Invited Paper

Coherent Random Lasing: Trapping of Light by the Disorder

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ABSTRACT

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. It suggests that in the situation, when the propagation of light is diffusive *on average*, the coherent feedback can be provided by the sparse disorder configurations that efficiently trap a photon, and thus, serve as random resonators. This scenario of random resonators has been substantiated experimentally by the ensemble averaging of the power Fourier transforms of the random emission spectra. In this paper the current status of the experiment and theory of coherent random lasing is reviewed.

Keywords: Random lasers, random resonators, power Fourier transform

1. INTRODUCTION

Conceptual importance of the recently discovered phenomenon of random lasing (RL) in various disordered materials with optical gain[1,2] is that laser action is due to coherent (amplitude) rather than non-coherent (power) optical feedback. The manifestation of the power feedback, first suggested by Letokhov,[3] is the gain-induced spectral narrowing typical of amplified spontaneous emission (ASE).[4–8] This process can be accurately described within the diffusive picture of light propagation. In contrast, the amplitude feedback manifests itself as a sequence of narrow lines,[1,2] each corresponding to *coherent* emission.[9,10] Clearly, the diffusive description, that neglects coherence and deals with *intensity* distribution, is unable to capture the physics underlying the amplitude feedback.

For strongly scattering media with a small light mean free path, l, of the order of the emission wavelength, λ, the amplitude feedback can be, at least qualitatively, accounted for by the proximity to the Anderson localization.[1,11] This is because a disorder configuration that localizes a photon mode can be viewed as an ideal resonator for light. However, when the disorder is weak, so that the light propagation is diffusive *on average*, the very existence of resonators, that are required to provide the amplitude feedback is not obvious. It was conjectured in[1,12] that the underlying mechanism of lasing was formation of closed loop paths of light with a characterisitic size comparable to λ which serve as resonant cavities. Needless to say, that more quantitative questions, concerning the dependence of the size and density of the resonators in a weakly scattering medium on the parameters of the disorder[13] remains largely unanswered.

Motivated by these experimental puzzles, in Sect. 2 of the present paper we report on our theoretical study of random resonators, which represent a new type of solutions[13,14] of the wave equation in a weakly disordered medium. We dubb these solutions as "almost localized" states. They describe a wave which is confined primarily to a small ring. In an *open* sample, of size L much smaller than the two-dimensional localization length ξ, the almost localized states correspond to sharp resonances, residing in the high-Q ring-shaped cavities, as discussed throughout the paper. However, in a *closed* sample -which would require perfectly reflecting walls- the resonances turn into true eigenstates, whose almost entire weight is located at the rings. In this respect the "almost localized" states differ from the "prelocalized" states, extensively studied in the context of electronic transport.[15–18] We provide a quantitative theory of the almost localized states and the associated random resonators, and point out their relevance for the phenomenon of random lasing. We stress, however, that these random resonators exist already in the *passive* medium, and gain is only needed "to make them visible". Moreover, the resonators are

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"self-formed", in the sense that no sharp features (like Mie scatterers or other "resonant entities") are introduced: the model is developed for a featureless dielectric medium with fluctuating dielectric constant.

Our study substantiates the intuitive image[1,12] of a resonant cavity as a closed-loop trajectory of a light wave bouncing between the point-like 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. 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. The reason why such rings are able to trap the light is that the constituting scatterers act as a *single entity*: only the coherent multiple scattering of light by *all* the scatterers in the resonator can provide trapping. We have also established that correlations in the fluctuating part of the dielectric constant highly facilitate trapping.

In the second part of the paper we review the experiments that provide the experimental evidence for the scenario of random resonanators.

2. THEORY: LIKELIHOOD OF RANDOM RESONATORS

2.1. Almost Localized Modes in Random Media

2.1.1. Basic Relations

In this paper we restrict our consideration to the two-dimensional case (a disordered film). We start from the scalar wave equation

$$\Delta_{\vec{\rho}}\Psi + \left[k^2 - U(\vec{\rho})\right]\Psi = 0, \tag{1}$$

where $\vec{\rho}$ is the in-plane coordinate. For a light wave with frequency ω, traveling in a medium with average dielectric constant ϵ, corresponding expressions for k^2 and $U(\vec{\rho})$ take the form

$$k^2 = \epsilon\left(\frac{\omega}{c}\right)^2 = \epsilon k_0^2\ , \qquad U(\vec{\rho}) = -\delta\epsilon(\vec{\rho})k_0^2 \tag{2}$$

where $\delta\epsilon(\vec{\rho})$ is the fluctuating part of the dielectric constant.

We assume that the flutuations are gaussian, so that the probability of the fluctuation $\delta\epsilon(\vec{\rho})$ is given by

$$\ln P = -\frac{1}{2\mathcal{W}^2}\iint d\vec{\rho}d\vec{\rho}'\,\delta\epsilon(\vec{\rho})\delta\epsilon(\vec{\rho}')\kappa(\vec{\rho}-\vec{\rho}'), \tag{3}$$

where the kernel $\kappa(\vec{\rho}-\vec{\rho}')$ is the inverse of the correlator $K(\vec{\rho}-\vec{\rho}')$

$$\int d\vec{\rho}'\,\kappa(\vec{\rho}-\vec{\rho}')K(\vec{\rho}'-\vec{\rho}_1) = \delta(\vec{\rho}-\vec{\rho}_1). \tag{4}$$

The correlator is defined as

$$\langle\delta\epsilon(\vec{\rho})\delta\epsilon(\vec{\rho}')\rangle = \mathcal{W}^2K(\vec{\rho}-\vec{\rho}'), \quad K(0) = 1, \tag{5}$$

so that $\mathcal{W}$ is the mean square fluctuation of the dielectric constant. Trapping of a light wave is provided by specific fluctuations, $\delta\epsilon(\vec{\rho})$, for which the solution, $\Psi(\vec{\rho})$ of Eq. (1) assumes anomalously large value within the area of the fluctuation. Outside the fluctuation, this solution propagates in a "normal" (diffusive) fashion, so that the "buildup" of $\Psi(\vec{\rho})$ within the fluctuation translates into the long trapping time, $\mathcal{T}$.

