Explicit asymptotic formulas for the positions, widths, and strengths of resonances in Mie scattering

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Explicit asymptotic formulas are derived for the positions, widths, and strengths of the morphology-dependent resonances in Mie scattering. These formulas are compared with numerical data and found to be highly accurate, especially for the low-order resonances most relevant to nonlinear processes. They permit the interpretation of experimental data on light scattering from microdroplets without resorting to the full apparatus of the Mie scattering formalism.

1. INTRODUCTION

The scattering of electromagnetic waves from a uniform dielectric sphere or droplet of refractive index n, also called Mie scattering, has long been a subject of interest both experimentally and theoretically.[1] The ability to produce micrometer-sized droplets in a controlled manner and the use of intense monochromatic laser illumination led to renewed interest over the past decade, with much attention devoted to nonlinear optical phenomena,[2] and to the nontrivial modification of molecular transitions by the droplet—the so-called cavity quantum electrodynamic effects.[3]

The most crucial variable describing interaction of light with droplets is the size parameter $x = ka$, where a is the droplet radius (typically tens of micrometers) and k is the wave number in vacuum. Current interest lies principally with large droplets ($x \gg 1$), for which the scattering exhibits a complicated angular pattern due to interference among many partial waves, and more important, also sharp peaks corresponding to morphology-dependent resonances (MDR's).[4] The resonances can be extremely narrow, with theoretical quality factor Q as high as 10^{20} or more for a perfectly round, homogeneous, and transparent droplet, and in practice Q values in the 10^6–10^7 region have been reported.[6] The small widths imply long storage times for photons and the possibility of strong optical feedback, making nonlinear and stimulated processes[2] specially interesting.

The detailed analysis of these optical processes usually makes use of the full apparatus of Mie scattering theory,[7] which is cumbersome algebraically and even computationally (especially for large x), and moreover tends to obscure the physical ideas, which are often simple. It should be possible to capture the main features of processes dominated by MDR's if one knows the positions, widths, and strengths (in a precisely defined fashion) of these resonances; indeed we have used information on the strengths, expressed as certain sum rules,[8] to give relatively simple descriptions of some optical processes in such droplets.[9] In this context, the present paper provides explicit and accurate asymptotic formulas for the MDR positions (Section 2), widths (Section 3), and strengths (Section 4); these formulas are valid for large droplets and allow the user to bypass the details of Mie theory in the interpretation of experiments.

For resonances with an angular momentum l, the size parameter x lies in the range $l/n \lesssim x \lesssim l$ (see Section 2), with narrow widths for $x \sim l/n$ (low-order resonances) and much larger widths for $x \sim l$. Historically, the main interest lies in elastic scattering; in this case, with the maximum scattering in each partial wave bounded by unitarity, the scattering signal for broadband incident light or in the absence of good wavelength resolution would be proportional to the width. Therefore, theoretical interest was also centered on this regime, and Probert-Jones[10] has obtained formulas for the resonance parameters that are accurate for $x \sim l$. More recent interest is on nonlinear optics, which typically goes as $\exp(g\tau)$, where g is a gain coefficient characteristic of the process, and τ is a storage time, given by $\tau \sim$ (width of resonance)$^{-1}$; therefore our attention will be placed on the narrow low-order resonances with $x \sim l/n$.

In Section 2, the derivation of the asymptotic formula for the position $x_{l,i}$ of an MDR of mode number l and mode order i, as well as the corresponding formula for the spacings $\Delta x_{l,i} = x_{l+1,i} - x_{l,i}$, are sketched. These formulas are compared with numerical results from Mie theory and with existing formulas in the literature.[10,11] The formulas given here are sufficiently accurate to be potentially useful for identifying the quantum numbers of the MDR's, which is important for understanding optical processes that are strongly dependent on the Q values.[12]

In an extended medium, the dispersion relation for a mode of excitation relates the frequency ω to the linear momentum $\mathbf{q}$: $\omega = \omega(\mathbf{q})$; in a droplet, $\mathbf{q}$ is naturally replaced by the angular momentum l, and ω is conveniently expressed by $x = \omega a/c$, so an explicit expression of x in terms of l can be regarded as a dispersion relation. In this sense, the formula is also potentially useful for understanding the conditions for phase matching in processes such as third-harmonic generation in a droplet.[13]

Section 3 provides the derivation of the widths, and a comparison with a formula in the literature.[10]

The strengths of the MDR's are best expressed in terms of a sum rule, which was previously discovered and verified

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Table 1. Roots α_i of Ai($-z$)

i	α_i
1	2.338
2	4.088
3	5.521
4	6.787
5	7.944
6	9.023
7	10.040
8	11.009
9	11.936
10	12.829
11	13.692
12	14.528
13	15.341
14	16.133
15	16.906

computationally[8]; this sum rule is proved in Section 4. The same result is also stated in terms of the coefficients c_l and d_l in the conventional formulation of Mie scattering.[14]

All derivations in this paper are given only for TE polarization, and the similar results for TM modes are stated without detailed proof.

