Random Resonators and Prelocalized Modes in Disordered Dielectric Films

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(Received 28 November 2001; published 13 June 2002)

We calculate the areal density of disorder-induced resonators with a high quality factor, $Q \gg 1$, in a film with fluctuating refraction index. We demonstrate that, for a given $kl > 1$, where k is the light wave vector and l is the transport mean-free path, when *on average* the light propagation is diffusive, the likelihood for finding a random resonator increases dramatically with increasing the correlation radius of the disorder. Parameters of *most probable* resonators as functions of Q and kl are found.

DOI: 10.1103/PhysRevLett.89.016802 PACS numbers: 73.20.Fz, 42.25.Dd, 42.60.Da

Introduction.—Recent discovery of the *coherent* lasing from various disordered materials adds a new dimension to the conventional physics of light propagation in multiply scattering media. In the experimental works [1–4], it was demonstrated that, above a certain excitation level, the emission spectra of ZnO powders [1,2], conjugated polymer films [3], and dye-infiltrated opals [4] exhibit a sequence of extremely narrow peaks, their widths being limited by the spectrometer resolution. Until recently, this finite resolution left some room for doubt as to whether the observed peaks indicated true lasing [5]. However, the latest experiments on photon statistics [6,7] have unambiguously established the coherence of the emitted light, thus proving conclusively that the underlying mechanism of random lasing in [1–4] involves the *amplitude* (coherent) rather than *power* (incoherent) feedback. (The latter is known to occur via the diffusion process of the light intensity, as proposed long ago by Letokhov [8] and observed first by Lawandy [9] and later in other numerous experiments.) The origin of coherent feedback, responsible for the lasing observed in [1–4], is a subject of controversy and debate. Such a feedback would naturally emerge if the light were localized [10,11]. However, the coherent backscattering measurements [1,12–16] carried out in parallel with the analysis of the emission spectra rule out this possibility. First, the values of kl extracted from these measurements turn out rather large. Second, the onset of the Anderson localization manifests itself in the rounding of the top of the backscattering cone [17]. No such rounding was observed in the experiments [1,12–16].

In the early work of Cao *et al.* [1,12,13], it was argued that, even in the diffusive regime of light propagation, a photon, scattered off a certain sequence of impurities (grains of, roughly, 100 nm size) can move on a closed loop, which thereby serves as a laser resonator. The picture of such ring cavities put forward in [1,12,13] was based on the experimental evidence [18] that the *recurrent* scattering processes contribute to backscattering albedo. However, since in each scattering act most of the energy gets scattered out of the loop, an unrealistically high gain would be required to achieve the lasing threshold condition for such a loop. This point was particularly emphasized by van Soest [16], who also argued that "impurity loops" are likely to generate a broad frequency spectrum rather than isolated resonances.

Certainly the picture of random cavities representing a certain spatial arrangement of *isolated* scatterers is too naive. This, however, does not rule out the entire concept of disorder-induced resonators. Although sparse, the disorder configurations that trap the light for a long enough time can occur in a sample of a large enough size, and a *single* such configuration is already sufficient for lasing to occur. Therefore, under the condition $kl \gg 1$, which implies that *overall* scattering is weak, the conclusion about the relevance of random cavities can be drawn only upon the *quantitative* calculation of their likelihood. This is the subject of the present paper.

We start with a remark that the issue of random resonators for light waves has its counterpart in the electronic transport. In particular, the modes with anomalously low radiative losses are analogous to the so-called *prelocalized* electronic states in diffusive conductors that are responsible for the long-time asymptotics of the current relaxation. Theoretical study of these states was launched more than a decade ago (see [19]) and later renewed in [20]. The results obtained until now are summarized in the review [21]. Following this analogy, we dub the modes of random resonators with anomalously high quality factor, Q, as *prelocalized* modes.

The principal outcome of our study is that, for a given $kl \gg 1$, the probability of formation of a high-Q random resonator depends crucially on the *size* of the scatterers, or, more precisely, on the correlation radius of the disorder, R_c. Similarly to the treatment in [2,13], we restrict our consideration to the two-dimensional case (a disordered film). Regarding the geometry of a random resonator, we adopt the idea proposed by Karpov [22] for trapping the acoustic waves in three dimensions. According to [22], the fluctuations responsible for trapping are the toroidal inclusions with reduced sound velocity. Correspondingly, in two dimensions, a random resonator represents a ring-shaped area (see Fig. 1a) within which the effective in-plane dielectric constant is enhanced by some small value ϵ_1 (compared to the background value ϵ). Then such a

