Photon Statistics and Coherence in Light Emission from a Random Laser

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We derive the photon number probability distribution of light emitted from a random multiple-light-scattering medium with gain, using a generalized master equation. Our model treats the random laser medium as a collection of low quality-factor cavities, coupled by random photon diffusion. We demonstrate that, with stronger scattering, the pumping threshold for the transition from chaotic to isotropic coherent light emission decreases and the local second order coherence, above threshold, increases.

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The prediction [1] and observation [2] of laserlike emission from random multiple-light-scattering media with gain have sparked broad interest [3–18] in the study of randomly disordered dielectric microstructures as novel light sources. Experimental studies in colloidal samples [3–6] and optically [7] and electrically [8] pumped semiconductor powders have revealed emission from these dielectric microstructures exhibiting spectral and temporal light intensity characteristics of a multimode laser oscillator. More recent experiments [9,10] have demonstrated that light emitted from random amplifying media, although uncollimated, exhibits coherence properties characteristic of true laser light. While several theoretical studies [11–13] have provided models for the overall input-output intensity characteristics of the random laser, a fundamental and comprehensive understanding of photon statistics and optical coherence [14–18] remains an open question.

In this Letter, we study the coherence properties of the random laser using a simple but novel approach, based on a generalized master equation formalism describing multiple light scattering and atomic excitation. Our description incorporates the probabilistic character of the photon emission and absorption processes needed to characterize optical coherence. Photon statistics can be described using a master equation for photon probability distribution function [19]. In our model, we replace the cavity-loss terms in the conventional laser master equation with terms that describe radiative transport and multiple light scattering. Our analysis attributes feedback to average wave transport leading to laser action throughout the random medium on average. Our generalized master equation model enables direct description of the random laser in terms of experimentally defined parameters, such as scatterer density and gain concentration. By evaluating the local second order degree of optical coherence, we infer that above a specific pumping threshold, the photon statistics changes from Bose-Einstein (characteristic of incoherent light) to Poissonian (characteristic of laser light). Moreover, the local coherence is increased and the lasing threshold decreased as the transport mean free path for the photon in the random medium is made smaller.

Light at a specific frequency ω and vacuum wavelength $\lambda = 2\pi c/\omega$ propagating through the random medium exhibits a photon transport mean free path l^*, the average spatial displacement of the photon for its direction of propagation to become nearly uncorrelated with its initial wave vector. We partition the sample into a collection of hypothetical cubic cells of side length l^* centered on the points of a cubic lattice with lattice constant $a = l^*$. These cells exchange photons with the neighboring cells through diffusion, and the number of photons within each cell fluctuates in time due to atomic emission and absorption events. Each cell is labeled by a "coarse-grained" position vector, $\boldsymbol{r}$. Assuming that $l^* \gg \lambda$, it is possible to simultaneously associate this approximate position $\boldsymbol{r}$ and arbitrary wave vector $\boldsymbol{k}$ with photons in the medium. A more precise formulation of this description from the true wave-field amplitude can be derived from the optical Wigner coherence function [15]. Each cell labeled by $\boldsymbol{r}$ is characterized by the joint distribution function $P_{\boldsymbol{r}}^{n,N}$, describing the probability that n photons are present (propagating in an arbitrary direction) and that N atoms within this cell are in their excited state. We define the photon probability distribution $P_{\boldsymbol{r}}^n \equiv \sum_N P_{\boldsymbol{r}}^{n,N}$ and the average number of excited atoms in the cell centered at $\boldsymbol{r}$, when precisely n photons occupy the cell: $N(n, \boldsymbol{r}) \equiv (\sum_N N P_{\boldsymbol{r}}^{n,N})/P_{\boldsymbol{r}}^n$. $P_{\boldsymbol{r}}^n$ changes with time due to absorption and emission of photons by atoms within the cell as well as transport of photons to and from neighboring cells. Here, we model the "atom" as a four-level system, in which the lower level of the excited state manifold is depopulated by radiative emission to the ground state manifold (lasing transition). We also assume that the emission and absorption spectra have negligible overlap so that the reabsorption of the emitted photons is neglected. As in a conventional laser master equation description, the rate at which photons are added to the cell by radiative emission when n photons are already present in the cell is given by $\gamma_{\mathrm{sp}}(n+1)N(n,\boldsymbol{r})P_{\boldsymbol{r}}^n$, where γ_{sp} is the single-atom spontaneous emission rate. This leads to a