For the benefit of readers who wish to bypass the derivations and use the results directly, the asymptotic formulas for the positions and widths are summarized next.

A. Positions

The MDR's are labeled by a mode number l and an order number i. The index l is defined by saying that for a TE (TM) mode, the electric (magnetic) field has an angular dependence $\mathbf{L}Y_{lm}$, where Y_{lm} are the spherical harmonics and $\mathbf{L} = \mathbf{r} \times (-i\nabla)$ is the angular momentum operator. The index i is essentially the number of radial nodes inside the droplet. We find that the position $x_{l,i}$ of such a mode in a droplet of refractive index n can be expressed as a series in $\nu^{-1/3}$, where $\nu = l + 1/2$:

$$nx_{l,i} = \nu + 2^{-1/3}\alpha_i\nu^{1/3} - \frac{P}{(n^2-1)^{1/2}} + \left(\frac{3}{10}2^{-2/3}\right)\alpha_i{}^2\nu^{-1/3} - \frac{2^{-1/3}P(n^2 - 2P^2/3)}{(n^2-1)^{3/2}}\alpha_i\nu^{-2/3} + O(\nu^{-1}), \tag{1.1}$$

where

$$P = \begin{cases} n & \text{for TE modes} \quad (1.2a) \\ 1/n & \text{for TM modes} \quad (1.2b) \end{cases}$$

and α_i are the roots of the Airy function Ai($-z$), presented in Table 1 for easy reference.

B. Widths

The corresponding FWHM in the x variable is given by

$$\Gamma_{l,i} = 2[Nx^2n_l(x)^2]^{-1}, \tag{1.3}$$

where n_l is the spherical Neumann function and

$$N = \begin{cases} n^2 - 1 & \text{for TE modes} \quad (1.4a) \\ (n^2-1)[\mu^2 + (\mu^2/n^2 - 1)] & \text{for TM modes} \quad (1.4b) \end{cases}$$

and

$$\mu = \nu/x, \tag{1.5}$$

with all x evaluated at $x_{l,i}$.

2. POSITIONS OF RESONANCES

A. Derivation

By going through the standard Mie scattering formalism[14] and imposing the condition that the phase shift δ satisfies $\exp(2i\delta) = -1$, the resonance condition for the TE case can be written as

$$nj_l'(nx)/j_l(nx) = n_l'(x)/n_l(x) \tag{2.1a}$$

or

$$nJ_\nu'(nx)/J_\nu(nx) = Y_\nu'(x)/Y_\nu(x), \tag{2.1b}$$

where the variable $\nu = l + 1/2$ comes about in translating the spherical Bessel and Neumann functions, j_l and n_l, to their cylindrical counterparts, and can be understood heuristically as the total angular momentum because $L^2 = l(l+1) = \nu^2 + O(1) \neq l^2 + O(1)$. What follows is nothing but a systematic asymptotic analysis of Eq. (2.1b).

It is useful to start with some physical insight. For a ray with wave number nk inside the droplet striking the droplet surface at an angle θ to the normal, the angular momentum is

$$\nu = nka \sin\theta = nx \sin\theta. \tag{2.2}$$

But $\sin\theta$ ranges from unity (glancing incidence) to $1/n$ (the limit of total internal reflection), so we expect $nx \gtrsim \nu \gtrsim x$. Thus for large droplets, ν scales with x, and it is therefore convenient to define $\mu = \nu/x (n \gtrsim \mu \gtrsim 1)$. Moreover, low-order resonances (i small) correspond to nearly glancing rays, so we expect $|nx - \nu|$ to be relatively small; in fact, this difference turns out to scale as $\nu^{1/3}$. In anticipation of this dependence, we define a variable t, expected to be $O(1)$, by

$$nx = \nu + t\nu^{1/3}. \tag{2.3}$$

Then the quantities in Eqs. (2.1) may be expressed as asymptotic series in powers of $\nu^{-1/3}$,[15]

$$J_\nu(nx) \sim \frac{2^{1/3}}{\nu^{1/3}}\mathrm{Ai}(-2^{1/3}t)\left[1 + \sum_{j=1}^{\infty} f_j(t)/\nu^{2j/3}\right] + \frac{2^{2/3}}{\nu}\mathrm{Ai}'(-2^{1/3}t)\sum_{j=0}^{\infty} g_j(t)/\nu^{2j/3}, \tag{2.4a}$$

$$Y_\nu(x) \sim -\frac{\exp[\nu(\beta - \tanh\beta]}{(\pi\nu\tanh\beta/2)^{1/2}}\left[1 + \sum_{j=1}^{\infty}(-)^j\frac{u_j(\coth\beta)}{\nu^j}\right], \tag{2.4b}$$

and similar expressions can be written down for the derivatives.[16] In Eqs. (2.4), Ai is the Airy funciton, f_j, g_j, and u_j are known polynomials,[15] and

$$\cosh\beta = \mu. \tag{2.5}$$

The asymptotic expansions are then put into Eqs. (2.1). We illustrate the main idea of our systematic analysis by showing the derivation to first nontrivial order. The two sides of Eqs. (2.1) are asymptotically