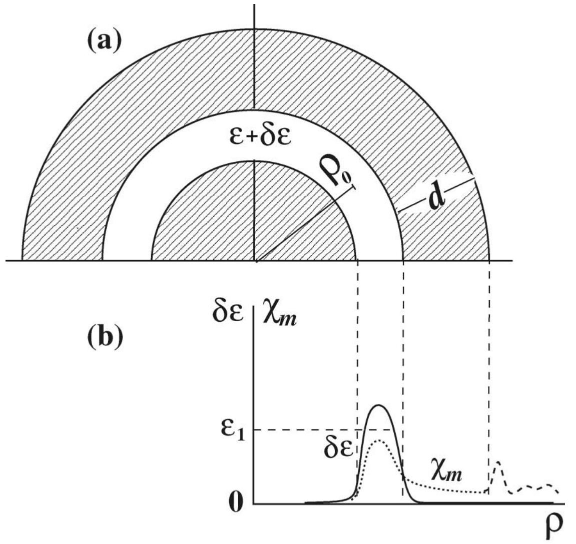

FIG. 1. (a) The structure of a two-dimensional resonator is illustrated schematically; only half of the ring-shaped waveguide (blank region) is shown. (b) Optimal fluctuation of the dielectric constant, $\delta\epsilon(\rho)$ (solid line), and the corresponding field distribution (dotted line) are shown. The dashed line outside the shaded region of a width, d, illustrates the evanescent leakage.

ring can be viewed as a waveguide that supports the modes of a whispering-gallery type. Because of the azimuthal symmetry, these modes are characterized by the angular momentum, m. Denote by $\mathcal{N}_m(kl, Q)$ the areal density of resonators with quality factor Q in the film with a transport mean-free path l. Here $k = \epsilon^{1/2}k_0$, where $k_0 = 2\pi/\lambda$, λ stands for the wavelength in vacuum. Obviously, in the diffusive regime, $kl > 1$, the density $\mathcal{N}_m(kl, Q)$ is exponentially small for $Q \gg 1$. In this domain $\mathcal{N}_m(kl, Q)$ can be presented as

$$\mathcal{N}_m(kl, Q) = \mathcal{N}_0 e^{-S_m(kl,Q)}, \tag{1}$$

where $\mathcal{N}_0$ is the prefactor.

Qualitative estimates.—Let us first give a qualitative estimate for S_m, which reveals its sensitivity to the strength and the range of the disorder (we assume Gaussian disorder with variance Δ^2 and range R_c). Since m is the number of wavelengths along the ring, its radius is $\rho_0 = m/k$. The ring waveguide can support a weakly decaying mode only if its width w satisfies the condition $w(\epsilon_1/\epsilon)^{1/2} \gtrsim k^{-1}$. The decay of the mode is due to the evanescent leakage—the optical analog of the under-the-barrier tunneling in quantum mechanics. A straightforward estimate for the decay time, i.e., the quality factor Q of the waveguide, results in $\ln Q \sim k\rho_0(\epsilon_1/\epsilon)^{3/2}$. Since the number $k\rho_0 = m$ is large, a relatively small fluctuation of the dielectric constant within the area $2\pi\rho_0 w$ of the ring can produce a large value of Q.

The probability P for creating the required fluctuation strongly depends on R_c. For a short range disorder ($k_0R_c \ll 1$), fluctuations of order ϵ_1 should occur *independently* in a large number, $N \sim \rho_0 w/R_c^2$, of spots within the ring, so that the probability $P \sim \exp(-N\epsilon_1^2/\Delta^2)$. In the other extreme of strongly correlated disorder, when $R_c \gg w$, the number of independent spots is much smaller, $N' \sim \rho_0/R_c$ (the number of disks of radius R_c needed to cover the ring). Correspondingly, the probability $P \sim \exp(-N'\epsilon_1^2/\Delta^2)$ is much larger than for a short range case. Finally, using the relation between ϵ_1 and Q, the probability $P = \exp(-S_m)$ can be rewritten in terms of Q, thus, yielding an estimate for S_m. For the short range case ($k_0R_c \ll 1$), we obtain $S_m \sim kl \ln Q$, where the mean-free path l is proportional to $(R_c\Delta)^{-2}$. In the opposite limit of a smooth disorder for the *transport* mean-free path, we have $l \sim R_c/\Delta^2$ (in two dimensions). Then we have $S_m \sim N'\epsilon_1^2/\Delta^2 \sim l(\ln Q)^{4/3}/(kR_c^2 m^{1/3})$.