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state with $(n+1)$ photons and a corresponding decay of $P_{\boldsymbol{r}}^{n}$ with time. The factor of $(n+1)$ in the overall emission rate is the usual enhancement factor when $n+1$ indistinguishable bosons appear in the final state [20]. On the other hand, $P_{\boldsymbol{r}}^{n}$ can increase with time if there are initially $(n-1)$ photons in the background and a single photon is emitted by one of $N(n-1,\boldsymbol{r})$ atoms. The rate of increase of $P_{\boldsymbol{r}}^{n}$ in this case is given by $\gamma_{\mathrm{sp}} n N(n-1,\boldsymbol{r}) P_{\boldsymbol{r}}^{n-1}$.

The new dynamics of the photon probability distribution in a random medium arise from the inflow and outflow of photons from a given cell due to random scattering processes. In a simple model of isotropic random scattering, the photon travels ballistically at the speed of light c over the length l^* after which its direction is randomized by scattering into a neighboring cell. The rate for this process is $w = c/l^*$. If there are initially n photons in the cell at $\boldsymbol{r}$, then the outflow of a photon would cause decay of $P_{\boldsymbol{r}}^{n}$. Since each of the n photons is leaving the cavity at the rate w, the overall decay rate of $P_{\boldsymbol{r}}^{n}$ is given by $-nwP_{\boldsymbol{r}}^{n}$. On the other hand, if there are initially $(n+1)$ photons in the cell, the outflow of a single photon will enhance $P_{\boldsymbol{r}}^{n}$. This enhancement could arise from any of the $(n+1)$ photons initially present and the rate of increase of $P_{\boldsymbol{r}}^{n}$ is given by $w(n+1)P_{\boldsymbol{r}}^{n+1}$. A more general description based on the optical Wigner coherence function, capable of describing anisotropic scattering, will be presented elsewhere [21].

In a conventional high-quality factor laser cavity, consisting of a pair of mirrors, w is the analogue of the leakage rate of light from the laser. While a conventional laser has a large number of extraneous nonlasing modes, in the random laser *all* modes can contribute equally on average to the overall lasing process. Light scattered in a random direction simply enters a neighboring cell, which participates with comparable probability to the buildup of laser radiation. In other words, the rate γ_{las} of photons emitted by atoms into a lasing mode is equal to the total rate γ_{sp} of photons spontaneously emitted. The actual lasing efficiency of excited atoms, however, is diminished by nonradiative relaxation described by a rate γ_{nr}, and we define an efficiency factor $\beta \equiv \gamma_{\mathrm{las}}/(\gamma_{\mathrm{sp}}+\gamma_{\mathrm{nr}})$. Moreover, the efficiency is severely offset by the fact that the cavity decay rate w is typically greater than the spontaneous emission rate. In the language of conventional lasers, the cells of the random laser act as "bad cavities."

The inflow of photons to the cell at $\boldsymbol{r}$ from a neighboring cell at $\boldsymbol{r}+\boldsymbol{\delta}$ containing n' photons occurs at a rate $(w/2)n'$. This rate must be weighted by the probability, $P_{\boldsymbol{r}+\boldsymbol{\delta}}^{n'}$, that there are in fact n' photons in the cell at $\boldsymbol{r}+\boldsymbol{\delta}$. This leads to an increase in $P_{\boldsymbol{r}}^{n}$, provided that there are initially $n-1$ photons in the cell at $\boldsymbol{r}$, and to a decrease in $P_{\boldsymbol{r}}^{n}$ if there are initially n photons in the cell at $\boldsymbol{r}$. Overall, the rate of increase of $P_{\boldsymbol{r}}^{n}$ due to a neighboring cell at $\boldsymbol{r}+\boldsymbol{\delta}$ is given by $\sum_{n'} n'(w/2)(P_{\boldsymbol{r}}^{n-1}-P_{\boldsymbol{r}}^{n})P_{\boldsymbol{r}+\boldsymbol{\delta}}^{n'}$. This must then be summed over all possible choices $\boldsymbol{\delta}$ of neighboring cells. For simplicity, we will consider only the case of plane wave pumping a slablike sample. In this case, the problem is one dimensional, and we consider the flow of photons normal to the slab faces. Putting all the above processes together, we arrive at the master equation:

