Photon Statistics of Random Lasers with Resonant Feedback

H. Cao, Y. Ling, J. Y. Xu, and C. Q. Cao
Department of Physics and Astronomy, Materials Research Center, Northwestern University, Evanston, Illinois 60208

Prem Kumar
Department of Electrical and Computer Engineering, Northwestern University, Evanston, Illinois 60208
(Received 14 August 2000)

We have measured the photon statistics of random lasers with resonant feedback. With an increase of the pump intensity, the photon number distribution in a single mode changes continuously from Bose-Einstein distribution at the threshold to Poisson distribution well above the threshold. The second-order correlation coefficient drops gradually from 2 to 1. By comparing the photon statistics of a random laser with resonant feedback and that of a random laser with nonresonant feedback, we illustrate very different lasing mechanisms for the two types of random lasers.

DOI: 10.1103/PhysRevLett.86.4524 PACS numbers: 42.50.Ar, 42.25.Fx, 42.55.–f, 71.55.Jv

Over the past two decades, there has been much progress in the study of light transport in a disordered dielectric medium. Most studies are within the framework of ray optics or wave optics. Light diffusion in the weak scattering regime can be modeled by ray optics. The enhanced backscattering and light localization can be explained by the interference effect in the wave optics [1–3]. However, quantum statistical properties of the light in a random medium have received little attention. Recent theoretical studies of photon statistics in random media aim to bridge the gap between random media and quantum optics [4]. In this Letter, we present an experimental study of photon statistics of random lasers and show that photon statistical behavior provides an insight into the lasing mechanism in a disordered medium.

There are two kinds of random lasers: one is with nonresonant (incoherent) feedback; the other is with resonant (coherent) feedback. From the ray optics point of view, lasing with nonresonant feedback is related to the instability of light amplification along open trajectories in a random medium, while lasing with resonant feedback corresponds to the instability of light amplification along closed loop paths. Through recurrent scattering, light may return to its original position through many different paths. All the backscattered waves interfere with each other, and their phase relationship determines the lasing frequencies. We investigate quantum statistical properties of random lasers with resonant feedback.

ZnO nanoparticles with an average diameter of 80 nm are cold pressed under a pressure of 200 MPa to form a pellet. The pellet is a disk of thickness 2 mm and diameter 1 cm. The transport mean free path l in the ZnO pellet is characterized in the coherent backscattering experiment [1]. From the angular width of the backscattering cone, we estimate $l \sim 2.3\lambda$ [5].

The ZnO pellet is optically excited by a train of 20 ps pulses separated by 100 ms from a frequency-tripled ($\lambda = 355$ nm) mode-locked Nd:YAG laser. The pump beam is focused by a lens to a spot of $\sim$15 μm in diameter on the sample surface. Another lens collects the sample emission in a single transverse mode and focuses it to the entrance slit of a 0.5 m Jarrell-Ash spectrometer. The output port of the spectrometer is connected to a Hamamatsu streak camera whose entrance slit is perpendicular to that of the spectrometer. The streak camera has a temporal resolution of 2 ps. Its photocathode width gives an observable spectral window of 6.7 nm with a spectral resolution of 0.1 nm. Partial output of the pump laser goes directly to a fast photodiode whose output signal triggers the streak camera. A Peltier-cooled charge-coupled-device (CCD) camera, operating at -50 °C for reduced dark noise, is used to record the streak image. The streak camera operates in the photon counting mode. A threshold is set to eliminate the contribution of the dark-current noise. Thus, in the absence of an input signal, no photons are counted.

By combining the spectrometer with the streak camera, we are able to separate different lasing modes and measure the temporal evolution of each mode. Figure 1 is

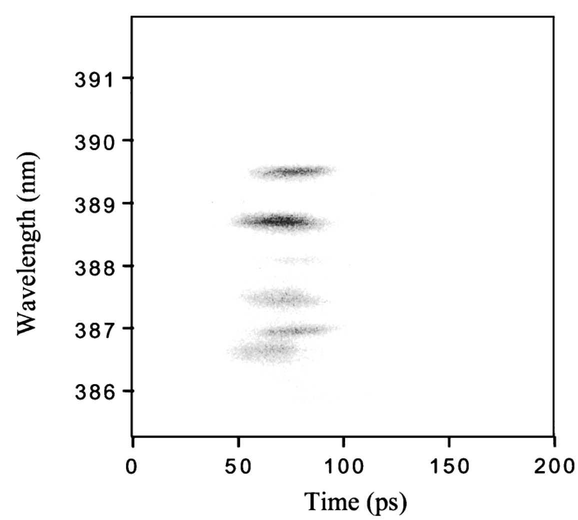

FIG. 1. The measured streak image of the emission from the ZnO pellet. The incident pump pulse energy is 4.5 nJ.