Calculations.—Now we briefly sketch an analytic calculation which supports the above qualitative estimates and enables us to obtain the precise numerical coefficients. We consider the scalar wave equation,

$$\nabla^2\Psi + k_0^2\delta\epsilon(\boldsymbol{r})\Psi = -k_0^2\epsilon\Psi, \tag{2}$$

where $\delta\epsilon(\boldsymbol{r})$ is the fluctuating part of the dielectric constant. For a Gaussian disorder, with rms Δ, the probability W of a certain realization $\delta\epsilon(\boldsymbol{r})$ is

$$\ln W = -\frac{1}{2\Delta^2}\iint d\boldsymbol{r}\, d\boldsymbol{r}'\, \delta\epsilon(\boldsymbol{r})\, \delta\epsilon(\boldsymbol{r}')\kappa(\boldsymbol{r} - \boldsymbol{r}'), \tag{3}$$

where the kernel $\kappa(\boldsymbol{r} - \boldsymbol{r}')$ is the inverse of the correlator $K(\boldsymbol{r} - \boldsymbol{r}')$, defined as

$$\langle\delta\epsilon(\boldsymbol{r})\delta\epsilon(\boldsymbol{r}')\rangle = \Delta^2 K(\boldsymbol{r} - \boldsymbol{r}'). \tag{4}$$

We search for a realization $\delta\epsilon(\boldsymbol{r})$, which produces the required solution of the wave equation, i.e., the wave-guided mode $\Psi(\rho, \theta) = (2\pi\rho)^{-1/2}\chi_m(\rho)\exp(im\theta)$, where $\chi_m(\rho)$ is the radial function. The required $\delta\epsilon(\rho, \theta)$ should be azimuthally symmetric, i.e., depend only on the radius ρ, and nonzero within the relatively narrow interval $\sim w$ (see Fig. 1a). On substitution of $\Psi(\rho, \theta)$ into Eq. (2), we obtain the following equation for χ_m:

$$\frac{d^2\chi_m}{d\rho^2} - \frac{m^2 - 1/4}{\rho^2}\chi_m + \delta\epsilon(\rho)k_0^2\chi_m = -\epsilon k_0^2\chi_m. \tag{5}$$

Since χ_m decays rapidly (i.e., at a distance $w \ll \rho_0$) away from ρ_0, we can introduce a coordinate $x = \rho - \rho_0$ and replace Eq. (5) by a one-dimensional equation ($-\infty < x < \infty$),

$$\hat{L}\chi_m = \frac{d^2\chi_m}{dx^2} + \delta\epsilon k_0^2\chi_m = \epsilon_1 k_0^2\chi_m, \tag{6}$$

where $\epsilon_1 = (\frac{m}{k_0\rho_0})^2 - \epsilon$. The shape of the required realization $\delta\epsilon(x)$ and its probability are found using the standard optimal fluctuation method [23,24]. Large m, in fact, simplifies matters since, due to the (narrow) ring-shaped geometry, we can replace $|\boldsymbol{r} - \boldsymbol{r}'|$ by $[(\rho - \rho')^2 + 4\rho_0^2\sin^2(\theta/2)]^{1/2}$ (θ being the angle between $\boldsymbol{r}$ and

$\mathbf{r}'$), to obtain a one-dimensional kernel $K_0(x - x') = \int_{-\infty}^{\infty} dy\, K(\sqrt{(x - x')^2 + y^2})$.

To proceed further, we have to specify the correlator, K. We have chosen the Gaussian form $K(\rho) = \exp(-\rho^2/R_c^2)$. We also adopt a simplified version of the optimal fluctuation method. Namely, we search for $\delta\epsilon$ and χ_m in the Gaussian form, $\delta\epsilon(x) = \epsilon_2 \exp(-\gamma_2 x^2)$ and $\chi_m(x) = \exp(-\gamma_1 x^2)$. In fact, this form of χ_m allows one to cover the entire range of correlation radii, from "white noise" ($R_c \to 0$) to the limit of a smooth disorder ($k_0 R_c \gg 1$). Indeed, for large R_c this form becomes exact [23]. In the opposite limit, $R_c \to 0$, using the above trial χ_m instead of exact $\chi_m \propto 1/\cosh(\gamma_1^{1/2} z)$ [24] leads to the overestimate of S_m by a factor $(\pi/3)^{1/2} \approx 1.023$. To find the variational parameters ϵ_2, γ_1, and γ_2, we minimize $|\ln W|$, defined by Eq. (3), under the constraint

$$\frac{\langle \chi_m \hat{L} \chi_m \rangle}{\langle \chi_m \chi_m \rangle} = \epsilon_1 k_0^2 = \left(\frac{3 \ln Q}{2m}\right)^{2/3} \epsilon k_0^2 . \tag{7}$$