$$\begin{aligned}\dot{P}_{\boldsymbol{r}}^{n} = &-\gamma_{\mathrm{sp}}(n+1)N(n,\boldsymbol{r})P_{\boldsymbol{r}}^{n} + \gamma_{\mathrm{sp}} n N(n-1,\boldsymbol{r})P_{\boldsymbol{r}}^{n-1} \\ &+ w[(n+1)P_{\boldsymbol{r}}^{n+1} - nP_{\boldsymbol{r}}^{n}] \\ &+ \frac{w}{2}\sum_{n'} n'(P_{\boldsymbol{r}}^{n-1}-P_{\boldsymbol{r}}^{n})[P_{\boldsymbol{r}-\boldsymbol{\delta}}^{n'} + P_{\boldsymbol{r}+\boldsymbol{\delta}}^{n'}]. \end{aligned} \tag{1}$$

This master equation is similar to that for the conventional laser [22], where the cavity-loss terms are replaced by terms that correspond to photon diffusion and supplemented by random diffusion of photons into the cell at $\boldsymbol{r}$ from neighboring cells. In the case of more general anisotropic scattering from wave vector $\hat{\mathbf{k}}$ to $\hat{\mathbf{k}}'$ with probability $w_{\hat{\mathbf{k}},\hat{\mathbf{k}}'}$, a similar master equation can be derived for the probability $P_{\boldsymbol{r},\hat{\mathbf{k}}}^{n}(t)$ that n photons traveling in direction $\hat{\mathbf{k}}$ exist in the cell at time t [21]. We also point out that the master equation (1) accounts for only the *average* properties of the random medium. More generally, the hopping rate w is itself a random variable that varies from location to location, depending on the statistical distribution of scatterers. Our mean-field theory containing a single average diffusion coefficient $D \equiv (w/2)(l^*)^2$ neglects contributions to random lasing arising from highly improbable local configurations of the scattering potential, which may contribute isolated "spots" of laser activity [23] below the threshold for lasing the entire medium on average.

Using the expression $\bar{n}(\boldsymbol{r}) = \sum_{n'} n' P_{\boldsymbol{r}}^{n'}$ for the average photon number in the cell centered at $\boldsymbol{r}$, the last group of terms in Eq. (1), in the continuum limit ($\boldsymbol{\delta}\to 0$), becomes $[D\nabla_{\boldsymbol{r}}^2\bar{n}(\boldsymbol{r}) + w\bar{n}(\boldsymbol{r})][P_{\boldsymbol{r}}^{n-1}-P_{\boldsymbol{r}}^{n}]$.

Multiplying Eq. (1) by n and then summing over n yields the diffusion equation

$$\dot{\bar{n}}(\boldsymbol{r}) = D\nabla_{\boldsymbol{r}}^2\bar{n}(\boldsymbol{r}) + \gamma_{\mathrm{sp}}[\bar{n}(\boldsymbol{r})+1]\bar{N}(\boldsymbol{r}), \tag{2}$$

where we made the additional mean-field factorization $\sum_{n,N} nNP_{\boldsymbol{r}}^{n,N} \approx \bar{n}(\boldsymbol{r})\bar{N}(\boldsymbol{r})$, and $\bar{N}(\boldsymbol{r}) \equiv \sum_n N(n,\boldsymbol{r})P_{\boldsymbol{r}}^{n}$ is the average number of excited atoms in the cell at $\boldsymbol{r}$.

For steady-state pumping, we obtain the steady-state solution for the photon distribution function corresponding to balancing of transitions between neighboring photon number states:

$$[\gamma_{\mathrm{sp}}(n+1)N(n,z) + D\nabla_{\boldsymbol{r}}^2\bar{n}(\boldsymbol{r}) + w\bar{n}(\boldsymbol{r})]P_{\boldsymbol{r}}^{n} = w(n+1)P_{\boldsymbol{r}}^{n+1}. \tag{3}$$

This detailed balance equation for the random laser has the solution:

$$P_{\boldsymbol{r}}^{n} = P_{\boldsymbol{r}}^{0}\prod_{k=0}^{n-1}\left[\frac{\gamma_{\mathrm{sp}}N(k,\boldsymbol{r})}{w} + \frac{D\nabla_{\boldsymbol{r}}^2\bar{n}(\boldsymbol{r}) + w\bar{n}(\boldsymbol{r})}{w(k+1)}\right]. \tag{4}$$