$$nJ_\nu'(nx)/J_\nu(nx) \sim -n(2/\nu)^{1/3}\,\mathrm{Ai}'(-2^{1/3}t)/\mathrm{Ai}(-2^{1/3}t) \tag{2.6a}$$

and

$$Y_\nu'(x)/Y_\nu(x) \sim -|\sinh \beta| = -(\mu^2 - 1)^{1/2}. \tag{2.6b}$$

Equating these and taking $\nu \to \infty$, with μ fixed ($n \gtrsim \mu \gtrsim 1$), we see that the powers of ν can only balance if

$$\mathrm{Ai}(-2^{1/3}t) = O(\nu^{-1/3}) \to 0, \tag{2.7}$$

or in other words, that the argument must be close to a root α_i of $\mathrm{Ai}(-z)$:

$$2^{1/3}t = \alpha_i + O(\nu^{-1/3}). \tag{2.8}$$

When this is put into Eq. (2.3) and the resultant value of the size parameter is denoted as $x_{l,i}$, we get

$$nx_{l,i} = \nu + 2^{-1/3}\alpha_i\nu^{1/3} + O(1), \tag{2.9}$$

yielding the first two terms of Eq. (1.1).

Incidentally, by reference to relation (2.4a), the fact that $|2^{1/3}t - \alpha_i| = O(\nu^{-1/3})$ means that, for a TE mode, the droplet surface is nearly a node of the electric field.

The (trivial) leading term in Eq. (2.9) essentially states that the photon follows a circle of radius $r = a$ and that its path length $2\pi a$ is an integral multiple of the wavelength λ/n: $2\pi a = l(\lambda/n)$. [The difference between l and ν can be absorbed into the $O(1)$ term in Eq. (2.9).] But in fact the mode energy is peaked some small distance d inside the droplet surface, so that the photon path is not $2\pi a(a - d)$. This argument would lead to the correction term in Eq. (2.9) if we heuristically identify

$$d/a \sim 2^{-1/3}\alpha_i\nu^{-2/3}. \tag{2.10}$$

We can obtain the higher-order corrections in Eq. (1.1) if write

$$2^{1/3}t = \alpha_i - \sum_{j=1}^{\infty} c_j\nu^{-j/3}, \tag{2.11}$$

expand all the quantities in Eqs. (2.4) in powers of $\nu^{-1/3}$, identify like terms, and thus determine the coefficients. For example, in Eq. (2.4a)

$$\begin{aligned}\mathrm{Ai}(-2^{1/3}t) &= \mathrm{Ai}(-\alpha_i + \Delta)\\ &= \Delta\mathrm{Ai}'(-\alpha_i) + (\Delta^2/2)\mathrm{Ai}''(-\alpha_i) + \ldots,\end{aligned}$$

where Δ is the sum in Eq. (2.11). By the Airy equation and the fact that $\mathrm{Ai}(-\alpha_i) = 0$, all the derivatives are proportional to $\mathrm{Ai}'(-\alpha_i)$, which cancels in forming the logarithmic derivative on the left-hand side of Eqs. (2.1). Similarly,

$$\cosh \beta = \frac{\nu}{x} = \frac{n}{1 + 2^{-1/3}\nu^{-2/3}(\alpha_i - \Delta)},$$

which again permits the quantities in relation (2.4b) to be expanded. Carrying out this evaluation explicitly up to terms of order $\nu^{-2/3}$ then gives Eq. (1.1).

B. Numerical Comparison

Figure 1 shows the pole positions in a plot of $nx - \nu$ versus $\nu^{1/3}$. The liens are the asymptotic formula Eq. (1.1), which would be straight lines to the extent that the $\nu^{-1/3}$ and $\nu^{-2/3}$ terms could be regarded as negligible, or as constant over a limited range of ν; the points are the exact Mie scattering results, obtained by solving Eqs. (2.1) numerically. The agreement is good, especially for the low-order resonances and for relatively large refractive indices.

The approximate locations of MDR's with $x \sim l$ were found to be given by the solution to the equation[10]

$$\tan \phi = \frac{n}{P\rho}\left[\left(\frac{2u}{\nu}\right)^{1/2} - \frac{1}{4u}\right], \tag{2.12}$$