a two-dimensional (2D) streak image taken by the CCD camera. The horizontal axis is the time, and the vertical axis is the wavelength. When the pump power exceeds a threshold, discrete peaks appear in the emission spectrum. The linewidth of these peaks is less than 0.2 nm. Simultaneously, the emission pulses are dramatically shortened from 200 ps below the threshold to less than 50 ps above the threshold. For different modes, lasing starts at different times and lasts for different periods of time. To measure the photon statistics of a single lasing mode, we draw on the streak image a rectangle whose one side is wavelength interval $\Delta\lambda$ and the other side is time interval Δt. The number of photons inside this rectangle is counted for each pulse. With one counting interval per emission pulse, the sampling rate is equal to the pulse repetition rate (10 Hz). The sampling time Δt is short compared to the emission pulse width. After collecting photon count data for a large number of pulses, the probability $P(n)$ of n photons within the wavelength interval $\Delta\lambda$ and the time interval Δt is obtained. Because the sampled radiation field is within a frequency interval $\Delta\nu = c\Delta\lambda/\lambda^2$, its relaxation must occur on the time scale longer than $1/\Delta\nu$. We set the sampling time $\Delta t < 1/\Delta\nu$ so that it is shorter than the coherence time of the radiation field. From another point of view, when $\Delta\nu \cdot \Delta t < 1$, the counting area corresponds to a single electromagnetic (EM) mode. For a single-mode coherent light, the photon number distribution $P(n)$ satisfies Poisson distribution $P(n) = \langle n\rangle^n e^{-\langle n\rangle}/n!$, where $\langle n\rangle$ is the average photon number. For a single-mode chaotic light, the photon number distribution $P(n)$ satisfies Bose-Einstein (BE) distribution $P(n) = \langle n\rangle^n/[1 + \langle n\rangle]^{n+1}$. Note that the above distribution holds only for a single mode. For a multimode chaotic light, the photon number distribution approaches Poisson distribution. From $P(n)$, we obtain the normalized second-order correlation coefficient $G_2 = 1 + (\langle(\Delta n)^2\rangle - \langle n\rangle)/\langle n\rangle^2$. For Poisson distribution, $G_2 = 1$. For BE distribution, $G_2 = 2$.

To check the reliability of our spectrometer-streak camera setup for photocounting, we have measured photon statistics of a coherent light. The ZnO sample is replaced by a pellet made of TiO_2 nanoparticles. Since the band gap energy of TiO_2 is larger than the pump photon energy, the pump laser light is not absorbed; instead it is scattered. The spectrometer is tuned to the pump wavelength 355 nm. Photon counting is done at the time of maximum intensity of each laser pulse, and the sampling time $\Delta t =$ 3.9 ps. The wavelength interval $\Delta\lambda = 0.1$ nm, and the corresponding $\Delta\nu = 2.4 \times 10^{11}$ Hz. Hence, $\Delta\nu \cdot \Delta t =$ 0.93. Figure 2(a) plots the measured photon count distribution of the scattered laser light. From the data of $P(n)$, we calculate the count mean $\langle n\rangle = \sum_n nP(n)$ and obtain the Poisson distribution for the experimental value of $\langle n\rangle$. As shown in Fig. 2(a), the measured photon count distribution is very close to the Poisson distribution with the same count mean. Using the data of $P(n)$, we calculate $G_2 = 0.97$.

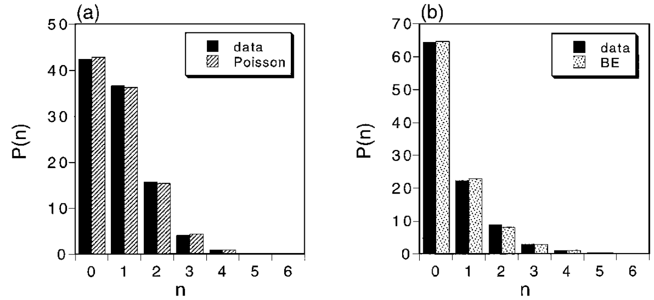

FIG. 2. (a) The solid columns are the measured photon count distribution of the laser light scattered by the stationary TiO_2 pellet. The dashed columns are the Poisson distribution for the same count mean. (b) The solid columns are the measured photon count distribution of the laser light scattered by the rotating TiO_2 pellet. The dotted columns are the Bose-Einstein distribution for the same count mean.

