

Photon Localization Laser: Low-Threshold Lasing in a Random Amplifying Layered Medium via Wave Localization

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We demonstrate low-threshold lasing in random amplifying layered medium via photon localization. Lasing is facilitated by resonant excitation of localized modes at the pump laser wavelength, which are peaked deep within the sample with greatly enhanced intensity. Emission occurs into long-lived localized modes overlapping the localized gain region. This mechanism overcomes a fundamental barrier to reducing lasing thresholds in diffusive random lasers, in which multiple scattering restricts the excitation region to the proximity of the sample surface.

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In conventional lasers, emission is stimulated into well-defined cavity modes and emerges as a coherent beam. Care is taken to suppress scattering within the cavity since this would shorten the photon residence time in the lasing mode and thereby raise the excitation power required to initiate lasing. In the opposite limit of a random amplifying medium, however, multiple scattering impedes the flow of light out of the gain region [1,2]. This has led to a search for lasing action in random amplifying media, which was observed in rare earth [3] and semiconductor [4] powders, colloidal suspensions in dye solutions [5,6], and organic films [7]. However, multiple scattering also impedes the flow of the incident pump light into the sample [8]. This creates a shallow gain region and allows subsequently emitted photons to escape promptly as diffuse luminescence. As a result, the lasing threshold in these diffusive systems is not appreciably lower than the power level at which stimulated emission surpasses spontaneous emission in a neat dye solution.

In this Letter, we show that a collimated laser beam is produced at greatly reduced lasing threshold in a stack of microscope cover slides with interspersed dye films. The threshold is sufficiently low for lasing to be induced with a quasi-cw illumination by a 3-watt laser beam from an argon-ion laser. Pump energy is deposited deep within the sample, where it is greatly enhanced, by the resonant excitation of spectrally narrow localized modes. Subsequent emission occurs into long-lived localized modes which spatially overlap the region of molecular excitation. This is demonstrated by the strong correlation of pump transmission and output laser power.

The sample is a stack of L partially reflecting glass slides of random thickness between 80 and 120 μm with an amplifying region or an air gap between the slides. The amplifying layers consisted either of Rhodamine 6G laser dye solution in ethylene glycol between slides separated by 25 μm diameter optical fibers, or of a single 0.3-mm-thick chromophore-doped plastic sheet. The stack was illumi-

nated at normal incidence by a 150 ns pulse from a frequency-doubled, Q -switched Nd:YAG laser at 532 nm focused to a 100 μm spot. A schematic picture of the experimental setup is shown in Fig. 1(a).

Light in this one-dimensional (1D) geometry is localized by multiple scattering from the parallel layers which returns the wave upon itself [9–12]. Emission spectra, recorded with a 0.07-nm-resolution spectrometer, are shown in Fig. 1. The broad emission spectrum of the neat dye solution [dashed curve in Fig. 1(b)] is compared with the emission spectrum from the random stack with interspersed dye layers slightly below and above the lasing threshold. Below threshold, the spectrum shows resolution-limited peaks of the electromagnetic modes of the system [lower curve in Fig. 1(b)]. Above threshold, a collimated emission beam perpendicular to the sample layers was observed [middle curve in Fig. 1(b), shifted up for clarity]. The abrupt change in the output power with increasing pump energy shown in the inset of Fig. 1(d) is characteristic of the lasing transition. At low pump energies lasing occurred in a single narrow line [Figs. 1(b) and 1(c)], while at higher energies multimode lasing was observed [Fig. 1(d)] with wavelength and intensity that vary randomly with the position of the pump beam on the sample surface. The lasing threshold depended strongly upon the dye concentration, sample thickness, and location of gain inside the stack. The lowest threshold of $\sim 0.25 \mu\text{J}$ was measured with a single dye layer with a concentration of 4 mg of dye in 1 ml of ethylene glycol at the center of a stack of 60 glass slides. This pulse energy corresponds to the peak pump intensity of $\sim 10 \text{ kW/cm}^2$, which can be obtained from a standard continuous-wave laser source. Indeed, single and multimode lasing was observed using the output of a 3-watt argon-ion laser which was chopped into 2 μs pulses by an acousto-optic modulator to prevent overheating of the sample [dashed curve in Fig. 1(c)].