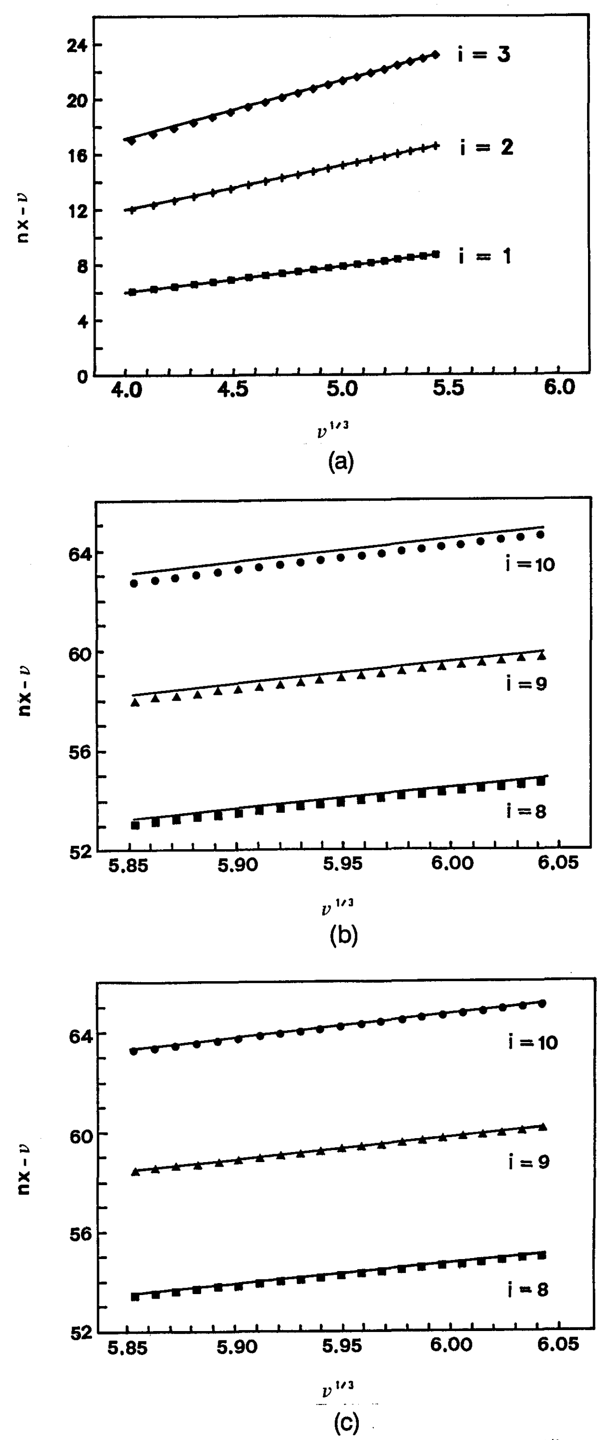

Fig. 1. $nx - \nu$ for TE resonances are plotted against $\nu^{1/3}$ for (a) $n = 1.33$, $i = 1, 2, 3$; (b) $n = 1.33$, $i = 8, 9, 10$, and (c) $n = 1.50$, $i = 8, 9, 10$.

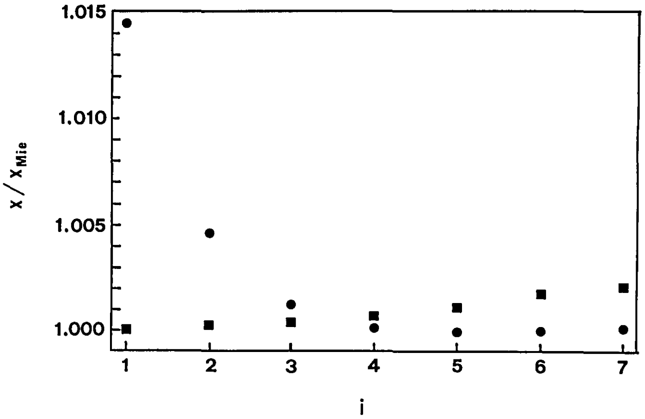

Fig. 2. Approximate locations of TE resonances obtained from our result, Eq. (1.1) (squares) and that of Probert-Jones, Eq. (2.12) (circles), are normalized to the exact values $x_{\rm Mie}$ and plotted against mode order i for $n = 1.33$ and $l = 130$.

where

$$\phi = \nu(\rho - \tan^{-1}\rho) - \rho u + \frac{u^2}{2\nu\rho} - \frac{\pi}{4}, \qquad (2.13)$$

and

$$\rho^2 = n^2 - 1, \qquad u = \nu - x. \qquad (2.14)$$

The locations predicted by this formula are compared numerically with the result of Eq. (1.1) in Fig. 2. The two formulas are seen to be complementary, with the result of Eq. (1.1) more accurate for the narrow low-order resonances and that of Probert-Jones [Eq. (2.12)] more accurate for the broader high-order resonances. However, Eq. (1.1) is explicit and shows the dependence on polarization.

C. Separation

For many purposes, for example the identification of the quantum numbers from the mode pattern,[12] the separation $\Delta x_{l,i} = x_{l+1,i} - x_{l,i}$ may be more useful. We obtain directly from Eq. (1.1) that

$$n\Delta x_{l,i} = 1 + \frac{2^{-1/3}}{3}\alpha_i\nu^{-2/3} - \frac{2^{-2/3}}{10}\alpha_i^2\nu^{-4/3} + \left[\frac{2^{\frac{2}{3}}}{3}\frac{P(n^2 - 2P^2/3)}{(n^2-1)^{2/3}} - \frac{2^{-1/3}}{9}\right]\alpha_i\nu^{-5/3} + O(\nu^{-2}). \qquad (2.15)$$