Next, the TiO_2 pellet is driven by a motor and starts rotating at 200 rpm. The part of the sample hit by the focused laser beam moves with a speed of ~100 mm/s. Since the distance it moves during the pulse duration (20 ps) is much less than the laser wavelength and the TiO_2 grain size (0.4 μm), the sample is practically at rest when illuminated by a single laser pulse. However, during the 100 ms interval between the pulses, the part of the sample which is hit by the laser pulse moves out of the focal region of the lens, and the next pulse hits a different part of the sample. Hence, the speckle pattern changes from pulse to pulse. In our setup, the detection area is less than the average speckle size. The counting parameters, e.g., Δt and $\Delta\lambda$, are the same as before. Every pulse of the scattered laser light is sampled only once, and the sampling time is shorter than the pulse width. After collecting many pulses, we obtain a random superposition of a great number of coherent beams. The photon number distribution is known to be BE distribution [6]. Figure 2(b) plots the measured photon count distribution and the BE distribution for the same count mean. The measured distribution follows the BE distribution closely and has a G_2 of 1.88.

By measuring the photon statistics of a coherent light and a "synthesized" chaotic light, we have confirmed the reliability of our apparatus for the photon statistics measurement. We then switch to the ZnO pellet and measure the photon statistics of random lasers with resonant feedback. The pump intensity is above the threshold where discrete spectral peaks appear, so that we can measure the photon statistics of a single peak. We pick up one of the brightest emission peaks and set $\Delta\lambda = 0.12$ nm around the center wavelength λ_0 of the peak. Photon counting is done every time the emission intensity at λ_0 reaches its maximum following a pump pulse. The sampling time $\Delta t = 3.9$ ps, and $\Delta\nu \cdot \Delta t = 0.95$. Since both radiative and nonradiative recombination times of ZnO are longer than a single pump pulse but much shorter than the interval between the pump pulses, the system is always in the transient regime. However, the emission intensity is nearly constant within a counting interval, because the sampling time is much shorter than the emission pulse width.

Figure 3(a) shows the measured photon statistics of ZnO emission at the threshold where discrete spectral peaks appear. The measured photon count distribution is almost identical to the BE distribution of the same count mean. The value of G_2 is 1.94. As we increase the pump intensity, the photon statistics of ZnO emission starts deviating from the BE statistics. As shown in Fig. 3(b), when the pump intensity is 1.5 times of the threshold, the measured photon count distribution is between the BE distribution and the Poisson distribution. G_2 becomes 1.51. When the pump intensity is increased to 3 times of the threshold, the photon count distribution of ZnO emission gets closer to the Poisson distribution [Fig. 3(c)]. G_2 is reduced to 1.19. Eventually, when the pump intensity is 5.6 times of the threshold, the photon count distribution is nearly identical to the Poisson distribution [Fig. 3(d)]. The corresponding G_2 is 1.06.

Figure 4 shows the value of second-order correlation coefficient G_2 as a function of pump intensity. As the pump intensity increases, G_2 decreases gradually from 2 to 1. Because we take only a finite number of pulses in the measurement, the rms error in G_2 is equal to $(2/K\langle n\rangle^2)^{1/2}$, where K is the number of pulses. The sampling error for G_2 is calculated and plotted for each data point in Fig. 4. Figures 3 and 4 illustrate that the photon statistics of the emitted light from the ZnO pellet changes continuously from BE statistics at the threshold to Poisson statistics well above the threshold.

