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Limitations on the photon storage lifetime in electromagnetic resonances of highly transparent microdroplets

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Thermal fluctuations lead to nonspherical distortions of liquid microdroplets with typical amplitude $\Delta \sim 0.1$ nm. The $2l+1$ modes of a multiplet are then split in frequency by a fractional amount of order $\Delta/a \sim 10^{-5}$, where a is the droplet radius. The width of the multiplet in the frequency domain implies a proportionate reduction in the photon storage lifetime and can limit the effective Q value defined by the photon lifetime to $Q_0 \sim (a/\Delta\varepsilon) \sim (10^5/\varepsilon)$, where ε $(0<\varepsilon<1)$ is process dependent and expresses the fraction of the modes of the multiplet effectively coupled in the process. The theory behind this scenario is presented.

I. INTRODUCTION

When light of vacuum wavelength λ is introduced into a microdroplet of radius a and refractive index $n > 1$, rays can be reflected around the rim. The dimensionless size parameter $x = 2\pi a/\lambda$ is a measure of the ratio of the roundtrip path to the wavelength, and when x assumes definite values (depending only on n), standing waves or resonances will appear. In geometric optics approximation (which is valid as $x \to \infty$), rays sufficiently near the rim undergo *total* internal reflection, so that the electromagnetic energy is completely confined and the quality factor Q of the corresponding resonance is theoretically infinite. However, for the physically interesting range $50 \lesssim x \lesssim 500$, corresponding to $5\ \mu\text{m} \lesssim a \lesssim 50\ \mu\text{m}$, one must go beyond the geometric optics approximation and describe these resonances in terms of electromagnetic modes with angular momentum (l,m), where, for rays near the rim

$$\begin{aligned} l &= (\text{momentum}) \times (\text{impact parameter}) \\ &\sim (nk)(a) = nx \end{aligned}$$

with $k = \omega/c = 2\pi/\lambda$. These modes have been computed for Mie scattering[1] and the internal density of states[2] and do show very sharp resonances. For transparent droplets (n real), the computed Q values increase very rapidly with x, especially for modes without a radial node inside the droplet, corresponding, in geometric optics language, to rays without radial momentum and hence propagating purely tangentially and grazing the droplet surface. For example, for $n = 1.45$ and $x = 170$, $Q \sim 10^{24}$.[1] In other words, the photon lifetime within the droplet can theoretically be extremely large, $\tau = Q/\omega \sim 10^{24}/10^{15}\ \text{s}^{-1} \sim 10^9$ s.

These modes, often called morphology-dependent resonances (MDR's), have been observed in many optical processes: Mie scattering,[3] fluorescence,[4] Raman scattering,[5] Brillouin scattering,[6] chemical energy transfer,[7] and lasing.[8] The large storage time τ is the key to the optical feedback and therefore central to the understanding of nonlinear process.[5,8,9] Recently, experiments using several different techniques[10] have found that the effective Q as defined by the photon lifetime is in fact limited to about 10^6. The droplets used are so transparent that absorption cannot be the principal reason for the reduction in Q and since some of the experiments concern low intensity phenomena, nonlinear processes are also not a likely cause. The purpose of this paper is to resolve this large discrepancy between the theoretical and measured Q values ($\sim 10^{24}$ vs $\sim 10^6$).

The main idea is that due to thermal fluctuations, the surface of the droplet is deformed to

$$r(\theta,\varphi) = a + \sum_{\substack{L \neq 0,1 \\ M}} \Delta_{LM} \sqrt{4\pi}\, Y_{L,M}(\theta,\varphi) \ . \tag{1}$$

The factor of $\sqrt{4\pi}$ is introduced for consistency with the notation in an earlier work[11] on which we shall rely heavily. The $L = 0$ term is excluded because volume changes are suppressed by the bulk modulus, while $L = 1$ terms are excluded because they correspond to pure translation of the droplet. The fluctuations are caused by thermal energy $k_B T$ (where k_B is the Boltzmann constant and T is the temperature) and restrained by the surface tension γ_S, and we shall show in Sec. II that for $L \neq 0, 1$, the thermal ensemble average (denoted by $\langle\ \rangle$) at equal times t is given by

$$\langle \Delta_{LM}(t) \Delta^*_{L'M'}(t) \rangle = \frac{k_B T}{4\pi\gamma_S} \frac{1}{L^2+L-2} \delta_{LL'} \delta_{MM'} \ . \tag{2}$$