This formula may be compared with a result by Chýlek[11]:

$$\Delta x = \frac{x}{l}\frac{\tan^{-1}[(nx/l)^2 - 1]^{1/2}}{[(nx/l)^2 - 1]^{1/2}} \qquad \text{for } |x - l| \gg 1/2, \qquad (2.16a)$$

$$\Delta x = \frac{\tan^{-1}[(n^2-1)^{1/2}]}{(n^2-1)^{1/2}} \qquad \text{for } x/l \sim 1. \qquad (2.16b)$$

Table 2 presents the exact separations obtained numerically from Mie theory, or result [Eq. (2.15)] and Chýlek's result [Eqs. (2.16)]; it is seen that Eq. (2.15) is slightly more accurate. More important, Eqs. (2.16) are explicit formulas in that the right-hand sides do not contain x, but instead depend on the mode number l, the order i, and the polarization. We therefore expect that Eqs. (2.16) may be more useful for the purpose of identifying these quantum numbers from experimental data on the resonance separations.

Generally, we expect that nx may be written asymptotically as

$$nx = \nu + a_1\nu^{1/3} + a_0\nu^0 + a_{-1}\nu^{-1/3} + \ldots, \qquad (2.17)$$

where Eq. (1.1) specifies the coefficients $a_1, a_0, a_{-1}, \ldots$, as presented in the first row of Table 3; these coefficients depend on polarization and mode order i, as indeed they should. We can also insert the formal expansion of Eq. (2.17) into Chýlek's expression to identify the implied values of $a_1, a_0, a_{-1}, \ldots$, as in the second row of Table 3, in which c is an undetermined constant. The fact that even the $\nu^{1/3}$ term in Eq. (2.17) is undetermined by Chýlek's formula is hardly surprising, because Eqs. (2.16) make no reference to the quantum numbers.

3. LINEWIDTH

A convenient method of finding the linewidth is to go to the complex k plane, and we define the complex size parameter $z = ka = x + iy$. The logarithmic derivative of the inside solution is $nj_l'(nx)/j_l(nx)$, while for the outside solution it would be $h_l^{(1)\prime}(z)/h_l^{(1)}(z)$ if we allow only an outgoing wave, where $h_l^{(1)}$ is the spherical Hankel function of the first kind. There will in general be a mismatch

$$M(z) \equiv h_l^{(1)\prime}(z)/h_l^{(1)}(z) - nj_l'(nz)/j_l(nz), \qquad (3.1)$$

Table 2. Position $x = x_{l,i}$ for a Mode with Mode Number l and Mode Order i, and $\Delta x = \Delta x_{l,i} = x_{l+1,i} - x_{l,i}$[a]

				Δx		
Mode	l	i	x	(a)	(b)	(c)
TE	50	1	38.0790	0.7087	0.7084	0.7041
	51	1	38.7877	0.7083	0.7080	0.7038
	45	2	38.1999	0.7351	0.7330	0.7276
	46	2	38.9350	0.7342	0.7323	0.7270
	41	3	38.3003	0.7581	0.7545	0.7478
	42	3	39.0584	0.7574	0.7534	0.7468
TM	50	1	38.5483	0.7093	0.7086	0.7067
	51	1	39.2576	0.7089	0.7082	0.7064
	45	2	38.6073	0.7382	0.7334	0.7299
	46	2	39.3455	0.7370	0.7326	0.7292
	41	3	38.5682	0.7663	0.7551	0.7492
	42	3	39.3345	0.7656	0.7539	0.7483

[a] The three columns for Δx refer to (a) exact value from Mie theory; (b) our result, Eq. (2.15); (c) Chýlek's result, Eq. (2.16a), and the refractive index n is 1.4746. Chýlek's result, Eq. (2.16b), would give $\Delta x = 0.7618$ uniformly.

Table 3. Analysis of Our Asymptotic Expansion and Chýlek's Result in Terms of the General Expression

Results	a_1	a_0	a_{-1}
Ours	$2^{-1/3}\alpha_i$	$-\frac{P}{(n^2-1)^{1/2}}$	$\frac{3}{10}2^{-2/3}\alpha_i^2$
Chýlek's	c[a]	$-\frac{1}{2}$	$\frac{3}{20}c^{2a}$

[a] Undetermined coefficient.

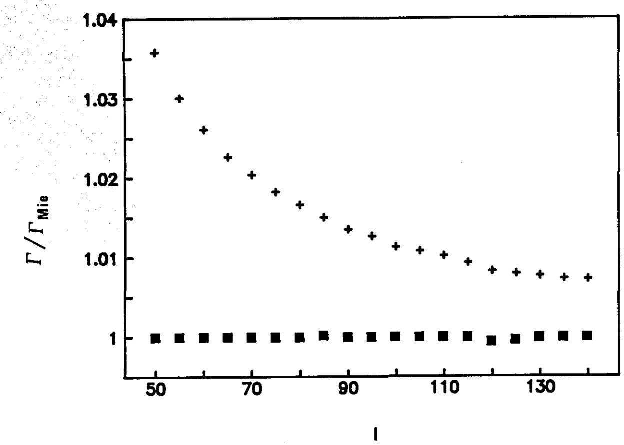

Fig. 3. Approximate widths Γ of TE resonances (normalized to exact values Γ_{Mie}) are evaluated from Eq. (1.3) by substituting approximate values of x given by Eq. (1.1) (crosses) and exact values of x (squares), and plotted against l for $n = 1.33$ and $i = 1$.