The statistical properties and coherence of the emitted radiation can be studied using the Fano-Mandel parameter [24] $F \equiv [\overline{n^2} - \bar{n}^2 - \bar{n}]/\bar{n}$, describing photon number fluctuations. Here $\overline{n^2}(\mathbf{r}) = \sum_n n^2 P^{n,\mathbf{r}}$ and $g^{(2)}(0) \equiv (\overline{n^2} - \bar{n})/(\bar{n})^2 = F/\bar{n} + 1$ is the well known degree of second order coherence [25]. For chaotic (incoherent) light $g^{(2)}(0) = 2$ and for Poissonian (coherent) light $g^{(2)}(0) = 1$. For a laser operating well below threshold in a weakly excited thermal state, as well as for a laser operating well above threshold in a coherent state, $F \rightarrow 0$. In a conventional laser with $\beta \approx 10^{-5}$, the Fano parameter exhibits a sharp peak as a function of pump intensity at the lasing threshold [19]. The large fluctuations in the threshold region are indicative of a phase transition. As the spontaneous emission factor β increases, this peak becomes smaller and wider.

We obtain photon statistics and optical coherence defined by F at various positions $\mathbf{r}$ within the random laser sample by evaluating $P^n_{\mathbf{r}}$. This requires knowledge of the mean number of photons $\bar{n}(\mathbf{r})$ determined by the diffusion equation (2) and its boundary conditions. However, $\bar{n}(\mathbf{r})$ and $\bar{N}(\mathbf{r})$ are also determined by $P^n_{\mathbf{r}}$, and the coupled nonlinear Eqs. (2) and (4) must be solved self-consistently. The steady-state atomic occupation number can be obtained from the rate equation for the atom number in the presence of a stationary photon distribution $P^n_{\mathbf{r}}$, consistent with a four-level laser scheme described above, as [22] $N(n, \mathbf{r}) = \mathcal{P}(\mathbf{r})/[\gamma_{\rm nr} + \gamma_{\rm sp}(n+1)]$. Here $\mathcal{P}(\mathbf{r})$ is the continuous wave pumping rate, which decreases within the sample from the incident value due to both absorption by the active medium and scattering. Substituting this expression into Eq. (4) corresponds to an "adiabatic elimination" of the atomic variables. Using the fact that $\bar{N}(\mathbf{r}) = \sum_{n'} P^{n'}_{\mathbf{r}} N(n', \mathbf{r})$ in Eq. (2) then leads to a closed set of equations for $P^n_{\mathbf{r}}$ and $\bar{n}(\mathbf{r})$. We note that the typical "cavity decay rate" w for a random laser is about 100 times faster than the atomic transition rates. As such, the adiabatic approximation is not rigorously justified. However, we have verified numerically that a more general treatment involving nonadiabatic atomic dynamics yields qualitatively similar results to those presented here [21].

For concreteness, we consider a slab in the xy plane, between the two planes $z = 0$ and $z = L$. We define $z < 0$ as the left region and $z > 0$ as the right region. A pumping beam is collimated perpendicular to the $z = 0$ plane from the left. The light emitted from the sample is measured by a detector on the left.

In Fig. 1, we plot the Fano-Mandel parameter as a function of incident pumping rate, at different depths within the random laser sample, for fixed scatterer density and gain concentration. As with a conventional laser, the Fano-Mandel parameter exhibits a fluctuation peak indicative of a transition from chaotic to coherent light. The magnitude of the fluctuation peak decreases deeper within the sample, due to attenuation of the local pumping intensity as it penetrates deeper into the sample. Deeper into the sample, the fluctuations also decrease

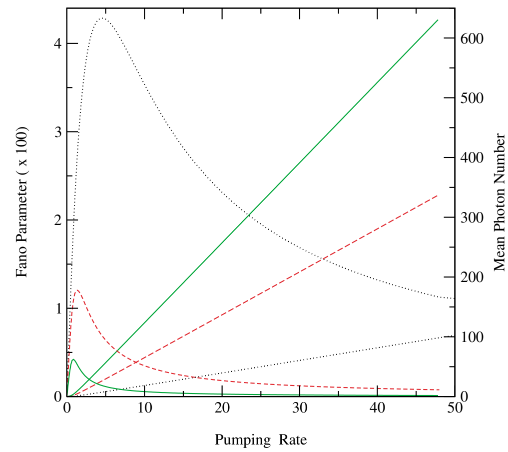

FIG. 1 (color online). Fano parameter and average photon number at different locations within the random laser sample, for a transport mean free path of 10^{-4} cm and absorption length of 1.5×10^{-2} cm. The position within the sample is $0.25l^*$ (dotted line), l^* (dashed line), and $2.5l^*$ (continuous line), respectively, from the front of the sample. We set $\beta = 0.1$ and $L = 1$ cm in the calculations, and the pumping rate is in units of $\Gamma \equiv \gamma_{\rm sp} + \gamma_{\rm nr}$.