In the presence of a shape distortion described by Δ_{LM}, the degeneracy of the $2l+1$ modes of a multiplet is lifted, so that the unperturbed frequency ω_0 becomes $\omega^\lambda = \omega_0 + \omega_1^\lambda$, where $\lambda = -l, \ldots, l$ labels the modes of the multiplet and ω_1^λ is the first-order frequency shift induced by Δ_{LM}. (If $M = 0$, the distortion maintains axial symmetry, then λ is identical with the azimuthal quantum number m; otherwise each mode λ is the linear combination of several m's.) Because these modes are dissipative, the frequencies have a small imaginary part. Ordinary perturbation theory, which is valid only for Hermitian systems with real eigenvalues, can no longer be applied, and the calculation of the frequency shifts becomes nontrivial. We have recently developed a first-order perturbation formalism for such dissipative systems[11] and we apply that formalism in Sec. III to show that the mean square splitting of the multiplet

$$\langle \Delta\omega^2\rangle \equiv \frac{1}{2l+1}\sum_{\lambda}(\omega_1^{\lambda})^2 \equiv \delta^2\omega_0^2 \tag{3}$$

can be expressed analytically in terms of a sum over $\langle \Delta_{LM}(t)\Delta^*_{LM}(t)\rangle$; and using (2),

$$\delta^2=\frac{k_BT}{\gamma_S a^2}C_l \tag{4}$$

where C_l are explicitly calculable numbers, with the asymptotic value

$$C_l \sim 0.04,\quad l \gg 1\ . \tag{5}$$

For processes involving all members of the multiplet in a coherent fashion, the uncertainty relation gives the following limit on the photon lifetime: $\tau \gtrsim \langle \Delta\omega^2\rangle^{-1/2}$. If the process involves only a *faction* of the multiplet, $\langle \Delta\omega^2\rangle^{1/2}$ is effectively reduced by a factor ε, $0<\varepsilon<1$. We therefore obtain

$$\begin{aligned} Q \sim \omega_0\tau &\gtrsim \frac{1}{\varepsilon\delta} \\ &\sim \frac{5}{\varepsilon}\left[\frac{\gamma_S a^2}{k_BT}\right]^{1/2} \equiv Q_0 \end{aligned} \tag{6}$$

where $Q_0 \sim 2\times10^5/\varepsilon$ for a glycerol droplet ($\gamma_S=0.06$, $\mathrm{N\,m^{-1}}$) of radius 10 μm at room temperature. The factor ε is process dependent, but it is reasonable to expect that in most circumstances it is at least ~ 0.1. So we expect that in this case Q_0 lies between 2×10^5 and 2×10^6, roughly in agreement with experiment. The expression for Q_0 is the principal result of this paper and the arguments leading to (6) as well as further discussions are given in Sec. IV.

II. SURFACE FLUCTUATIONS

Consider a droplet surface described by (1), where Δ_{LM} are functions of time. The amplitudes Δ_{LM} of the surface modes satisfy, for $L\neq 0,1$,[12]

$$\ddot{\Delta}_{LM}+\Gamma_L\dot{\Delta}_{LM}+\Omega_L^2\Delta_{LM}=F_{LM}(t)\ . \tag{7}$$

The damping coefficient is

$$\Gamma_L=(2L+1)(L-1)\Gamma\ , \tag{8}$$

$$\Gamma=\frac{2\mu}{\rho a^2} \tag{9}$$

where μ is the viscosity and ρ is the density of the liquid. The restoring force is provided by surface tension γ_S:

$$\Omega_L^2=L(L^2+L-2)\Omega^2\ , \tag{10}$$

$$\Omega^2=\frac{\gamma_S}{\rho a^3}\ . \tag{11}$$

The term $F_{LM}(t)$ on the right-hand side represents the force due to thermal fluctuations, with a white-noise spectrum

$$\langle \tilde F_{LM}(\omega)\tilde F^*_{L'M'}(\omega')\rangle = A_L 2\pi\delta(\omega-\omega')\delta_{LL'}\delta_{MM'}\ , \tag{12}$$

$$F_{LM}(t)=\int (d\omega/2\pi)\tilde F_{LM}(\omega)e^{-i\omega t}\ , \tag{13}$$

and A_L is some normalization constant.