As compared to the qualitative consideration, the factor $3/2$ in the second identity accounts for the linear shape of the "tunneling barrier" in the domain of large $x \sim d = \epsilon_1 m/(2\epsilon k)$ (see Fig. 1a). Minimization of $|\ln W|$ can be carried out analytically, yielding $|\ln W|_{\min} = S_m$, where

$$S_m = 2^4 3^{-3/2} \pi^{1/2} m \left(\frac{\epsilon_1^3}{\epsilon}\right)^{1/2} \frac{\Phi(\epsilon_1^{1/2} k_0 R_c)}{(\Delta k_0 R_c)^2} , \tag{8}$$

where $\epsilon_1 = \epsilon(3 \ln Q/2m)^{2/3}$. The analytical expression for the function $\Phi(u)$ is the following:

$$\Phi(u) = \frac{3^{3/2}}{2^6} \frac{(5 + \sqrt{9 + 16u^2})^{5/2}}{(3 + \sqrt{9 + 16u^2})^{3/2}} . \tag{9}$$

It is shown in Fig. 2. Recall that we are interested in the density of random resonators at a *given* value of kl. The remaining task is to express the transport mean-free path in terms of Δ and R_c. With $K(\rho) = \exp(-\rho^2/R_c^2)$, the expression simplifies in two limits,

$$kl|_{k_0 R_c \ll 1} = \frac{4\epsilon}{\pi (k_0 R_c \Delta)^2} , \qquad kl|_{k_0 R_c \gg 1} = \frac{4\epsilon^{5/2} k_0 R_c}{\pi^{1/2} \Delta^2} . \tag{10}$$

We are now able to specify the numerical coefficients in the qualitative derivation. For the short range case, $R_c \to 0$, we have $\Phi(u) \to 1$. Then, combining Eqs. (8) and (10), we obtain

$$S_m(k_0 R_c \ll 1) = 2\left(\frac{\pi^3}{3}\right)^{1/2} kl \ln Q . \tag{11}$$

To trace the change of S_m with increasing R_c, it is convenient, after using Eq. (10), to present Eq. (8) in the form

$$\frac{S_m(k_0 R_c > 1)}{S_m(k_0 R_c \ll 1)} = \frac{\Phi(\epsilon_1^{1/2} k_0 R_c)}{\pi^{1/2} (\epsilon^{1/2} k_0 R_c)^3} . \tag{12}$$

It is seen from Eq. (12) that S_m falls off rapidly with increasing R_c. In the domain $k_0 R_c > 1$, but $\epsilon_1^{1/2} k_0 R_c \lesssim 1$, we have $\Phi \approx 1$, so that $S_m \propto (k_0 R_c)^{-3}$. For larger R_c we have $\Phi(u) \propto u$. In this domain S_m decreases slower with R_c: $S_m \propto (k_0 R_c)^{-2}$. We emphasize that Eqs. (11) and (12) apply *for a given* kl value, so that the decrease of S_m with R_c *leaves the backscattering cone unchanged.*

Discussion.—Equation (11) quantifies the effectiveness of trapping of light in a random medium with pointlike scatterers. It follows from Eq. (11) that the likelihood of a high-Q cavity is really small. Indeed, even for rather strong disorder, $kl = 5$, the exponent, S_m, in the probability of having a cavity with a quality factor $Q = 50$ is close to $S_m = 120$. We emphasize that, in the two-dimensional case under consideration, this exponent does not depend on m and, thus, on the cavity radius $\rho_0 = m/\epsilon^{1/2} k_0$. To estimate the degree to which a finite size of scatterers ($\sim R_c$) improves the situation, we choose $k_0 R_c \approx 2$, which already corresponds to the limit $k_0 R_c \gg 1$ in Eq. (10), but still allows one to set $\Phi = 1$. Then for $Q = 50$, $kl = 5$ we obtain $S_m \approx 1.1$, suggesting that the resonators with this Q are quite frequent. In the latter estimate we have set $\epsilon = 4$.

A natural question to address is what values of Q are feasible for a given kl and $k_0 R_c$. To address this question, we inspect the argument $u = \epsilon_1^{1/2} k_0 R_c$ of the function Φ. It can be presented in the form $u = \epsilon^{1/2} k_0 R_c [\frac{3 \ln Q}{2m}]^{1/3}$. Since $\Phi(u)$ increases monotonically (see Fig. 2), the latter expression suggests that Q can be increased at the expense of larger m values. In the example considered above, in order to keep Φ smaller than, for example 1.5, m should be bigger than 50. However, due to slow dependence $u \propto m^{-1/3}$, we get a rather small value $S_m \approx 3$ for m as small as $m = 15$. Certainly, the allowed values of m are limited from above. This limitation originates from "vulnerability" of waveguiding to the fluctuations of the dielectric constant around optimal ringlike distribution. The dangerous fluctuations are those that enhance the evanescent leakage.