The distinct spontaneous emission and lasing peaks, shown in Fig. 1, contrast with the continuous emission

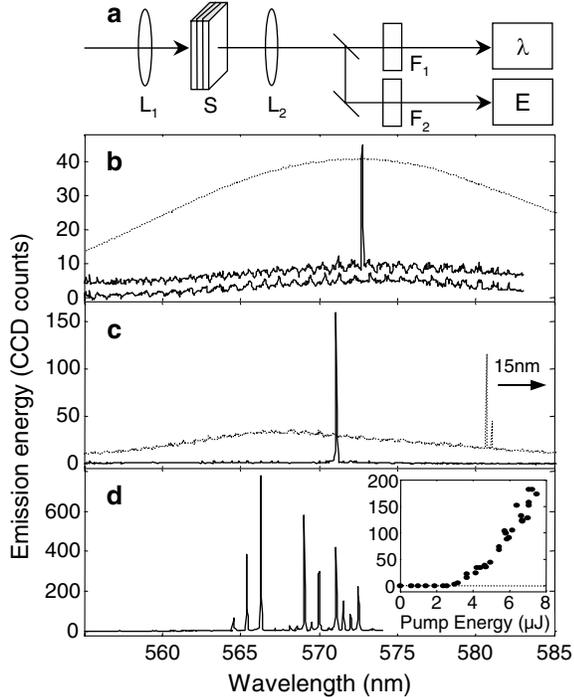


FIG. 1. (a) Scheme of the experimental setup: $L_{1,2}$, focusing and collection lenses; S, a stack of glass slides with gain medium inside; $F_{1,2}$, color filters for blocking the pump and fluorescence light, respectively; λ , spectrometer; E, energy meter. (b)–(d) Emission spectra of the neat dye solution and of localization laser (see text), for the following experimental parameters: (b) single $25 \mu\text{m}$ dye layer (2 mg of dye in 1 ml of ethylene glycol) and $L = 50$ glass slides; pump energy, $E_{\text{pump}} = 110 \mu\text{J}$ and $130 \mu\text{J}/\text{pulse}$ for the lower and middle curves, respectively. (c),(d) $L = 100$ slides, every other air gap filled with the same dye solution; $E_{\text{pump}} = 3.2 \mu\text{J}$ and $4.9 \mu\text{J}/\text{pulse}$ for (c) and (d), respectively. Dashed curve in (c) corresponds to different gain medium (colored plastic sheet) in the middle of a stack excited by a chopped Ar-laser beam. The inset in (d) shows the total emission energy integrated over the spectral region 550–590 nm.

spectrum of the diffusive random laser [5] and is characteristic of localized electromagnetic modes. In localized media, the spectral width of a mode, $\delta\nu$, which is the inverse of the escape time of photons from the medium, is smaller than the typical spacing between modes, $\Delta\nu$. This gives a value below unity for the Thouless number, δ , which is the ratio of the level width to the level spacing, $\delta \equiv \delta\nu/\Delta\nu < 1$ [10,12,13]. Narrow emission modes have also been observed recently in micron-sized diffusive ZnO clusters [14] and organic thin films [7]. Although $\delta > 1$ in these samples, its value approaches unity when the photon excursion volume approaches the coherence volume $V_c = (\lambda/2)^3$. In the presence of substantial gain, emission into a few long-lived modes of the microscopic sample is enhanced, giving rise to distinct spectral peaks [15]. In contrast, the overlap of many modes in macroscopic diffusive lasers results in nonresonant feedback and a continuous emission spectrum [2]. Because of the large number of

competing modes within the excitation volume, the number of stimulated photons in each mode is low. As a result, the lasing threshold is high, and only a modest spectral narrowing is achieved.

The small value of δ in random layered media reflects weak coupling between adjacent regions of a localized sample, which leads to an exponential falloff of the average transmittance with sample thickness, $\langle T(L) \rangle$ [9,10]. Excellent agreement is found in a comparison of the measured average transmittance over a 10 mm^2 area of the sample and scattering-matrix simulations [16] for random ensembles of 1D samples for $L \leq 60$ slides (Fig. 2). The measured transmittance contains many realizations of longitudinal disorder as a result of the transverse inhomogeneity of the glass slides of $\sim 200 \text{ nm}/\text{mm}$. This produces a random speckle pattern in the transmitted field. In samples with $L \leq 60$ slides, the observed width of the speckle spots is greater than the spread of the wave as it propagates through the sample. As a result, the transmitted field explores an area over which it is coherent, and the disorder explored by a wave traversing the sample may appropriately be treated as one dimensional. In contrast, the wave spread in thicker samples as determined either experimentally or from calculations utilizing the measured transmission linewidth [16] becomes bigger than the speckle size. A single exponential fit to the measured transmittance, $\langle T(L) \rangle \sim \exp(-L/2\xi)$, for $25 \leq L \leq 60$ slides gave the localization length $\xi = 13.5$ slides.