The pressure discontinuity across the surface is

$$p=\gamma_S\left[\frac{2}{a}+(\hat{\mathbf{L}}^2-2)\frac{h}{a^2}\right] \tag{14}$$

where $h(\theta,\varphi)=r(\theta,\varphi)-a$ is the sum in (1) and $\hat{\mathbf{L}}=\mathbf{r}\times(-i\nabla)$ is the angular momentum operator. The second term in (14), proportional to Δ_{LM}, is the excess due to the thermal fluctuations. The potential energy due to the fluctuations is

$$V=\tfrac{1}{2}\int dS\, ph\ . \tag{15}$$

For the contribution of a single mode LM, the excess pressure is

$$\begin{aligned} p&=\gamma_S(\hat{\mathbf{L}}^2-2)\frac{\Delta_{LM}(t)\sqrt{4\pi}Y_{L,M}}{a^2} \\ &=\frac{\gamma_S}{a^2}(L^2+L-2)\Delta_{LM}(t)\sqrt{4\pi}Y_{L,M} \end{aligned} \tag{16}$$

and

$$h=\Delta^*_{LM}(t)\sqrt{4\pi}Y^*_{L,M} \tag{17}$$

giving the potential energy

$$V_{L,M}=\tfrac{1}{2}(4\pi\gamma_S)(L^2+L-2)|\Delta_{LM}(t)|^2\ . \tag{18}$$

Obviously different modes do not interfere. Identifying (18) with $k_BT/2$ (which is valid even for overdamped modes provided the temperature is high[13]) and noting that different modes are independent then gives (2). The characteristic amplitude is

$$\Delta \equiv \left[\frac{k_BT}{4\pi\gamma_S}\right]^{1/2} \tag{19}$$

independent of droplet size, with a value of 0.07 nm for glycerol at room temperatures.

This normalization determines A_L in (12), which, together with (7), can be used to evaluate the *unequal* time correlations $\langle \Delta_{LM}(t)\Delta^*_{L'M'}(t')\rangle$ in a straightforward manner. However, these are not needed for the present problem and will not be shown.

III. FREQUENCY SHIFTS

Since the measured photon storage time is $\tau \sim Q/\omega_0 \sim 10^6/10^{15}\ \mathrm{s^{-1}} \sim 10^{-9}$ s, and the frequency of the surface fluctuations is $\Omega \sim 10^6\ \mathrm{s^{-1}}$, for all practical purposes the electromagnetic modes experience a *static* distortion of the droplet with *constant* values of Δ_{LM}. Moreover $\Delta_{LM}/a \sim 10^{-5} \ll 1$, so first-order time-independent perturbation is valid for calculating the frequency shifts. We have shown[11] that for an electromagnetic mode λ within a multiplet of angular momentum l, the relative frequency shift $\omega_1^\lambda/\omega_0$ is given by the eigenvalue equation

$$\bar{U}^{\alpha\beta}a_\beta^\lambda = \left[\frac{\omega_1^\lambda}{\omega_0}\right] a_\beta^\lambda \tag{20}$$

where

$$\bar{U}^{\alpha\beta} = -\sum_{L,M} \frac{\Delta_{LM}}{a}\sqrt{4\pi}\int d\Omega\, \mathbf{X}^*_{l,\alpha}\cdot Y_{L,M}\mathbf{X}_{l,\beta}\ . \tag{21}$$

In (21) and all subsequent formulas, $L=0,1$ are understood to be excluded. [In Ref. 11, the derivation was only for one term Δ_{LM}, but it is evident that in first-order perturbation theory, the effects of several perturbations are additive. Also, (20) actually applies to the *complex* frequency ω_0 and its *complex* first-order shift ω_1^λ. However, the change in the imaginary part is negligible and will be ignored, so henceforth ω_0 and ω_1^λ may be taken to be just the respective real parts.] The eigenvector $a_\beta^\lambda = (a_1^\lambda, a_2^\lambda, \ldots)$ determines the spatial dependence of the electromagnetic eigenfunctions, and need not concern us here. In (21),

$$\mathbf{X}_{l,\alpha} = \frac{1}{\sqrt{l(l+1)}}\hat{\mathbf{L}}Y_{l,\alpha}$$

are the vector spherical harmonics. The surprising feature of this result is that it is totally independent of the refractive index n and the radial function describing the unperturbed mode (which is in general complicated). The result applies exactly to TE modes, and to within corrections of $O(L^2/l^2)$ for TM modes. Since (2) implies that large L modes are suppressed, the corrections are unimportant.

