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Lasing with resonant feedback in random media ☆

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Abstract

We studied experimentally lasing with resonant feedback in disordered media. The lasing threshold and the number of lasing modes exhibit strong dependence on the mean free path. Using a spectrally resolved speckle technique, we obtained the spatial field correlation function for individual lasing states. The electric field amplitude of a lasing state falls exponentially with the transverse coordinate at the sample surface. The spatial extent of a lasing state shrinks as the scattering length decreases.

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Since the pioneer work of Letokhov [1], there have been many studies on lasing in disordered media [2–12]. The so-called random laser is a laser based on disorder-induced scattering. There are two kinds of random lasers: one is with non-resonant (incoherent) feedback, the other is with resonant (coherent) feedback.

In an active random medium, light is scattered and undergoes a random walk before leaving the medium. Because of gain, a photon may induce the stimulated emission of a second photon as it travels in the medium. There are two characteristic length scales. One is the gain length: the distance a photon travels before generating a second photon. The other is the average path length that a photon travels in the gain medium. With an increase in scattering strength, the average path length of photons in the medium increases. When it is equal to the gain length, on average every photon generates another photon before leaving the medium. The photon number increases with time. This process is lasing with nonresonant (incoherent) feedback. It is similar to the neutron scattering in combination of nuclear fission.

When scattering gets stronger, after multiple scattering light may return to the scattering center from which it was scattered before, forming a closed-loop path for light. When the amplification along the loop reaches the loss, lasing oscillation occurs in the loop which serves as a cavity. The lasing frequency is determined by the requirement of constructive interference, i.e., the phase delay along the loop is equal to $2\pi m$, where m is an integer. Such laser is a random laser with resonant (coherent) feedback. Of course, the picture of a single closed-loop path for light is intuitive but

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naive. Light may return to its original position through many different paths. All the backscattered light interferes and their phase relationship determines the lasing frequencies. Therefore, such random laser is a randomly distributed feedback laser. The feedback is provided by the disorder-induced scattering.

From the ray optics point of view, lasing with nonresonant feedback is related to the instability of light amplification along open trajectories, while lasing with resonant feedback corresponds to the instability of light amplification along closed-loop paths. Therefore, the contribution of optical scattering to these two types of random lasers is very different. In random laser with incoherent feedback, scattering merely increases the path length of light in the gain medium so that light is amplified more. In the random laser with coherent feedback, scattering brings light back to its original position, and light interference leads to frequency selection.

To understand the effect of scattering on lasing with resonant feedback, we investigated the dependence of the lasing threshold and the number of lasing modes on the transport mean free path [13]. The random medium used in our experiment is Poly(methyl methacrylate) (PMMA) containing rhodamine 640 perchlorate dye and titanium dioxide (TiO_2) microparticles. The dye serves as gain medium and the microparticles are scattering centers. The average size of TiO_2 particle is 400 nm. By varying the particle density, we changed the mean free path. The transport mean free path l was measured in the coherent backscattering experiment. The output from a He:Ne laser was used as the probe light, because its wavelength is very close to the emission wavelength of rhodamine 640 perchlorate dye. To avoid absorption of the probe light, we used the polymer containing only TiO_2 particles but not dye. From the angular width w of the backscattering cone, we calculate $kl \simeq 0.7n(1-R)/w$, where k is the wave vector in the sample, n is the effective refractive index of the medium, and R is the diffuse reflection coefficient at the sample–air interface.

In the photoluminescence experiment, the samples were pumped by the second harmonics of a pulsed Nd:YAG laser (532 nm, 25 ps, 10 Hz). The pump beam spot on the sample surface is about 50 μm in diameter. Emission from the polymer sheet is collected by a fiber bundle and directed to a 0.5-m spectrometer with a cooled CCD array detector. Above the lasing threshold, discrete narrow peaks emerge in the emission spectrum. The emission intensity increases much more rapidly with the pump intensity. Fig. 1(a) plots the incident pump pulse energy at the lasing threshold versus the transport mean free path. The dye concentration is fixed at 50 mM. As the transport mean free path decreases, the lasing threshold is reduced. The strong dependence of the lasing threshold on the mean free path clearly illustrates the important contribution of scattering to lasing. With an increase in the amount of optical scattering, the feedback provided by

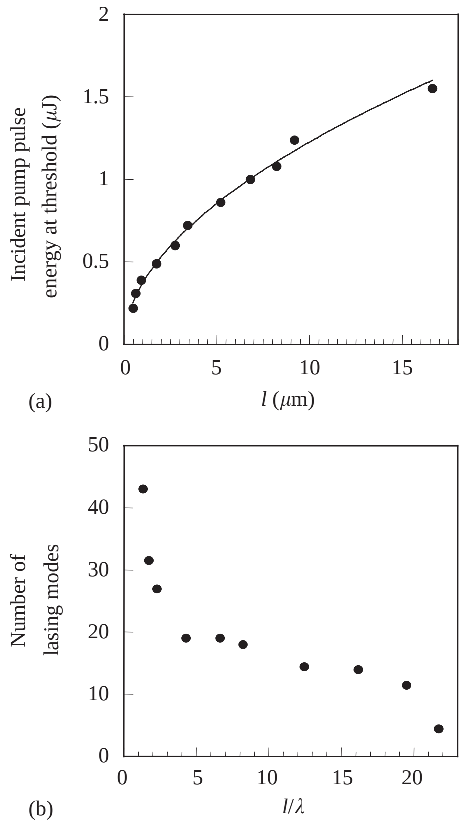

Fig. 1. (a) The incident pump pulse energy at the lasing threshold versus the transport mean free path. (b) The number of lasing modes versus the transport mean free path.