Even though the average optical transport is suppressed, resonant tunneling through localized states provides spectrally narrow modes, in which transmission can be high [10,12,17]. A large number of narrow modes was observed in measurements [Fig. 3(a)] and in scattering-matrix calculations [Fig. 3(b)] of transmittance for random stacks of 50 glass slides. The laser beam was focused to an $80 \mu\text{m}$ diameter spot. Stronger focusing was avoided in order to prevent spectral line broadening due to the angular spread of the incident beam. High spectral resolution was obtained using a single-mode tunable dye laser with a linewidth of $\sim 1 \text{ MHz}$ ($\Delta\lambda \approx 10^{-6} \text{ nm}$). In the spectrum shown in

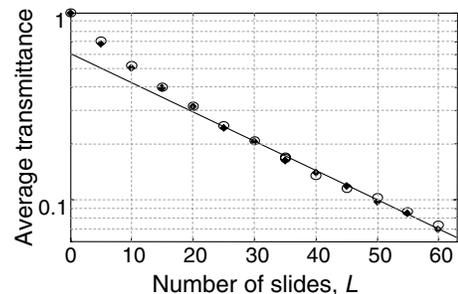


FIG. 2. Average transmittance through a stack of glass slides without intervening dye solution. Experimental results (circles) are compared with the results of numerical calculations (diamonds), in which averaging was performed over 100 sample realizations of the glass stack with uniform distribution of the slide thickness between 80 and $120 \mu\text{m}$.

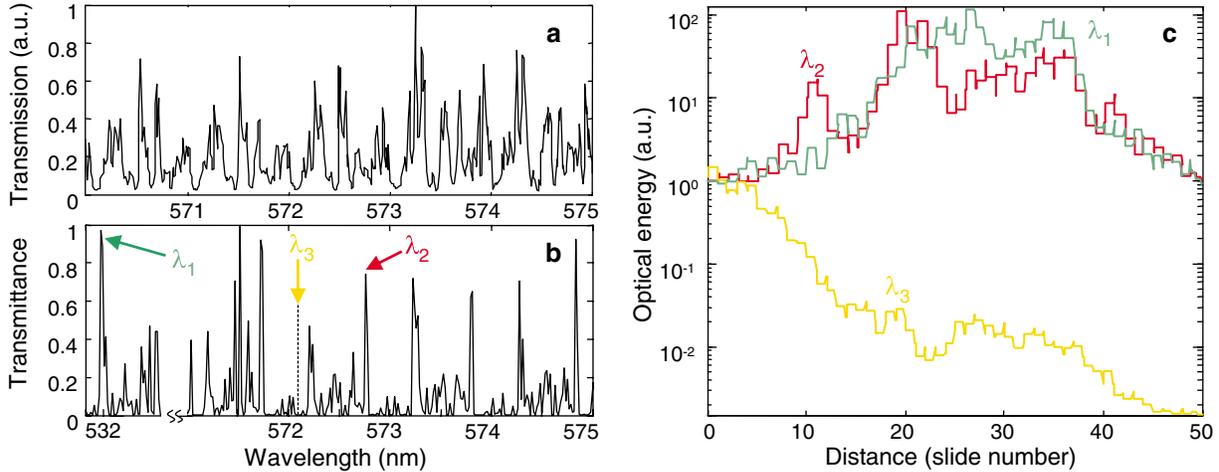


FIG. 3 (color online). (a) High-resolution experimental spectrum of transmission through a stack of 50 glass slides. (b) Results of a scattering-matrix calculation of the transmittance spectrum for a stack of 50 slabs of random thickness illuminated by a plane wave, with the instrumental resolution taken into account. (c) Calculated spatial energy distribution of a plane wave incident upon the sample in (b) for the three wavelengths indicated. A stepwise change in energy density occurs at the air-glass interfaces.