The average relative shift is

$$\frac{1}{2l+1}\sum_\lambda \left[\frac{\omega_1^\lambda}{\omega_0}\right] = \frac{1}{2l+1}\mathrm{tr}\bar{U}$$
$$= -\frac{1}{2l+1}\sum_{L,M}\frac{\Delta_{LM}}{a}\sqrt{4\pi}\left[\sum_\alpha \int d\Omega\, \mathbf{X}^*_{l,\alpha}Y_{L,M}\mathbf{X}_{l\alpha}\right] . \tag{22}$$

If we consider the thermal ensemble average, of course $\langle \Delta_{LM}\rangle = 0$. But even without this average, we have

$$\sum_\alpha \mathbf{X}^*_{l,\alpha}\cdot\mathbf{X}_{l,\alpha} = \frac{2l+1}{4\pi} \tag{23}$$

with zero projection on $Y_{L,M}, L\neq 0$. Thus the average shift is zero even for a given static perturbation. This property is readily verified using the explicit results in Ref. 11.

Next we calculate the mean square relative shift, averaged over the thermal ensemble:

$$\delta^2 = \frac{1}{2l+1}\sum_\lambda \left\langle \left[\frac{\omega_1^\lambda}{\omega_0}\right]^2\right\rangle = \frac{1}{2l+1}\langle \mathrm{tr}\bar{U}^2\rangle$$
$$= \frac{1}{2l+1}\sum_{L,M}\sum_{L',M'}\frac{\langle \Delta_{LM}\Delta^*_{L'M'}\rangle}{a^2}\sum_{\alpha,\beta}\sqrt{4\pi}\int d\Omega\, \mathbf{X}^*_{l,\alpha}(\Omega)\cdot Y^*_{L,M}(\Omega)\mathbf{X}_{l,\beta}(\Omega)$$
$$\times\sqrt{4\pi}\int d\Omega' \mathbf{X}^*_{l,\beta}(\Omega')\cdot Y^*_{L',M'}(\Omega')\mathbf{X}_{l,\alpha}(\Omega')\ . \tag{24}$$

We use (2) for the ensemble average $\langle \Delta_{LM}\Delta^*_{L'M'}\rangle$ and the identity

$$\int d\Omega\, \mathbf{X}^*_{l,\alpha}(\Omega)\cdot Y_{L,M}(\Omega)\mathbf{X}_{l,\beta}(\Omega)$$
$$= \left[1-\frac{L(L+1)}{2l(l+1)}\right]\int d\Omega\, Y^*_{l,\alpha}(\Omega)Y_{L,M}(\Omega)Y_{l,\beta}(\Omega)\ . \tag{25}$$

Next apply the addition theorem

$$\sum_\alpha Y^*_{l,\alpha}(\Omega)Y_{l,\alpha}(\Omega') = \frac{2l+1}{4\pi}P_l(\cos\gamma) \tag{26}$$

where γ is the angle between Ω and Ω', and similarly to the other two sums

$$\sum_M Y^*_{L,M}(\Omega)Y_{L,M}(\Omega'),\quad \sum_\beta Y^*_{l,\beta}(\Omega')Y_{l,\beta}(\Omega)$$

to get, finally

$$\delta^2 = \left[\frac{k_BT}{\gamma_S a^2}\right]\frac{1}{2l+1}\sum_L \frac{1}{L^2+L-2}\left[1-\frac{L(L+1)}{2l(l+1)}\right]^2$$
$$\times\frac{2L+1}{4\pi}\left[\frac{2l+1}{4\pi}\right]^2$$
$$\times\int d\Omega\, d\Omega' P_L(\cos\gamma)[P_l(\cos\gamma)]^2 . \tag{27}$$

The double integral in (27) is

$$\frac{(4\pi)^2}{2L+1}[C(llL;000)]^2 \tag{28}$$

where $C(l_1l_2l_3;m_1m_2m_3)$ are Clebsch-Gordan coefficients,[14] and in the present case, vanishes for odd L, while for even $L \le 2l$

$$C(llL;000)=(-1)^{l-L/2}\left[\frac{2L+1}{2l+L+1}\right]^{1/2} \times \frac{\tau(2l+L)}{\tau(2l-L)}\frac{1}{[\tau(L)]^2} \tag{29}$$

where

$$\tau(x)=\frac{(x/2)!}{\sqrt{x!}} \; . \tag{30}$$

Then we get (4), with

$$C_l=\frac{2l+1}{4\pi}\sum_{\substack{L>0\\ \text{even}}}^{2l}\frac{1}{L^2+L-2}\left[1-\frac{L(L+1)}{2l(l+1)}\right]^2\frac{2L+1}{2l+L+1} \times \left[\frac{(l+L/2)!}{(l-L/2)!}\right]^2\frac{(2l-L)!}{(2l+L)!} \times \left[\frac{L!}{[(L/2)!]^2}\right]^2 . \tag{31}$$

Table I shows some selected C_l, and since we are typically interested in $l \sim nx \gg 1$, it will be adequate to use the asymptotic value in (5). So for glycerol at room temperatures, $\delta \sim 5\times 10^{-6}$.