more rapidly with the increasing of the pump above threshold. This suggests that the light emitted from deeper inside the sample, although weaker in intensity, is more coherent than that emitted from near the front face of the sample. These modes have a smaller contribution to the total laser radiation detected outside of the sample. The laser light measured by a detector outside the sample is given by a weighted sum of the light emitted from various points within the sample. A microscopic derivation of the output electric field fluctuations is beyond the scope of our optical diffusion model. In our model, light intensity from different spatial regions is assumed uncorrelated and it is convenient to define an averaged output Fano parameter [18,26]:

$$F_{\rm output} = \frac{1}{l^*} \int_0^L dz h(z) F(z), \tag{5}$$

where $F(z)$ is the Fano parameter for light at a depth z within the sample, $h(z) = \bar{n}(z)\rho^2(z)/[(1/l^*) \times \int_0^L dz \bar{n}(z)\rho(z)]$, $\bar{n}(z)$ is the average number of photons emitted from a depth z within the sample, $\rho(z) = 1/(4\pi) \int d\hat{\mathbf{k}}_f \exp[-z/(|\hat{\mathbf{k}}_f \cdot \hat{\mathbf{z}}|l^*)]$, and $\mathbf{k}_f = \hat{\mathbf{k}}_f(\omega/c)$ is the wave vector of the emerging light. Here $\exp[-z/(|\hat{\mathbf{k}}_f \cdot \hat{\mathbf{z}}|l^*)]$ represents the fraction of the radiation at depth z that emerges from the sample without being further scattered [27].

The averaged output Fano parameter for various values of the scatterer density and gain concentration is presented in Fig. 2. Once again we observe a transition from chaotic to coherent light at a specific pump threshold. This pump threshold is identical to the threshold for laser oscillation (defined as the value of pump that marks

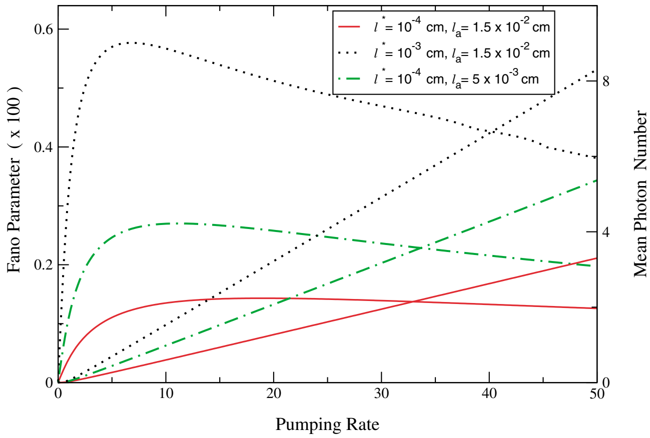

FIG. 2 (color online). Averaged Fano parameter and average photon number for the output radiation, for various scatterer and gain parameters. The other sample parameters are the same as for Fig. 1.

a transition of the slope in the input-output characteristics). It has been shown [11,28] that the lasing threshold decreases with the addition of scatterers in the system, since, as the transport mean free path becomes shorter, there is a higher probability that photons emitted in a given region would return to that same region through the "random walk" process (rather than leave the sample), thereby contributing coherent feedback. It is also apparent from Fig. 2 that optical coherence, above threshold, is enhanced in samples with shorter mean free path (stronger scattering) and higher gain molecule concentration within the range of parameters studied.

In conclusion, we have developed a theoretical model to describe photon number statistics of a random laser. Our results suggest that stronger scattering not only lowers the threshold for laser action, but also diminishes the noise with respect to the Poissonian value. These results may be used to explain recent experiments, where, for a lower scatterer density [9], the radiation was found to be weakly coherent, while for a system with stronger scatterers [10], the radiation became highly coherent above threshold. These results also suggest that dramatic enhancement in random laser light emission may occur in stronger scattering samples approaching the photon localization threshold [29].

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