and the resonance position, which we denote as $z_0 = x_0 + iy_0$, is defined by $M(z_0) = 0$. In other words, at such a value of z, the solution can satisfy all the following conditions: (a) regular at the origin, (b) only an outgoing wave at infinity, (c) continuity of the wave function, and (d) continuity of the normal derivative; whereas at other values of z we must give up at least one of these conditions, say (d), expressed as a nonzero value of $M(z)$. (A generalization of the mismatch M has been used recently to derive corrections to the positions and widths of the MDR's when the droplet is slightly perturbed.[17])

Now for any real size parameter x near z_0, $M(x) \simeq (x - z_0)M'(z_0)$, and moreover it can be obtained from Bessel's equation that $M'(z_0) = n^2 - 1$, hence in particular at $x = x_0$,

$$M(x_0) = -i(n^2 - 1)y_0. \qquad (3.2)$$

Next, if we use the Wronskian and the fact that $|j_l| \ll |n_l|$ in the domain $l > x \gg 1$, we can show that

$$h_l^{(1)\prime}(x)/h_l^{(1)}(x) \approx \frac{n_l'(x)}{n_l(x)}\left[1 + \frac{i}{x^2 n_l(x) n_l'(x)}\right]. \qquad (3.3)$$

The real part of this equation leads to Eqs. (2.1), while the imaginary part yields

$$\mathrm{Im}\ M(x_0) \approx \frac{1}{{x_0}^2 n_l(x_0)^2}. \qquad (3.4)$$

Because the FWHM Γ in the x variable is given by $\Gamma/2 = -y_0$, Eq. (1.3), for the width follows trivially.

The derivation for TM modes is similar, except that the factor $n^2 - 1$ in Eq. (3.2) is replaced by $(n^2 - 1)[\mu^2 + (\mu^2/n^2 - 1)]$.

The result Eq. (1.3) can be used either by substituting the exact positions x (obtained numerically from Mie theory or from experiment) or by substituting the asymptotic values of x as given by Eq. (1.1). Both yield good agreement with the exact widths, as shown in Fig. 3.

The widths increase with the mode order i, and also in general the TM modes are wider. These dependences are expressed implicitly through the variation of the value of x in Eq. (1.3) with i and polarization.

Probert-Jones[10] has also given an approximate formula for the linewidths, which follows using our notation:

$$\frac{\Gamma}{2} = \frac{n}{P(n^2 - 1)}\left[\left(\frac{2u_0}{\nu}\right)^{1/2} - \frac{1}{4u_0}\right]\exp\left[-\frac{4u_0}{3}\left(\frac{2u_0}{\nu}\right)^{1/2}\right], \qquad (3.5)$$

in which u_0 is the value of $u = \nu - x$ at the resonance position. Figure 4 shows the comparison of this formula as well as our result with the exact widths, indicating that, in contrast to Eq. (1.3), Eq. (3.5) is least accurate for large angular momenta and low order—the regime of interest to nonlinear optics.

4. STRENGTH

The strength of the MDR's also satisfies a simple rule; this rule can be expressed in several equivalent ways, depending on the normalization convention.

A. Sum Rule

Consider a particular polarization (say TE) and angular momentum l, m, and let the most general solution be described by the electric field