Thus the $2l+1$ modes which are originally degenerate at ω_0 become split in frequency by $\sim \omega_0\delta$.

TABLE I. Values of C_l as defined by (31).

l	C_l $(\times 10^{-2})$
4	2.2779
6	2.7766
8	3.0525
10	3.2262
20	3.5914
30	3.7182
40	3.7825
50	3.8213
60	3.8474
70	3.8660
80	3.8800
90	3.8910
100	3.8997
150	3.9260
200	3.9392
250	3.9471
300	3.9524
400	3.9590
500	3.9630

IV. PHOTON STORAGE LIFETIME

Strictly speaking, the effect of mode splitting on the time dependence of an optical process will depend on the details of the process. In this paper, in order not to obscure the main ideas and to emphasize the commonality among different processes, we shall sketch only the general considerations applicable to all processes. In the simplest terms, the increased width in the frequency domain of the multiplet as a whole leads, via the uncertainty principle, to a proportionate reduction in the lifetime, as discussed in Sec. I.

To be more concrete, assume that only a $(2l+1)$-fold multiplet of modes is dominant. In general, we may characterize the experiments in terms of emission (or input coupling) to modes λ with amplitudes $a(\lambda)$, followed by detection (or output coupling) with amplitudes $b(\lambda)$, $\lambda=-l,\ldots,l$. For example, in Mie scattering $a(\lambda)$ and $b(\lambda)$, respectively, describe the projection of the incident plane wave and the detected scattered wave into the relevant modes. In chemical energy transfer, $a(\lambda)$ describes the emission of a virtual photon by a donor molecule (say at position $\mathbf{r}_1$), while $b(\lambda)$ denotes the absorption of the virtual photon by an acceptor molecule (say at position $\mathbf{r}_2$). Then schematically, the amplitude A for the process goes as

$$A \sim \sum_{\lambda} a(\lambda)b(\lambda)e^{-i(\omega_0+\omega_1^{\lambda})t}e^{-\gamma t/2} \tag{32}$$

where $\gamma/2$ is the imaginary part of the frequency, for simplicity assumed to be unaffected by the droplet distortion. To appreciate the time dependence of the amplitude A, it is useful to first consider two extreme cases.

A. Single mode processes

Consider a hypothetical situation in which the droplet suffers a definite distortion (say the "droplet" is a deformed glass bead, with $\Delta_{20}\neq 0$, all other $\Delta_{LM}=0$), without distribution over an ensemble. Consider a specially arranged "Mie scattering" experiment where the incoming wave is not a plane wave, but consists of a single eigenstate λ, and the detection apparatus is designed to accept the same eigenstate λ only. Then the sum in (32) has only *one* term, and we have

$$|A|^2 \sim |a(\lambda)b(\lambda)|^2 e^{-\gamma t} \; . \tag{33}$$

In other words, under these very special circumstances, the photon lifetime is $\tau=1/\gamma$, essentially *unaltered* by the shape distortion. The Q value should be the same as that computed for a perfect sphere, say $\sim 10^{24}$ if absorption could indeed be ignored.

Selection of a single mode can be naturally achieved in some nonlinear processes.[15] In typical experiments,[3–6,15] the droplet, in falling through air, is deformed into an oblate spheroid, typically by $\sim 1\%$, i.e., there is a *static* term $\Delta_{20} \sim 0.1-0.5$ μm in addition to the thermal fluctuations with $\Delta_{LM} \sim 0.1$ nm. The static distortion causes the $2l+1$ modes of a multiplet to split, again by $\sim 1\%$. It is then possible to preferentially excite a few modes, and for these modes, now nondegenerate (at the 1% level), not to mix under the thermal fluctuations (which are at the 10^{-5} level).