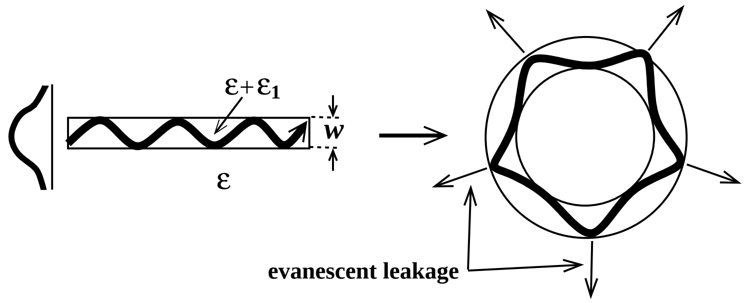

Figure 1. Rationale for the structure of the resonator. Upon wrapping a stripe with enhanced dielectric constant into a ring, a waveguided mode transforms into a whispering-gallery mode.

2.1.2. Trapping Configuration of the Disorder

The key idea concerning the geometry of the trap was proposed by Karpov for trapping acoustic waves[14] and electrons[19] in three dimensions. Below we explain this idea for the two-dimensional geometry.

Suppose that within a certain stripe the effective in-plane dielectric constant of a film is enhanced by some small value $\epsilon_1 \ll \epsilon$. Then such a stripe can play a role of a *waveguide*, i.e., it can capture a transverse mode, as it is illustrated in Fig. 1. There is no threshold for such a waveguiding, which means that the transverse mode will be captured even if the width of the stripe is small. Now, in order to form a trap, one has to roll the stripe into a ring. Upon this procedure, the mode propagating along the waveguide transforms into a whispering-gallery mode of a ring. An immediate consequence of the curving of the waveguide is emergence of the evanescent leakage - the optical analog of the under-the-barrier tunneling in quantum mechanics (see Fig. 2). This leakage is responsible for a finite lifetime, $\mathcal{T}$, of the whispering-gallery mode.

Due to the azimuthal symmetry, the modes of the ring are characterized by the angular momentum, m. Denote by $\mathcal{N}_m(kl, \omega\mathcal{T})$ the areal density of traps which capture the light wave for a long period of time $\mathcal{T} \gg \omega^{-1}$ in the film with a transport mean free path l. In the diffusive regime, $kl > 1$, the density $\mathcal{N}_m(kl, \omega\mathcal{T})$ is exponentially small. In this domain $\mathcal{N}_m(kl, \omega\mathcal{T})$ can be presented as

$$\mathcal{N}_m(kl, \omega\mathcal{T}) \propto \exp\left[-S_m(kl, \omega\mathcal{T})\right] \ , \tag{6}$$

with $S_m(kl, \omega\mathcal{T}) \gg 1$. The latter condition justifies the application of the optimal fluctuation approach[20,21] to the calculation of S_m.

2.1.3. Optimal Fluctuation Approach

We search for fluctuations of a type shown in Fig. 2, *i.e.*, $\delta\epsilon$ is azimuthally symmetric (depends only on the radius, ρ) and is non-zero within the relatively narrow ring of width $w \ll \rho_0$, where ρ_0 is the radius corresponding to the middle of the ring (see Fig. 2a). For such a fluctuation the wave equation for field distribution corresponding to the angular momentum, m, has a general solution of the form

$$\Psi(\rho, \theta) = (2\pi\rho)^{-1/2}\chi_m(\rho)\exp(im\theta), \tag{7}$$

where $\chi_m(\rho)$ is the radial function. The fact that $\delta\epsilon$ is non-zero within a narrow interval of ρ around ρ_0 allows to simplify the equation for $\chi_m(\rho)$ to

$$\hat{L}\chi_m = \frac{d^2\chi_m}{dx^2} + \delta\epsilon\ k_0^2\chi_m = \epsilon_1\ k_0^2\chi_m \ , \tag{8}$$

where $x = \rho - \rho_0$. The "eigenvalue" in the r.h.s. of Eq. (8) is defined as $\epsilon_1 = \left(\frac{m}{k_0\rho_0}\right)^2 - \epsilon$. It is independent of x by virtue of the condition $\rho_0 \gg w$.

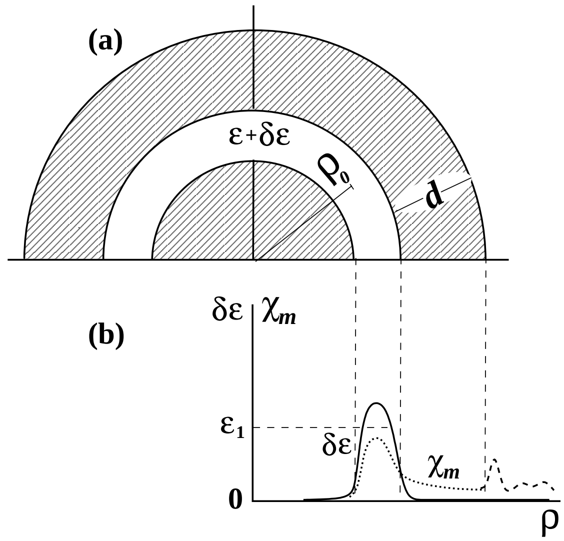

Figure 2. (a) The structure of a two-dimensional resonator is illustrated schematically; only a half of the ring-shaped waveguide (blank region) is shown. (b) Optimal fluctuation of the dielectric constant, $\delta\epsilon(\rho)$ (solid line), and the corresponding field distribution (dotted line) are shown. Dashed line outside the shaded region of a width, d, illustrates the evanescent leakage.