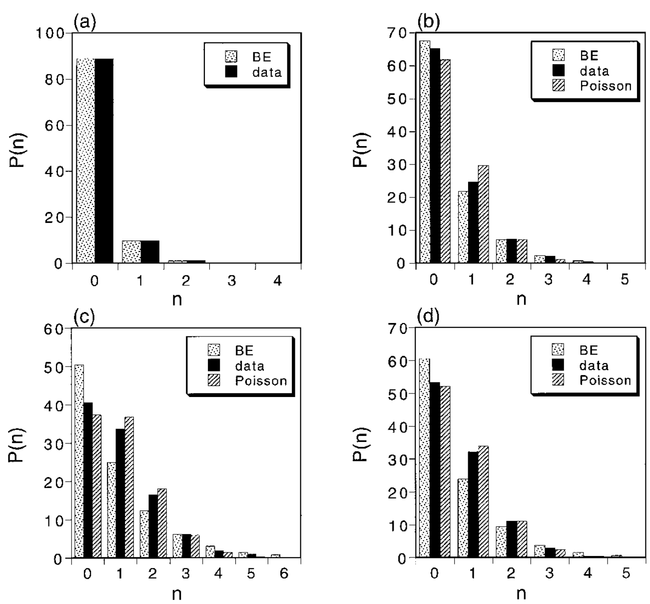

FIG. 3. The solid columns are the measured photon count distribution of the emission from the ZnO pellet. The dotted (dashed) columns are the Bose-Einstein (Poisson) distribution for the same count mean. The incident pump intensity is (a) 1.0, (b) 1.5, (c) 3.0, and (d) 5.6 times of the threshold intensity where discrete spectral peaks appear.

In principle, the measured photon count distribution is not necessarily equal to the actual photon number distribution inside the random medium. Let β be the probability of a photon escaping through the boundary of the random medium and being counted by our detector. The value of β is estimated to be less than 10^{-4} in our experiment. Fortunately for our experiment, the BE distribution and the Poisson distribution are not affected by β [7]. Namely, when the original photon number distribution inside the random medium is BE distribution (or Poisson distribution), the measured photon count distribution remains BE distribution (or Poisson distribution) regardless of the value of β. Therefore, our data illustrate that the photon statistics of the light field inside the random medium changes from BE statistics at the threshold to Poisson statistics well above the threshold.

Our numerical simulation of random lasers with resonant feedback also confirms the generation of coherent light above the threshold. We calculate the classical EM field in an active random medium by solving the Maxwell equations [8]. A seed pulse is launched inside the random medium of finite size. Below the lasing threshold, the seed pulse dies away from the random medium into the absorbing boundary layers. Only when the optical gain exceeds a threshold, the EM field builds up inside the random medium. Since the classical EM field represents the coherent part of the quantum field, our simulation result indicates that the quantum field in the random medium has no coherent part below the threshold; its coherent component appears only above the threshold.

The quantum statistical properties of light reveal very different lasing mechanisms for the two types of random

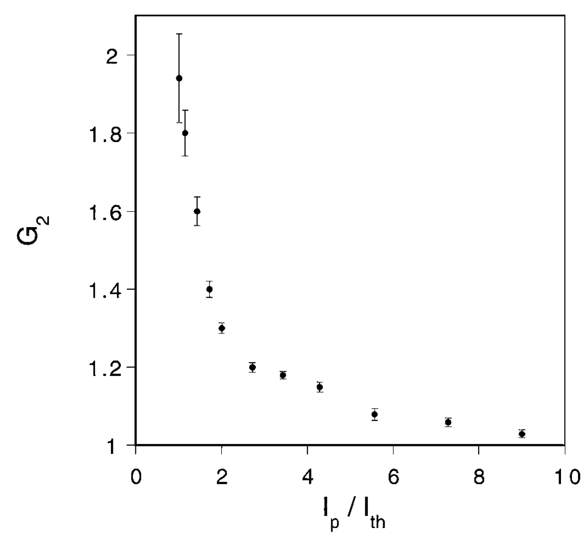

FIG. 4. The second-order correlation coefficient G_2 as a function of the ratio of the incident pump intensity I_p to the threshold intensity I_{th}.

lasers. The photon statistics of a random laser with resonant feedback is very different from that of a random laser with nonresonant feedback. For a random laser with nonresonant feedback, the fluctuation of the total number of photons in all modes of laser emission is smaller than that of blackbody radiation with the same number of modes [9,10]. However, the photon number distribution in a single mode remains BE distribution even well above the threshold. Based on these experimental results, we propose a qualitative explanation for the random lasers with the concept of quasistates. The quasistates are the eigenmodes of the Maxwell equations in a finite-sized random medium. The boundary condition for quasistates is the absence of any incoming waves [11]. The eigenenergies are complex numbers, whose imaginary parts represent the decay rates. The decay of a quasistate results from light leakage through the boundaries of the random medium and loss of its photon to other quasistates. When $kl > 1$ (k is a wave vector; l is the transport mean free path), the average decay rate of a quasistate is larger than the average frequency spacing of adjacent quasistates. Hence, the quasistates are spectrally overlapped, giving a continuous emission spectrum.