Preferential excitation depends on the mode functions, which are most easily visualized as follows. In geometric optics language, the photons run around the circumference along great circles, with **L** normal to the great circle. Thus modes with $|m|\sim l$ correspond to photons going around the equator, and have mode functions concentrated there; modes with $m\sim 0$ correspond to photons going along a meridian, with the mode functions not concentrated near any latitude. Thus if the incident radiation is focused onto a spot on the equator, then it will couple predominantly to the $|m|\sim l$ modes. In nonlinear processes such as stimulated Raman scattering (SRS), the difference in the coupling strength to different modes m leads to a corresponding difference in the intensity thresholds $I_{\text{th}}(m)$ for stimulation, in this case with $I_{\text{th}}(l)<I_{\text{th}}(l-1)<\cdots<I_{\text{th}}(0)$. Thus if the incident intensity I_0 lies in the range $I_{\text{th}}(l)<I_0<I_{\text{th}}(l-1)$, only *one* mode of the multiplet will exhibit SRS, with consequently a storage time that is in principle *not* limited by the thermal fluctuations. Experimental clarification of this issue would be interesting. However, such a strong selection of modes cannot occur for linear processes.

B. Coherent superposition over all modes

It is clear that in most circumstances, especially for processes not involving stimulation, $a(\lambda)\neq 0$, $b(\lambda)\neq 0$ and in fact the product $a(\lambda)b(\lambda)$ must be the same order for all λ. This is especially so since

$$a(\lambda)b(\lambda)\sim\langle\text{out}|\lambda\rangle\langle\lambda|\text{in}\rangle \tag{34}$$

and $|\lambda\rangle$, under the ensemble average, is a *random* vector in the $(2l+1)$-dimensional Hilbert space spanned by the resonant modes. Then the uncertainty principle would give a characteristic decay time

$$\tau\sim\langle(\omega_1^\lambda)^2\rangle^{-1/2} \tag{35}$$

for the sum

$$\sum_\lambda a(\lambda)b(\lambda)e^{-i\omega_1^\lambda t}\ .$$

Since in all cases of interest $\gamma\tau\ll 1$, this is also the characteristic decay time for A. Expressing the right-hand side of (35) in terms of δ and using (4) then leads to the expression for Q_0 in (6) with $\varepsilon=1$.

In short, because the thermal fluctuations are random, the input and output coupling would connect "equally" to all λ. Because of the splitting in frequency, interference causes a relatively rapid decay with $\tau\sim O((\omega_1^\lambda)^{-1})$, the mechanism being the same as in mode locking.

Any actual experimental situation must lie between these two extreme cases, in that more than a single mode is involved but not all modes are equally coupled. This means that $\langle\Delta\omega^2\rangle^{1/2}$, now defined with respect to the terms in the sum in (32), will be reduced by some factor ε, $0<\varepsilon<1$, relative to that in (35). This then leads to (6) in its general form. The precise value of ε can only be found if each process is calculated in detail. But, except in stimulated processes with the intensity just above one threshold, it is unlikely that ε is less than ~ 0.1, so we have found a very useful estimate for Q_0, especially in view of the fact that the discrepancy to be explained is $\sim 10^{18}$ (10^{24} vs 10^6).

Assuming $\varepsilon\gtrsim 0.1$, there is clearly broad agreement with experimental values, showing that the main reason for the reduction in photon lifetime is readily explained through shape perturbations, though the simultaneous existence of other effects, such as Brillouin scattering from density fluctuations, is not excluded. A more detailed comparison would require a careful analysis of the individual experimental conditions [in other words evaluation of $a(\lambda)$ and $b(\lambda)$ and hence ε], and will be given elsewhere.

We end by clarifying the meaning of Q in the frequency domain, in terms of which $Q\sim\omega_0/(\text{width})$. When the multiplet is split, there are two distinct widths: (a) the width γ of a single mode, which is essentially unaltered, and (b) the width of the multiplet as a whole, say $\omega_0\delta$, with δ given by (4), or more generally the width of the fraction ε of the multiplet which is effectively coupled, say $\omega_0\delta\varepsilon$. The former width defines $Q_1\sim\omega_0/\gamma$, say $Q_1\sim 10^{24}$ if absorption were indeed negligible. The latter width defines $Q_2\sim 1/\delta$, or more generally $Q_2\sim 1/(\delta\varepsilon)$, which has a reduced value, say $Q_2\sim 10^6$, on account of thermal fluctuations. The reduction in Q discussed in this paper refers to the effective Q defined by the photon storage time, $Q\sim\omega_0\tau$, which is equal to Q_2 and not Q_1, simply because the process sees the multiplet as a whole (or at least a fraction ε of the multiplet) and not a single mode. In particular, we emphasize that Q_1 is *not* reduced by thermal fluctuations.

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