The next step is to find the "most probable" distribution $\delta\epsilon(x)$ for a given trapping time $\mathcal{T}$. This is equivalent to finding the most probable $\delta\epsilon(x)$ for a given eigenvalue ϵ_1, since, with logarithmic accuracy, ϵ_1 uniquely defines $\mathcal{T}$ as we demonstrate below

Lifetime of a Trapped Mode. It is obvious that at large distances, $\rho \gg \rho_0$ (see Fig. 2b), the behavior of χ_m is oscillatory, $\chi_m \propto \exp(i\epsilon^{1/2}k_0\rho)$, manifesting that the waveguided mode of a ring has a finite lifetime. In other words, due to evanescent leakage, the eigenvalue, ϵ_1, in Eq. (8) has an imaginary part, $\tilde{\epsilon}_1$. The quality factor is inversely proportional to $\tilde{\epsilon}_1$, namely, $\omega\mathcal{T} = \epsilon/\tilde{\epsilon}_1$. The leading contribution to $\tilde{\epsilon}_1$ comes from the region of a width, d, adjacent to the waveguide (see Fig. 2a). To find d we have to take into account that the r.h.s in Eq. (8) does in fact depend on x. This is because the precise form of the r.h.s is not $\left(\frac{m}{\rho_0}\right)^2 - \epsilon k_0^2$, but rather $\left(\frac{m}{\rho_0+x}\right)^2 - \epsilon k_0^2$. In the region outside the waveguide, when $x \gg 1/\epsilon_1^{1/2}k_0$ (but still $x \ll \rho_0$) Eq. (8) takes the form

$$\frac{d^2\chi_m}{dx^2} = \epsilon_1 k_0^2\left(1 - \frac{x}{d}\right)\chi_m\ , \tag{9}$$

where the width of the decay region is given by $d = \epsilon_1 k_0^2 \rho_0^3/2m^2$. Equation (9) is of Airy-type. Semiclassical calculations with exponential accuracy yields for the rate of evanescent leakage $\tilde{\epsilon}_1 \propto \exp[-(2m/3)(\epsilon_1/\epsilon)^{3/2}]$, and hence

$$\ln(\omega\mathcal{T}) = \frac{2m}{3}\left(\frac{\epsilon_1}{\epsilon}\right)^{3/2} . \tag{10}$$

Eq. (10) is the sought relation between the evanescent lifetime and the magnitude of the fluctuation, ϵ_1.

Once the lifetime is expressed through ϵ_1, the search for the most probable $\delta\epsilon(x)$ reduces to a standard problem, for which the optimal fluctuation approach[20,21] prescribes minimization of auxiliary functional

$$\Xi\{\delta\epsilon\} = \ln P - \lambda\left[\langle\chi_m|\hat{L}|\chi_m\rangle - \epsilon_1 k_0^2\langle\chi_m|\chi_m\rangle\right], \tag{11}$$

where λ is the Lagrange multiplier. In Eq. (11) $\delta\epsilon$ and χ should be varied independently. It is convenient to choose $\lambda = 2\pi\rho_0/\mathcal{W}^2k_0^2$. Then the minimization of the auxiliary functional Eq. (11) yields

$$\delta\epsilon(x) = \int dx_1 K_0(x - x_1)\chi_m^2(x_1). \tag{12}$$

The above expression reminds the standard result[20] for a correlated random potential. The difference is, however, that, due to the angular integration in Eq. (3), the kernel in Eq. (12) is given by a function K_0, which is related to the correlator, $K(\vec{\rho})$, as $K_0(x_1 - x_2) = \int_{-\infty}^{\infty} dy K(\sqrt{(x_1-x_2)^2 + y^2})$. A natural x-scale for the eigenmode χ_m is $(\epsilon_1^{1/2} k_0)^{-1}$. Thus, we present the outcome of the optimal fluctuation approach in terms of a dimensionless variable $z = \epsilon_1^{1/2} k_0 x$.

$$S_m = \frac{\pi \rho_0}{\epsilon_1 k_0^2 \mathcal{W}^2} \iint dz_1 dz_2 \chi_m^2(z_1) \chi_m^2(z_2) K_0(z_1 - z_2) \ , \tag{13}$$

where the dimensionless equation for the function χ_m reads

$$\frac{d^2 \chi_m(z)}{dz^2} + \frac{\chi_m(z)}{\epsilon_1^{3/2} k_0} \int dz_1 K_0(z - z_1) \chi_m^2(z_1) = \chi_m(z) \ . \tag{14}$$

2.1.4. Variational Solution

A very accurate analytical solution of Eq. (14) can be obtained by employing the variational approach. The function $\chi_m(z)$ corresponds to the minimum of the following functional

$$\mathcal{F}\{\chi\} = \left(\int dz \chi_m^2(z) \right)^{-1} \left[\int dz \left(\frac{d\chi_m(z)}{dz} \right)^2 - \frac{1}{2\epsilon_1^{3/2} k_0} \int \int dz_1 dz_2 \chi_m^2(z_1) K_0(z_1 - z_2) \chi_m^2(z_2) \right]. \tag{15}$$