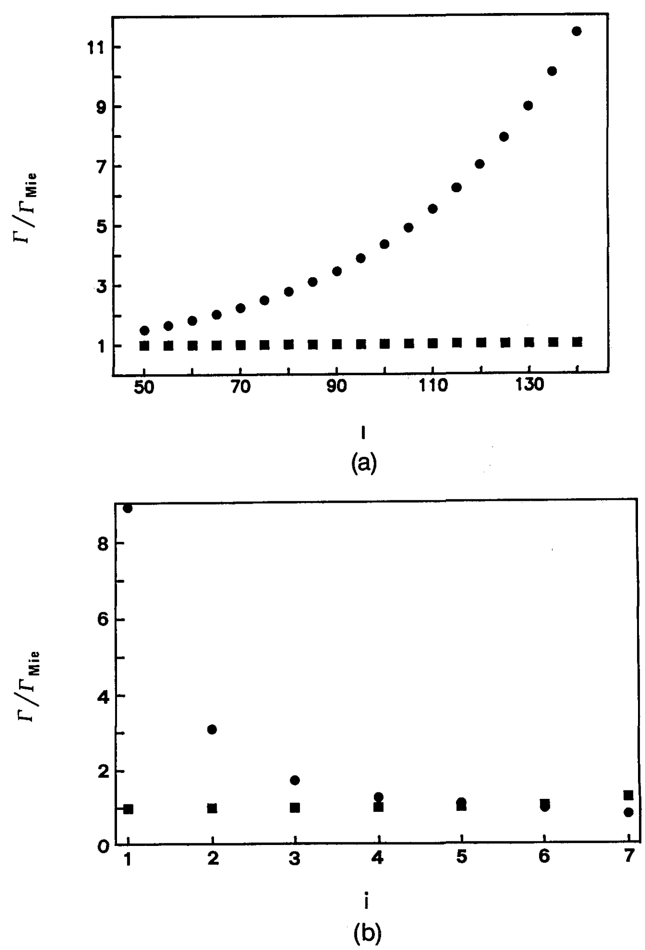

Fig. 4. Approximate widths Γ of TE resonances (normalized to exact values Γ_{Mie}) are evaluated from Eq. (1.3) (squares) and Eq. (3.5) (circles). (a) $\Gamma/\Gamma_{\mathrm{Mie}}$ versus l for $i = 1$; and (b) $\Gamma/\Gamma_{\mathrm{Mie}}$ versus i for $l = 130$. The refractive index is $n = 1.33$ in both figures. Note the difference in vertical scale between Figs. 3 and 4.

$$\mathbf{e}(\mathbf{r}) = \begin{cases} \beta j_l(nkr)\,\mathbf{L}Y_{lm}(\theta,\phi) & r < a \\ [\gamma^{(1)}h_l^{(1)}(kr) + \gamma^{(2)}h_l^{(2)}(kr)]\mathbf{L}Y_{lm}(\theta,\phi) & r > a \end{cases}. \tag{4.1}$$

Here we regard k as real, as is appropriate to a scattering configuration. We enclose the entire system in a universe, which is taken to be a large sphere of radius $\Lambda \gg a$, concentric with the droplet.[8] Requiring the field to vanish at $r = \Lambda$ then restricts k to a discrete set, with spacing $\Delta k = \pi/\Lambda$, and consequently

$$\sum_k = \frac{\Lambda}{\pi}\int \mathrm{d}k\,. \tag{4.2}$$

Now we choose to normalize the mode to unity in the entire universe, i.e.,

$$\int_{r\le\Lambda} n^2(\mathbf{r})|\mathbf{e}|^2\mathrm{d}V = 1\,, \tag{4.3}$$

where $n^2(\mathbf{r})$ is n^2 (1) inside (outside) the droplet. Because $\Lambda \gg a$, the integral is dominated by the outside solution and serves to determine $\gamma^{(i)}$:

$$|\gamma^{(1)}|^2 = |\gamma^{(2)}|^2 = \frac{k^2}{2l(l+1)\Lambda}\,. \tag{4.4}$$

The fact that $|\gamma^{(1)}| = |\gamma^{(2)}|$ comes from the conservation of probability.

Given this normalization convention on Eq. (4.1), we then ask for the magnitude of β, which should then be a measure of the strength of the MDR. It turns out to be even more natural to ask about the electric part of the density of states in the cavity C defined by $r < a$.

$$\rho_{\mathrm{E}}^{\mathrm{C}}(k) = \int_{r\le a} n^2|\mathbf{e}|^2\mathrm{d}V. \tag{4.5}$$

Our result can now be simply stated as

$$\sum_k \rho_{\mathrm{E}}^{\mathrm{C}} = \frac{\Lambda}{\pi}\int \rho_{\mathrm{E}}^{\mathrm{C}}(k)\mathrm{d}k \approx 1\,, \tag{4.6}$$

where the integral is over a range of k covering an MDR. The right-hand side of Eq. (4.6) would become $2l + 1$ if we sum over the degenerate solutions labeled by different m. Note that the factors of Λ in Eqs. (4.2) and (4.4) have canceled, so that the final result is independent of the size of the universe, as indeed it should be.

On account of Eq. (4.3), $\rho_{\mathrm{E}}^{\mathrm{C}}(k)$ is the fraction of the mode of wave number k in the cavity, so the expression in Eq. (4.6) can be regarded as

$$\sum_{\text{modes}} \text{(fraction of each mode in C)} = \text{total number of modes in C}\,.$$

Therefore Eq. (4.6) states simply that, in this sense, an MDR has exactly the strength of one mode. It is necessary to involve the region outside the droplet, i.e., the rest of the universe, because the droplet or cavity by itself is not a Hermitian system,[17,18] so that a mode (in the sense of an eigenfunction of a Hermitian operator) is not a well-defined concept for the droplet on its own.

The statement of the strength of the MDR in terms of the sum rule, Eq. (4.6), is intuitively appealing in this sense. It was first discussed in Ref. 8, and the numerical accuracy of the sum rule was also presented. Now we are in a position to present an analytic proof of the sum rule.