scattering becomes stronger. In other words, the laser cavities formed by recurrent light scattering have lower loss. Thus the lasing threshold is reduced. Through curvefitting, we find the lasing threshold is proportional to the square root of the transport mean free path. Fig. 1(b) shows the number of lasing modes in the samples with different transport mean free path at the same pump intensity. The stronger the scattering is, the more lasing modes emerge. This is because in a sample of stronger scattering strength, there are more low-loss cavities formed by recurrent scattering. When the optical gain is fixed, in more cavities the gain exceeds the loss, and lasing oscillation occurs. An interesting feature in Fig. 1 is that when the transport mean free path approaches the optical wavelength, the lasing threshold pump intensity drops quickly, and the number of lasing modes increases dramatically.

To understand the nature of the lasing states, it is essential to know their spatial profile. We used the spectrally resolved speckle technique to obtain the spatial field correlation function for individual lasing states. The far-field speckle pattern of a lasing state is determined by its field pattern at the sample surface. The angular distribution of the outgoing field $E(q)$ is the Fourier transform of the field pattern $E(x)$ at the surface: $E(q) = \int E(x)\exp(i2\pi qx)\,dx$, where $q \equiv \sin\theta/\lambda$, θ is the angle between the emission direction and the normal to the sample–air interface, x represents the transverse coordinate at the sample surface. The angular distribution of the emission intensity $I(q) \equiv |E(q)|^2$ can be obtained experimentally from the far-field speckle pattern. The spatial field correlation function at the sample surface $C_E(x) \equiv \int E^*(x')\,E(x+x')\,dx'$ is the Fourier transform of $I(q)$, i.e., $C_E(x) = \int I(q)e^{-i2\pi xq}\,dq$. Therefore, the speckle pattern of a lasing state gives its spatial field correlation function.

Experimentally, the emission from a PMMA sample was collimated by a lens and directed to a spectrometer. The two-dimensional CCD array detector, mounted at the exit port of the spectrometer, recorded the speckle image. From the image, we extracted the speckle pattern for each lasing peak. Using the discrete Fourier transform, we obtained the spatial field correlation function

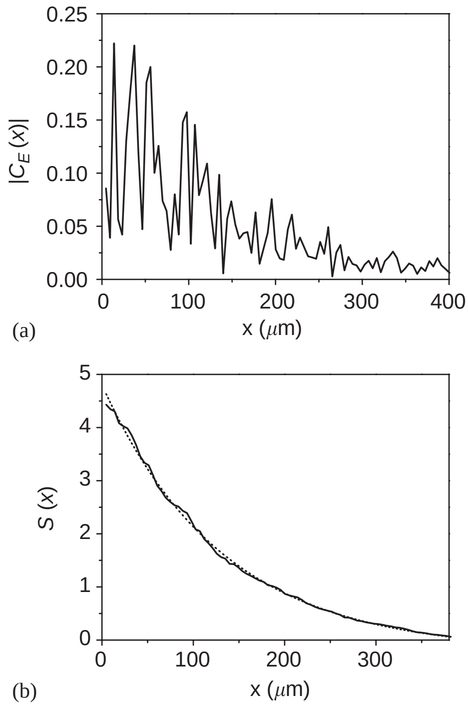

Fig. 2. (a) The amplitude of spatial field correlation function $C_E(x)$ for a lasing state. (b) The solid line is $S(x)$ obtained by integrating $|C_E(x)|$ in (a). The dotted line represents the fitting of an exponential decay function.

$C_E(x)$. Fig. 2(a) shows the amplitude of $C_E(x)$ for a lasing state at $\lambda = 622.8$ nm. The profile of $|C_E(x)|$ has some peaks and valleys. It reflects the field pattern $E(x)$. By integrating $|C_E(x)|$, we get a smoother function $S(x) \equiv \int_x^\infty |C_E(x')|\,dx'$, as shown in Fig. 2(b). The dotted line in Fig. 2(b) represents the fit of $S(x)$ with an exponential decay function, $S(x) = A\exp(-x/l_d)$, where A and l_d are the fitting parameters. $S(x)$ fits very well with the exponential decay function, $\chi^2 = 0.0026$. The decay length $l_d = 131.8$ μm. We also fit $S(x)$ with an algebraic decay function $S(x) = c_1/x^{c_2}$, where c_1 and c_2 are the fitting parameters. The fit is very bad, $\chi^2 = 0.46$. For different lasing peaks, the far-field speckle pattern is different, so is the spatial field correlation function. Nevertheless, $S(x)$ always fits well with an exponential decay function, but not an algebraic decay function. The decay length varies from peak to peak. The exponential decay of $S(x)$ suggests that the envelope of the spatial field correlation function falls exponentially with x, i.e., $|C_E(x)| \sim \exp(-x/l_d)$. In the samples with shorter transport mean free path, the average decay length of the lasing states is shorter.

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