To proceed further we have to specify the correlator, K. We have chosen the Gaussian form $K(\rho) = \exp\left(-\rho^2/R_c^2\right)$. The form of the function $\chi_m = \mathcal{A}(R_c) \exp[-\zeta(R_c) z^2]$ allows 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. In the opposite limit, $R_c \to 0$, using the above trial function instead of the exact solution $\chi_m \propto 1/\cosh(z)$ leads to the overestimate of S_m by a factor $(\pi/3)^{1/2} \approx 1.023$. The parameter $\mathcal{A}(R_c)$ and $\zeta(R_c)$ of the trial function can be found analytically. Evaluation of Eq. (13) reduces to the Gaussian integration which for a given $m \approx \epsilon^{1/2} k_0 \rho_0$ yields

$$S_m = 2^4 3^{-3/2} \pi^{1/2} m \left(\frac{\epsilon_1^3}{\epsilon} \right)^{1/2} \frac{\Phi(\epsilon_1^{1/2} k_0 R_c)}{(\mathcal{W} k_0 R_c)^2} \ , \tag{16}$$

where the dimensionless function $\Phi(u)$ is defined as

$$\Phi(u) = \frac{3^{3/2}}{2^6} \frac{(5 + \sqrt{9 + 16u^2})^{5/2}}{(3 + \sqrt{9 + 16u^2})^{3/2}}. \tag{17}$$

This function is plotted in Fig. 3. It is seen that at $u > 1$ it increases roughly linearly from $\Phi(0) = 1$ with a slope ≈ 0.3. The analysis of the result Eq. (16) is given in the next subsection.

2.1.5. Likelihood of Trapping as a Function of the Scattering Strength

The remaining task is to express the transport mean free path in terms of $\mathcal{W}$ and R_c. Using the golden rule, the product kl can be expressed through the correlator, K, as follows

$$\begin{aligned} (kl)^{-1} &= \frac{\mathcal{W}^2}{4\pi\epsilon^2} \int dq \ d\phi \ q^3 \delta(q^2 + 2kq\cos\phi) \int d^2\rho \ K(\rho) \exp(iq\rho) = \\ &= \frac{2\mathcal{W}^2 k_0^2}{\epsilon} \int_0^{\pi/2} d\alpha \ \sin^2\alpha \int_0^{\infty} d\rho \ \rho \ K(\rho) \ J_0(2k\rho\sin\alpha), \end{aligned} \tag{18}$$

where J_0 is the Bessel function of zero order. With a gaussian correlator $K(\rho) = \exp(-\rho^2/R_c^2)$ the above integral can be evaluated analytically. Substituting this form into (18), we obtain

$$(kl)^{-1} = \pi \left(\frac{\mathcal{W} k_0 R_c}{2\epsilon^{1/2}} \right)^2 F\left(\frac{k^2 R_c^2}{2} \right), \tag{19}$$

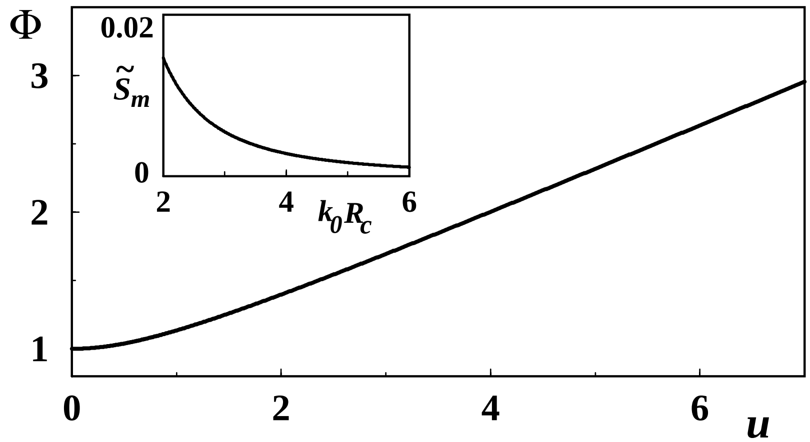

Figure 3. Dimensionless function $\Phi(u)$ defined in Eq. (17) is plotted. Inset: Normalized modulus of the log-density of random resonators, $\tilde{S}_m = S_m(k_0R_c)/S_m(0)$, calculated from Eq. (24) for $\epsilon = 4$, $Q = 50$, and $m = 15$ is plotted versus the dimensionless correlation radius, k_0R_c.

where the dimensionless function F is defined as

$$F(x) = e^{-x}\Big[I_0(x) - I_1(x)\Big]. \tag{20}$$

Here I_0 and I_1 are the modified Bessel functions of zero and first order, respectively. The above expression simplifies in two limits corresponding to the short-range and to the smooth disorder.

$$kl = \frac{4\epsilon}{\pi(k_0R_c\mathcal{W})^2} \quad \text{for} \quad k_0R_c \ll 1 \ ; \tag{21}$$

$$kl = \frac{4\epsilon^{5/2}k_0R_c}{\pi^{1/2}\mathcal{W}^2} \quad \text{for} \quad k_0R_c \gg 1 \ . \tag{22}$$

Eq. (21) allows us to cast the result Eq. (16) for the exponent in the density of traps into a remarkably simple form. Indeed, upon expressing ϵ_1 from Eq. (10) and $\mathcal{W}$ from Eq. (21), and substituying both into Eq. (16), we obtain

$$S_m(k_0R_c \ll 1) = 2\left(\frac{\pi^3}{3}\right)^{1/2} kl|\ln\omega\mathcal{T}| \ , \tag{23}$$

Equation (23) quantifies the effectiveness of trapping of light in a random medium with short-range flutuations of the dielectric constant. It follows from Eq. (23) that the likelihood of the efficient trap is really small. Indeed, even for rather strong disorder, $kl = 5$, the exponent, S_m, in the probability of having a trap with $\omega\mathcal{T} = 50$ is close to $S_m = 120$. We emphasize that in two-dimensional case under consideration, this exponent does not depend on m and, thus, on the radius $\rho_0 = m/\epsilon^{1/2}k_0$ of the ring-shape fluctuation trapping the light.