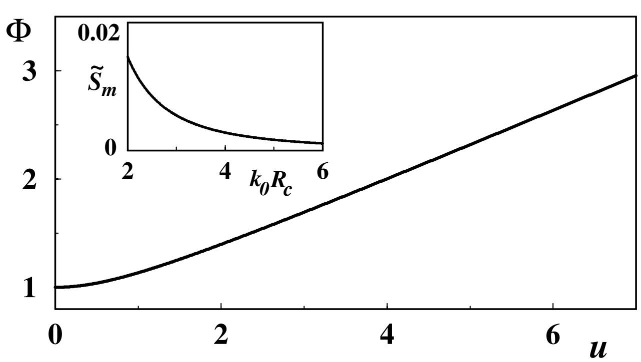

FIG. 2. Dimensionless function $\Phi(u)$ defined in Eq. (9) is plotted. Inset: Normalized modulus of the log density of random resonators, $\tilde{S}_m = S_m(k_0 R_c)/S_m(0)$, calculated from Eq. (12) for $\epsilon = 4$, $Q = 50$, and $m = 15$ is plotted versus the dimensionless correlation radius, $k_0 R_c$.

Note that these fluctuations do not affect the main exponent S_m in the density of resonators. It is obvious that the bigger the area $A_m = 2\pi\rho_0 d \propto m^{4/3}$, responsible for evanescent leakage (see Fig. 1a), the harder it is to "protect" the waveguiding. Since the fluctuations $\delta\epsilon(\boldsymbol{r})$ have the spatial scale R_c, the probability that the waveguiding "survives" can be roughly estimated as $\exp(-A_m/R_c^2)$. The condition that the exponent A_m/R_c^2 does not dominate over the principal exponent S_m can be cast into $m \lesssim (\epsilon kl)^{3/5}(\ln Q)^{2/5}$. For the example $kl = 5$ and $Q = 50$, addressed above, we get $m \lesssim 10$. Rigorous calculation of the "survival probability" is of the same complexity as calculation of the prefactor [25] in the functional integral.

Conclusion.— In the present paper, we provided a quantitative theory of random resonators that substantiates the intuitive image [1,12] of a resonant cavity as a closed-loop trajectory of a light wave bouncing between the pointlike scatterers. The intuitive picture in [1,12] assumed that light can propagate along a loop of scatterers by simply being scattered from one scatterer to another. Such a picture, however, is unrealistic due to the scattering out of the loop [16]. We have demonstrated that the scenario of light traveling along closed loops can be remedied. In our picture the "loops," i.e., the random resonators, can be envisaged as rings with dielectric constant larger than the average value. On a microscopic level, these resonators correspond to certain arrangements of scatterers (grains). The main point, however, is that a resonator acts as a *single entity*: Only the coherent multiple scattering of light by *all* the scatterers in the resonator can provide trapping. We also point out that correlations in the fluctuating part of the dielectric constant (due to finite grain size) highly facilitate trapping.

The effectiveness of light trapping is expressed by Eq. (12). This expression describes the statistics of the quality factors which determines the distribution of the threshold gain for random lasing. In particular, Eq. (12) yields a quantitative prediction for the decrease of the threshold upon adding disorder to the sample. Shortening of the mean-free path, as the concentration of scatterers increases, can be inferred from the broadening of the backscattering cone [7]. Another quantitative prediction of our theory, which can be tested experimentally, is the decrease of the threshold with the size of the excitation spot, D^2. Since occurrence of a random resonator is a rare event, it is easy to show that the average threshold gain is $\propto \ln D$ with the proportionality coefficient determined by Eq. (11).

Our consideration pertains to the *passive* films; i.e., we neglect the effect of gain on the spatial distribution of the light intensity [2,13,26]. Random resonators considered in the present paper are sparse, so that there is no spatial overlap between the modes of different resonators, the situation opposite to [27,28]. We have also treated scatterers as frequency-independent fluctuations, $\delta\epsilon(\boldsymbol{r})$, of the dielectric constant. The entirely different scenario of the collective mode formation emerges for *resonant* scatterers [29].

Two of the authors, M. E. R. and B. S., acknowledge the hospitality of the University of Köln and of the Institute for Theoretical Physics at UCSB where the parts of this work were completed. This research was supported in part by the National Science Foundation under Grants No. PHY99-07949 and No. DMR-0202790, and by Petroleum Research Fund under Grant No. 37890-AC6.

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