [11], the wavelength of the laser which illuminated the droplets was fixed, while the droplets gradually evaporated, so that the radii (and hence the size parameters) were changing. Although our calculations are done for a fixed radius and a variety of frequences, they apparently correspond to the same phenomenon.

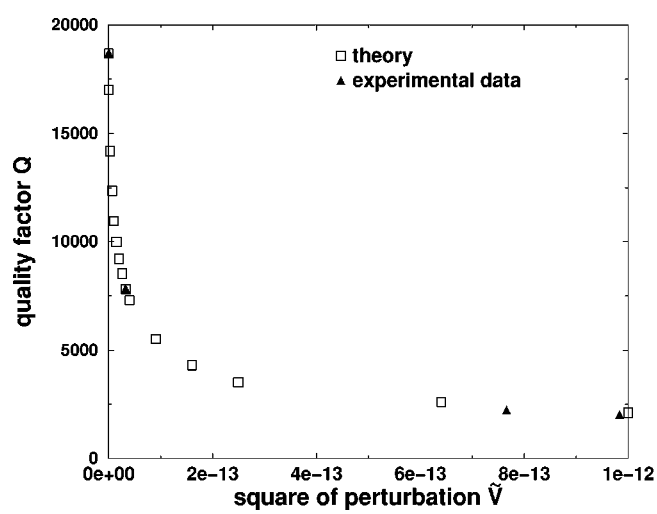

FIG. 6. Comparison of the experimental and theoretical broadenings of TE^2_{47}.

In the experiment of Ref. [11] a droplet of glycerol $m=1.4746$ was seeded by a microscopic $r_0=10–100$ nm powder of latex $m=1.41$ [19]. Unfortunately, the low signal-to-noise ratio typical of experiments with a single object (without an ensemble average) does not allow us to state that the observed line shapes are identical to the calculated ones, although some tendencies toward such a similarity can be traced. But the width of the resonances which manifests itself in the quality factors of the resonances, can be measured as well as calculated. In Fig. 6 we present the experimental results of Ref. [11] along with the calculations of quality factors performed for the profiles of TE_{47}^2 resonance (Fig. 4).

The correspondence between the mean-squared perturbation $\tilde{V}^2$ and the size of inclusions is established by Eqs. (24) and (34). One sees a very good agreement between these data.

VIII. CONCLUSION

We have demonstrated that transformation of the line shapes and experimentally observed quality factors of scattering resonances in dielectric droplets with internal disorder can be described with the help of random matrix theory. However, such a description requires us to take into account the variation of the random coupling with the frequency of the incident light. We have proposed a model where the mean-square coupling $W_{nn'}(\vec{k},\vec{k}')$ of modes characterized by wave vectors $\vec{k}$ and $\vec{k}'$ has the factorized form $W_{nn'}(k,k')=\tilde{V}^2|a_n(k)|^2|a_{n'}(k')|^2$. This allows us to take into account the main features of the random ensemble, and explain the experimentally observed broadenings. It also predicts a specific two-hump shape of narrow resonances, which cannot be obtained in the framework of a standard random-matrix model.

Apart from a practical interest for nonlinear and quantum optics and high-resolution spectroscopy, this result seems to have general interest as an example of a type of profile that can exist in random media, not conforming to standard models (Gaussian orthogonal ensemble or Gaussian unitary ensemble) of Gaussian disorder. One may observe such a profile, for instance, in the line shapes of light emitted by microdroplets of an active media.

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[14] For a complete analysis, one also should include the transverse magnetic (TM) mode. It gives two coefficients $a_n'(k)$ and $b_n'(k)$ which can be found by analogy with the transverse electric (TE) mode, such that Eq. (10) is slightly modified. The coefficients a_n' in principle also have to be included in Eq. (27); however, here we write only the TE part, not only to make the notation shorter but also since TM resonances are far enough from TE resonances to neglect their contribution in the particular case of TE_{47}^2, which we have considered for the comparison with experiment: the width of the resonance is 1×10^{-2} on the scale of size parameter, and the nearest TM resonance is at a distance of 38×10^{-2}.

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[19] We are grateful to Sondes Trabelsi for providing us with these data.