The effectiveness of trapping is strongly enhanced when disorder is correlated, i.e., when the correlation radius, R_c, exceeds the wavelength, $2\pi/k_0$. To see this, consider the ratio of the exponents S_m for the cases of the long-range and short-range disorder that correspond to the *same* transport mean free path, l. From Eq. (16) we obtain

$$\frac{S_m(k_0R_c > 1)}{S_m(k_0R_c \ll 1)} = \frac{\Phi(\epsilon_1^{1/2}k_0R_c)}{\pi^{1/2}(\epsilon^{1/2}k_0R_c)^3} \ . \tag{24}$$

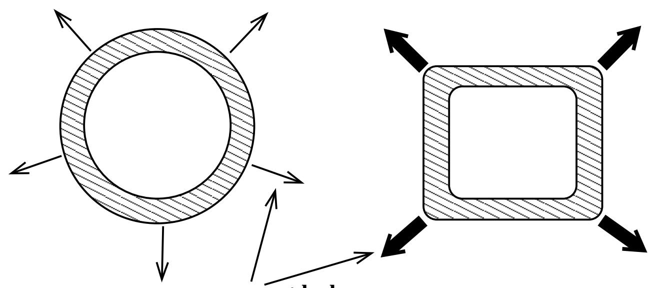

Figure 4. This drawing illustrates the optimal character of the ring-shape resonator.

We see that, with increasing R_c, the exponent, S_m, falls off roughly as R_c^{-3}, making the formation of the efficient traps exponentially more likely than in the case of the short-range disorder. This is illustrated in the inset in Fig. 3, where the ratio Eq. (24) is plotted. To estimate quantitatively the degree to which finite correlation radius facilitates the trapping, we choose $k_0 R_c \approx 2$, which already corresponds to the limit $k_0 R_c \gg 1$ in Eq. (22), but still allows to set $\Phi = 1$. Then for $\omega\mathcal{T} = 50$, $kl = 5$ we obtain $S_m \approx 1.1$, suggesting that the resonators with this $\omega\mathcal{T}$ are quite frequent. In the latter estimate we have set $\epsilon = 4$. In conclusion of this subsection we would like to emphasize that the likelihood of traps increases with correlation radius for a *given* value of the scattering strength kl.

2.2. Qualitative discussion

2.2.1. Why Rings?

The answer to this question is illustrated in Fig. 4. The distinguishing property of a ring is that the local curvature radius is the same at each point. Upon any deviation from the ring geometry, the curvature in a certain region of the fluctuation would be higher than in all other regions. Since the evanescent losses are governed by this curvature, the quality factor of the resonator would be determined exclusively by this region (see Fig. 4), so that the remaining low-curvature part would be "unnecessary", in the sense, that a ring with a radius corresponding to the maximal curvature would have the same quality factor as a square in Fig. 4 but significantly higher probability of formation. It is also quite obvious that, for the purpose of supporting a wave-guided mode of the whispering-gallery type, a ring is much superior to a disk of the same radius: indeed, the internal area of the disk remains unused in the guiding process, whereas a heavy penalty in terms of probability is paid in creating this area.

2.2.2. Why Smooth Disorder Facilitates Trapping?

At the qualitative level, the enhancement of the probability of formation of the cavity with increasing R_c can be understood for a toy model of the disorder, illustrated in Fig. 5. Suppose that all the disks, that model the scatterers, are identical. Then R_c scales with the radius of the disk, R. Since the disks cannot interpenetrate, the ring-shaped cavity corresponds to their arrangement in the form of a necklace. The probability of formation of such a cavity can be estimated as follows. Suppose that a sector, $\delta\phi$, is "allocated" for a single disk. The probability to find a disk within this sector, at the distance ρ_0 from the center , is $\sim n(\rho_0 \delta\phi)^2$, where n is the concentration of the disks. Thus, the probability of formation of the necklace is $\exp\left[-\frac{2\pi}{\delta\phi}\ln\left(\frac{1}{n\rho_0^2(\delta\phi)^2}\right)\right]$, where $\frac{2\pi}{\delta\phi}$ is the number of sectors. It is obvious that if a necklace is "loose", the quality factor of the corresponding cavity would be low. In order for Q to be high, neighboring disks must almost touch each other. This implies that $\delta\phi \approx 2R/\rho_0$. Then the above estimate for probability takes the form $\exp\left[-\frac{\pi\rho_0}{R}\ln\left(\frac{1}{f}\right)\right]$, where $f = n\pi R^2$ is the filling fraction. This probability increases exponentially with R, i.e., with R_c, reflecting the fact that, for a given ρ_0, the number of disks to be arranged is smaller when R is larger. The above estimate was based on the assumption that the positions of the disks are uncorrelated, i.e., $f \ll 1$. We have used the model of *hard* disks as an easiest illustration of the role of R_c.

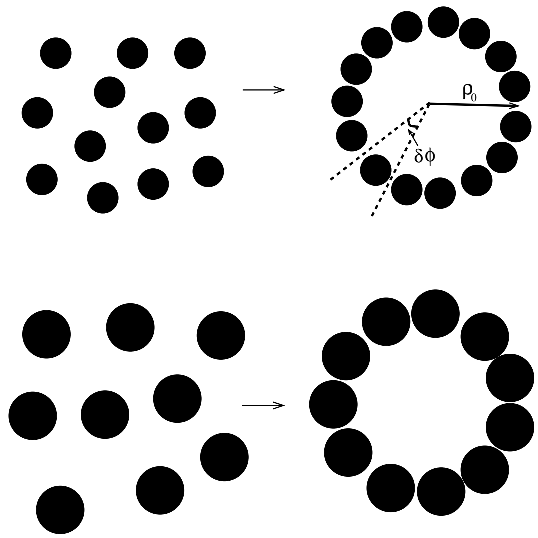

Figure 5. A schematic illustration explaining why larger correlation radius for a fixed filling fraction facilitates trapping. A sector, shown with dashed lines, illustrates the tolerance in the arrangement of disks into a necklace.