In the case of weak scattering, the quasistates decay fast, and they are strongly coupled. Because of photon exchange among the quasistates, the loss of a set of interacting quasistates is much lower than the loss of a single quasistate. In an active random medium, when the optical gain for a set of interacting quasistates at the frequency of gain maximum reaches the loss of these coupled quasistates, the total photon number in these coupled states builds up. This process is lasing with nonresonant feedback [12]. The drastic increase of photon number at the frequency of gain maximum results in a significant spectral narrowing [13]. Well above the threshold, gain saturation quenches the total photon number fluctuation. However, strong coupling of the quasistates prevents stabilization of the photon number in a single state.

With an increase in the amount of optical scattering, the dwell time of light in the random medium increases, and the mixing of the quasistates is reduced. Thus the decay rates of the quasistates decrease. When the optical gain increases, it first reaches the threshold for lasing in a set of coupled quasistates at the frequency of gain maximum. As the optical gain increases further, it exceeds the loss of a quasistate that has a long lifetime. Then, lasing occurs in a single quasistate. The spectral linewidth of the quasistate is reduced dramatically above the lasing threshold. A further increase of optical gain leads to lasing in more low-loss quasistates. Laser emission from these quasistates gives discrete peaks in the emission spectrum. This process is lasing with resonant feedback [14,15]. When the scattering strength increases further, the decay rates of the quasistates and the coupling among them continue decreasing. Because of large dispersion of the decay rates of quasistates, the threshold gain for lasing in individual low-loss quasistates becomes lower than the threshold gain for lasing in the coupled quasistates at the frequency of gain maximum. Thus, lasing with resonant feedback occurs first. Because of weak coupling among the quasistates, the photon number fluctuation in each lasing state is quenched by the gain saturation effect well above the threshold.

We thank A. L. Burin for stimulating discussion. This work is supported partially by National Science Foundation under Grant No. ECS-9877113. H. C. acknowledges support from the David and Lucile Packard Foundation and the Alfred P. Sloan Foundation.

[1] M. P. van Albada and A. Lagendijk, Phys. Rev. Lett. **55**, 2692 (1985); P. E. Wolf and G. Maret, *ibid.* **55**, 2696 (1985).
[2] D. S. Wiersma, P. Bartolini, A. Lagendijk, and R. Righini, Nature (London) **390**, 671 (1997); F. J. P. Schuurmans, D. Vanmaekelbergh, J. van de Lagemaat, and A. Lagendijk, Science **284**, 141 (1999).
[3] A. A. Chabanov, M. Stoytchev, and A. Z. Genack, Nature (London) **404**, 850 (2000).
[4] C. W. J. Beenakker, Phys. Rev. Lett. **81**, 1829 (1998); M. Patra and C. W. J. Beenakker, Phys. Rev. A **60**, 4059 (1999).
[5] J. X. Zhu, D. J. Pine, and D. A. Weitz, Phys. Rev. A **44**, 3948 (1991).
[6] F. T. Arecchi, Phys. Rev. Lett. **15**, 912 (1965).
[7] M. Sargent, M. O. Scully, and W. E. Lamb, *Laser Physics* (Addison-Wesley, New York, 1974), p. 302.
[8] H. Cao *et al.,* Phys. Rev. Lett. **84**, 5584 (2000).
[9] R. V. Ambartsumyan, P. G. Kryukov, V. S. Letokhov, and Yu. A. Matveets, Sov. Phys. JETP **26**, 1109 (1968).
[10] G. Zacharakis, N. A. Papadogiannis, G. Filippidis, and T. G. Papazoglou, Opt. Lett. **25**, 923 (2000).
[11] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics; Non-relativistic Theory* (Pergamon, Oxford, 1977).
[12] V. S. Letokhov, Sov. Phys. JETP **26**, 835 (1968).
[13] N. M. Lawandy, R. M. Balachandran, A. S. L. Gomes, and E. Sauvain, Nature (London) **368**, 436 (1994).
[14] H. Cao *et al.,* Appl. Phys. Lett. **73**, 3656 (1998); H. Cao *et al.,* Phys. Rev. Lett. **82**, 2278 (1999).
[15] S. V. Frolov, Z. V. Vardeny, K. Yoshino, A. Zakhidov, and R. H. Baughman, Phys. Rev. B **59**, R5284 (1999).