By using Eq. (4.1), the normalization condition on $\gamma^{(i)}$, and the matching of the inside and outside solution to give the ratio $|\beta/\gamma^{(1)}|$, it is readily shown that[8]

$$\rho_{\mathrm{E}}^{\mathrm{C}}(k) = \frac{2n^2k^2}{\Lambda}\frac{\int_0^a r^2 j_l(nkr)^2\mathrm{d}r}{x^4|W[j_l(nx), h_l^{(1)}(x)]|^2}\,, \tag{4.7}$$

where W is akin to a Wronskian

$$W[f(x),g(x)] = f(x)g'(x) - f'(x)g(x)\,, \tag{4.8}$$

but W does not assume a simple form because the functions $j_l(nx)$ and $h_l^{(1)}(x)$ satisfy slightly different differential equations. However, W can be expressed in terms of the mismatch M introduced in Section 3, and, from the fact that for a real size parameter x near a resonance,

$$\begin{aligned} M(x) &\approx (n^2-1)[x-(x_0+iy_0)] \\ &= (n^2-1)[(x-x_0)+i\Gamma/2]\,, \end{aligned} \tag{4.9}$$

we find

$$\begin{aligned} |W[j_l(nx), h_l^{(1)}(x)]|^2 &= |j_l(nx)h_l^{(1)}(x)|^2|M(x)|^2 \\ &= |j_l(nx)h_l^{(1)}(x)|^2(n^2-1)^2[(x-x_0)^2+\Gamma^2/4]\,. \end{aligned} \tag{4.10}$$

For the numerator in Eq. (4.2), the integral can be expressed as j_l^2 multiplied by a sum of 1, j_l'/j_l, and $(j_l'/j_l)^2$. At resonance, the logarithmic derivative can be related to n_l'/n_l by Eqs. (2.1), and because n_l is essentially an exponential function in this domain [see relation (2.4b)], n_l'/n_l is a constant. These considerations lead to

$$\int_0^a r^2 j_l(nkr)^2\mathrm{d}r \approx \frac{a^3}{2}j_l(nx)^2(n^2-1)/n^2. \tag{4.11}$$

Moreover, in this domain $|j_l| \ll |n_l|$, so

$$|h_l^{(1)}(x)|^2 \approx |n_l(x)|^2. \tag{4.12}$$

Substituting relations (4.10), (4.11), and (4.12) into Eq. (4.7) then yields

$$\rho_{\mathrm{E}}^{\mathrm{C}}(k) \approx \frac{a}{\Lambda}\frac{\Gamma/2}{(x-x_0)^2+(\Gamma/2)^2}\,, \tag{4.13}$$

and integrating over the Lorentzian then yields the sum rule, Eq. (4.6).

B. Internal Energy in Mie Scattering

Another way to state the strength of the MDR is to consider Mie scattering with a circularly polarized incident wave ($m = +1$ or -1) of amplitude E_0, with wave number k chosen to be near a resonance of mode number l. Then the incident part of the external field coefficient for the resonant mode is given by

$$\gamma^{(2)} = i^l\left[\frac{\pi(2l+1)}{2l(l+1)}\right]^{1/2} E_0\,. \tag{4.14}$$

Because we have effectively found $|\beta/\gamma^i|$, we can then calculate the average $\mathbf{E}^2$ inside the droplet due to the resonant mode:

$$\frac{\langle E^2\rangle_{\mathrm{res}}}{E_0^2} = \frac{3}{4}\frac{(2l+1)}{x_0^2}\frac{\Gamma/2}{(x-x_0)^2+(\Gamma/2)^2}. \tag{4.15}$$

C. Mie Scattering Coefficients c_l and d_l

Another way is to give the coefficients c_l and d_l for the internal fields, where these are as conventionally defined.[14] Near a TE resonance, we have

$$c_l \approx \frac{n_l(x_0)}{j_l(x_0)}\frac{\Gamma/2}{(x-x_0)+i\Gamma/2}, \tag{4.16}$$

whereas near a TM resonance,

$$d_l \approx \frac{1}{n}\frac{n_l(x_0)}{j_l(x_0)}\frac{\Gamma/2}{(x-x_0)+i\Gamma/2}. \tag{4.17}$$

5. CONCLUSION

We have developed systematic asymptotic formulas for both the positions and the linewidths of the MDR's, which have been checked against exact numerical data with good agreement, especially for low-order resonances of large angular momenta, which are the ones of experimental interest. These explicit formulas will greatly simplify studies on the distribution and characteristics of the MDR's, on the identification of quantum numbers from an experimental pattern, on their phase matching, etc., all of which are important for laser-droplet interactions.

The sum rule on the density of states, which is a convenient and intuitive way of expressing the strength of the MDR's, has been proved analytically. Together with knowledge of the positions and widths, this relation yields a complete characterization of the MDR's (in other words the real and imaginary parts of the pole position of the S matrix, as well as the residue of the pole), and this characterization will be useful for interpreting experimental data without the complication of the full Mie theory.

Another by-product of the present analysis is the development of a geometric-optics picutre, with corrections to the strict geometric-optics limit for glancing rays [e.g., as represented by the nonleading terms in Eq. (1.1)]. Such a picture, in the present study restricted to noninteracting photons, becomes all the more useful as an aid to understanding for the case of nonlinear optics, in which photons interact.

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