2.2.3. "Vulnerability" of the Ring-Shaped Cavities.

The value S_m given by Eq. (16), which was derived within the optimal fluctuation approach, is the exponent in the probability of formation of an *ideal* ring. Obviously, any actual disorder realization is not ideal, in the sense, that actual distribution of dielectric constant differs from the optimal. For the same reason, the probability of formation of ideal necklace of the type shown in Fig. 5 is zero. In order for the probability to be finite, we should allow a certain *tolerance* in the positions of the centers of the disks, as illustrated in Fig. 5. In the conventional applications of the optimal fluctuation approach,[22] deviations from the optimal distribution do not affect the value of the exponent, S_m. However, in application to the random cavities, we have searched for the fluctuation which is optimal *for a given trapping time*, $\omega^{-1}Q$. In this particular application, a "normal" gaussian deviation from the optimal geometry can have a catastrophic effect on trapping by scattering the light wave out of the whispering-gallery trajectory. This scattering is discussed below.

Scattering within the plane. Two-dimensional picture adopted throughout this paper, implies that electromagnetic field is confined within a thin film in the z-direction. This confinement results from the fact that the average dielectric constant of the film is higher than in the adjacent regions. Then the filed distribution, $\mathcal{E}_0(z)$, along z corresponds to a transverse waveguide mode. For a given frequency, ω, the almost localized state on a ring can be destroyed due to the scattering into states with the *same* distribution of the field in the z-direction, which propagate freely along the film. More precisely, the almost localized state with a given angular momentum m, which is protected from the outside world by the centrifugal barrier, can be scattered out to the continua of states with smaller m's, for which there is no barrier. It is essential to estimate the lifetime, τ, with respect to these scattering processes and to verify that it is feasible to have τ larger than the prescribed trapping time, $\omega^{-1}Q$, so that the almost localized state is not destroyed.[23]

The effect can be also illustrated with the model of randomly positioned hard disks (Fig. 5), although the disorder in this model is non-gaussian. It is seen from Fig. 5, that spacings between the rings, which are due to tolerance, open a channel for the light escape, that is different from evanescent leakage. A *typical* lifetime with respect to such an escape is quite short, i.e., even a small tolerance, which affects weakly the exponent in the probability of the cavity formation, seems to be detrimental for trapping. At this point we emphasize that, in calculation of the scattering rate out of the whispering-gallery trajectory, the disks constituting the necklace must be considered as a *single entity*. As a result, for a *given* configuration of the disks, the rate of scattering out caused by a *single* disk must be multiplied by the following *form-factor*

$$\mathcal{F} = \int \frac{d\varphi_{\vec{k}}}{2\pi} \left| \sum_i \exp\left(i\vec{k}\vec{\rho}_i\right) \right|^2 = \sum_{i,j} J_0\left(k|\vec{\rho}_i - \vec{\rho}_j|\right), \tag{25}$$

where $\vec{\rho}_i$ is the position of the center of the i-th disk in the necklace. The form-factor, $\mathcal{F}$, is the sum of $\left(\frac{\pi\rho_0}{R}\right)^2 \gg 1$ terms. Out of this number, $(kR)^{-1}\left(\frac{\pi\rho_0}{R}\right)$ terms (for $kR < 1$) and $\frac{\pi\rho_0}{R}$ terms (for $kR > 1$), for which $k|\vec{\rho}_i - \vec{\rho}_j| < 1$, are close to unity. The portion of these terms is small. Other terms have random sign. This leads us to the important conclusion that, for certain realizations of the necklace in Fig. 5, the form-factor can take anomalously small values. For these realizations the quality factor will be still determined by the evanescent leakage. The "phase volume" of these realizaions is exponentially small and depends strongly on the model of the disorder.

Scattering out of the plane. Compared to the previous case, two modifications are in order. Firstly, since the final state of the scattered cavity mode is a plane wave with the wave vector pointing in a certain direction within the solid angle 4π, the expression Eq. (25) for the form-factor should be replaced by

$$\tilde{\mathcal{F}} = \sum_{i,j} \frac{\sin\left(k_0|\vec{\rho}_i - \vec{\rho}_j|\right)}{k_0|\vec{\rho}_i - \vec{\rho}_j|}. \tag{26}$$

Secondly, for $kR > 1$, i.e., when the disorder is smooth, scattering out of the plane that is caused by a single disk, requires a large wave vector transfer, $\sim k$. Thus, the corresponding rate is suppressed as compared to the in-plane scattering.

3. EXPERIMENT: CONFIRMATION OF THE RANDOM RESONATORS SCENARIO

It is crucial to establish experimentally to what extent the scenario of random resonators is viable to *various* weakly scattering media. The ability to test this scenario relies on the fact that the occurrence of a random resonator is a *rare event*, which corresponds to a certain disorder configuration that is capable to capture light for a long time, $\mathcal{T}$. Then it immediately follows that for a given $\mathcal{T}$, there exists a *dominant* disorder configuration that can provide the trapping. In other words, all random resonators with a given $\mathcal{T}$, are approximately *identical*. Since for a weakly scattering medium, light trapping is feasible only within spatial scales much larger than the wavelength, the identity of different resonators implies almost equal *pathlengths* $L_0 \gg \lambda$ of light in all resonators. This suggests the following strategy[24–26] for testing the scenario of the random resonators in random lasing.

Firstly, to overcome loss, the vale of L_0 is determined by the threshold condition. Secondly, the modes of a large-size resonator are close and almost even-spaced in frequency. If the width of the gain spectrum exceeds the spacing between the modes, then a single resonator would contribute more than one mode to the laser emission. As a result a "hidden" order would be present in random lasing spectra of the random media. Discerning the hidden order would provide the evidence for random resonators in random lasing. To unravel the hidden order, we have employed, following our earlier studies,[24–26] the ensemble-averaging of the power Fourier transform (PFT) of the random lasing emission spectra. Note, that a similar procedure has been previously used in electron transport. In particular, the analysis of the PFT of magnetoresistance fluctuations of metallic mesoscopic rings allowed to reveal the underlying fundamental period, h/e.[27] Ensemble-averaging of PFT of magnetoresistance in a semiconductor ring allowed to unravel even more delicate feature – the splitting of the fundamental period caused by spin-orbit coupling.[28] Our analysis of the averaged PFT spectrum of random lasing indicates that rather than converging into a smooth curve similar to that of regular ASE, it contains a sharp, well-resolved Fourier component and its harmonics, which are characteristic of a well-defined laser resonator.

We have studied coherent random lasing in three *different* organic media with optical gain in the weak scattering regime. The PFT averaging was done over the sample area or illumination time, depending on the specific organic gain medium. Three different media studied are: (i) a π-conjugated polymer films of poly (dioctyloxy) phenylene vinylene (DOO-PPV),[29] (ii) a laser dye (Rhodamine 6G [R6G,[30]]) in methanol infiltrated into a synthetic opal,[31] and (iii) a methanol suspension of TiO_2 balls in R6G enclosed in a cuvette. A homemade

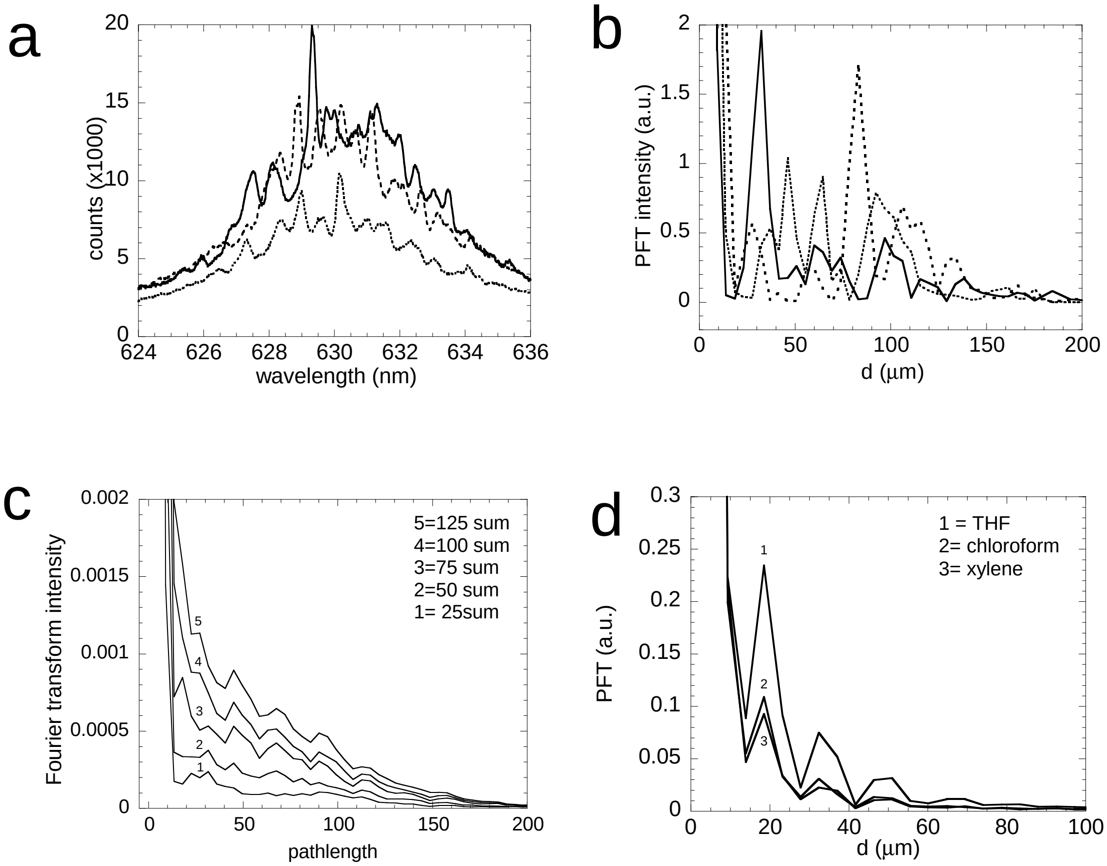

Figure 6. Ensemble average power Fourier transform spectroscopy of random lasing in polymer films. (a) Three random lasing emission spectra of a DOO-PPV polymer film spin casted from a toluene solution, which were collected from three different illuminated stripes on the film. (b) The PFT spectra of the emission spectra shown in (a). (c) Ensemble-averaged PFT spectra of random lasing emission spectra such as in (a), which were averaged from different illuminated stripes over the film area. The numbers in the upper right corner denote the number of random lasing spectra collected in the averaging process. (d) Ensemble-averaged PFT spectra of 125 different random lasing emission spectra of DOO-PPV films spin casted from solutions of different solvents.

coherent backscattering (CBS) apparatus[32] was used to measure the light mean free path, l. Each of the obtained l for the three samples falls in the region of $l \gg \lambda$; in fact l/λ ranged between 300 and 5000.[32]

The samples were excited under inert atmosphere, using a beam from the second harmonic of a pulsed Nd: YAG regenerative laser amplifier, which was operated at wavelength of $532nm$, $100Hz$ repetition rate, with pulses of about $0.1J$ energy/pulse and $100ps$ time duration.[29] The excitation beam was focused on the samples through a cylindrical lens to form a narrow illuminated stripe of $2mm \times 100mm$. The emission light was collected using a fiber and sent to a 0.5-meter spectrometer, where a charged coupled device camera recorded the emission spectrum; the overall spectral resolution of the collecting system was $0.02nm$.[10] PFT was performed for each spectrum with about $5\mu m$ resolution. For the polymer film and dye infiltrated opal, averaging of the PFT spectra was done by changing the illuminated stripe on the film. However for the dye/TiO_2 suspension PFT was performed for the emission spectrum collected for each excitation pulse. Averaging was then simply done over time from the *same* excitation stripe since *self-averaging* occurs with time due to the low viscosity of the suspension.

We demonstrate the PFT averaging technique using the π-conjugated polymer film. At low excitation inten-

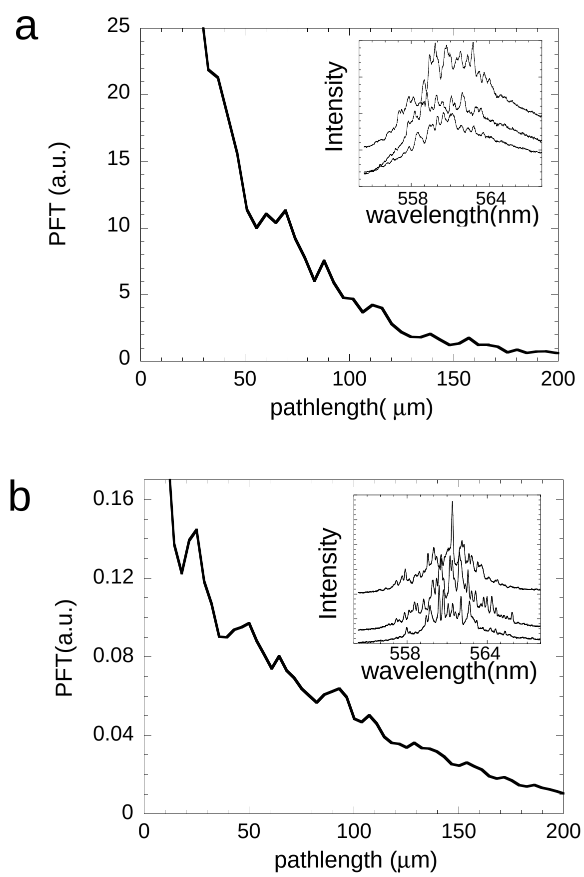

Figure 7. Ensemble average PFT spectroscopy of 125 random lasing spectra of a R6G dye infiltrated into synthetic opal (a), and a methanol suspension of R6G dye with TiO2 balls (b). The insets show three individual random lasing emission spectra from the respective gain medium; the spectra were vertically displaced for ease of comparison.

sity $I < 20$ nJ/pulse, the PL emission band of the DOO-PPV film is approx. $150nm$ wide.[9] This band collapses into a narrow ASE band of about $7nm$ wide at moderately high excitation intensities, $30 < I < 100$ nJ/pulse; the emission intensity in this intensity range depends exponentially[2] on I. However, at $I > 100$ nJ/pulse the emission spectrum develops narrow, laser-like lines as shown in Fig. 6a, which are superimposed on the ASE band,[2,10] and the emission intensity depends linearly[2] on I. Withint this excitation regime the random lasing is due to the amplitude feedback,[1,2] since the narrow lines were shown to be coherent radiation that is typical of conventional lasers.[3,10] We emphasize that the entire spectrum is reproducible at the same intensity and illuminated stripe on the film. However the emission spectrum dramatically changes when different stripes on the film are illuminated.[2] An example of uncorrelated random lasing emission spectra for three different illuminated stripes of a DOO-PPV film is shown in Fig. 6a. Fig. 6b shows the calculated PFT of the three random lasing emission spectra shown in Fig. 6a. Recall, that for a well-defined laser cavity, the PFT shows even-spaced peaks d_m, given by $d_m = mLn/\pi$.[29] Although each PFT spectrum in Fig. 6b contains sharp Fourier components, there is no apparent correlation between the individual PFT spectra.

To find out whether the hidden order is present in the random lasing spectra we performed ensemble averaging of many PFT's measured from different illuminated stripes of the DOO-PPV film. Fig. 6c shows the ensemble averaged PFT spectra of increasing numbers, j, of random lasing emission spectra ranging from $j = 25$ to $j = 125$. As the number j of averaged spectra increases it is seen that the background FT increases; this is expected, since with increasing j, more random FT components contribute to the average PFT. The background

FT spectrum is similar to $1/f$ noise in resistance fluctuations,[28] and it can be fit with a stretched exponential function.

The most striking phenomenon, however, is that the averaged PFT spectrum does not smooth out with increasing j, but instead develops rather sharp features at $d_1 \approx 18\mu m$ with about five harmonics up to $d_6 \approx 110\mu m$. This observation, revealing the hidden order in the random lasing spectra in three different media, is our central result. It is obvious that, in terms of disorder, different illuminated spots are *statistically independent*. Therefore, the almost periodic structure in the averaged PFT indicates that: (i) the random lasing is due to well-defined laser cavities, and (ii) the pathlengths of all cavities responsible for random lasing are almost indetical. Using $n = 1.8$ for DOO-PPV we obtain from $d_1 = nL_0/\pi$ an ensemble-averaged random cavity pathlength, $L_0 \approx 30\mu m$. Also from the CBS albedo cone of the DOO-PPV polymer film we obtained $l \approx 5.2\mu$m. We thus have for the random lasing resonator $l \gg \lambda$ that falls in the category of weak light scattering, the case considered in Sect. 2.

We emphasize, that not only the presence of the features in the average PFT spectra is common for *different* random media studied, but the *shapes* of the average PFT's are *identical* for these media. This observation provide a conclusive experimental evidence that random lasing in the weak scattering regime is due to random resonators.

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