Fig. 3(a), the laser was scanned in 0.01 nm steps. Measurements of the width of a typical narrow isolated transmission line using scans with step size of 10^{-4} nm gave $\delta\lambda = 4.3 \times 10^{-3}$ nm ($\delta\nu = 4.0$ GHz). This spectral width is smaller than an average mode spacing in a 5-mm-thick stack, corresponding to the expected $\delta < 1$. Narrow transmittance lines, which correspond to long-lived spatially localized modes [12,18], occur at both the excitation and emission wavelengths. The highest transmittance found in simulations is of order unity and occurs at the wavelength corresponding to the peaks of the narrowest lines at which the intensity is exponentially enhanced near the center of the layered medium, as shown in Figs. 3(b) and 3(c). The spatial distribution of the pump energy at wavelength λ_1 inside the layered structure demonstrates the deep penetration of the pump energy into the sample's interior when the pump laser is tuned to resonance with a localized mode. Energy absorbed from this mode is subsequently emitted into long-lived localized modes which fall within the dye emission spectrum, e.g., at wavelength λ_2 . Stimulated emission is enhanced when the spatial energy distributions at both the excitation and emission wavelengths overlap. In contrast to the resonantly excited modes at λ_1 and λ_2 , the energy of nonresonant light at wavelength λ_3 falls exponentially within the sample. The deposition of pump energy deep within the sample and its efficient coupling to long-lived emission modes removes a major barrier to achieving low-threshold lasing in the presence of disorder. Serendipitously, the increased penetration of the excitation light entails an exponential increase in the pump intensity near the center of the sample. This produces population inversion even at low incident power.

Since lasing occurs via the excitation of narrow localized modes, large fluctuations in the output lasing power may be expected as the laser is focused on different parts of

the sample, corresponding to different realizations of disorder [18]. This is seen in the plot of the total energy emitted into a solid angle of 0.2 sr following excitation by 10 μ J pulses versus translation of the sample in the plane of the glass slides (Fig. 4). The total laser power was integrated over the wavelength range of 555–590 nm. The simultaneously measured pump transmission (dashed curve) is seen to be strongly correlated with the output laser power. This is contrary to the expectation for a diffusive amplifying medium where strong transmission is associated with low output laser power since it indicates weak absorption of the pump laser. In the present case, strong transmission is expected when the pump laser resonantly excites a localized mode which is spatially peaked near the center of the sample. Pump energy is therefore exponentially enhanced within the sample (Fig. 3) and is efficiently transferred to the gain medium. Since excitation in the center of the sample is likely to escape via emission into localized modes with long photon residence time, the opportunity for stimulated emission is enhanced and the laser output power is high. Because both strong transmission at the pump wavelength and high output laser power are associated with the excitation of localized modes in the center of the sample, these quantities are strongly correlated. Unlike the case of random diffusion lasers, low transmission in this system corresponds to strong back-reflection of the pump beam, and therefore to low absorption in the gain medium and inefficient lasing.

The crossover from nonresonant to resonant feedback in random lasers as the parameter δ falls below unity parallels the transition between wave diffusion and localization in random media at the localization threshold of $\delta = 1$ [10]. Indeed, the inverse of the Thouless number δ is the average number of returns of a wave to a coherence volume V_c within a disordered sample [13]. Thus, the probability of a wave trajectory returning upon itself is small in a diffusive

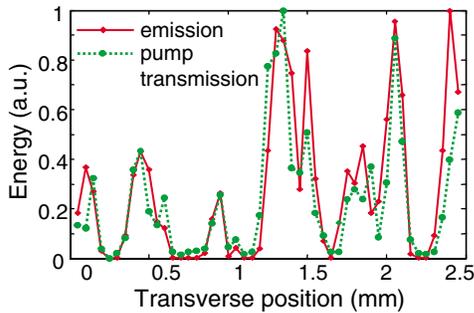


FIG. 4 (color online). Spatial variation of transmitted energy of the pump beam with fixed input power of $10 \mu\text{J}/\text{pulse}$ (dashed curve), and of the total laser energy (solid curve) for a stack of 50 glass slides with a single layer of dye solution in the center.

medium with $\delta > 1$. Wave interference may therefore be neglected in calculating average optical propagation. The evolution of light within such an amplifying diffusive medium may then be described as a random walk of photons at the pump and emission wavelengths, corresponding to nonresonant feedback [19]. In contrast, the condition $\delta < 1$ corresponds to multiple returns to a coherence volume which lead to photon localization due to wave interference. At the same time, $\delta < 1$ (i.e., $\delta\nu < \Delta\nu$) indicates that modes are decoupled and that lasing may occur due to resonant feedback in a single well-defined mode.

The photon localization laser described here is of simple design, which can be literally slapped together. A collimated beam emerges normal to the surface since lasing is initiated only in the longest-lived modes, which spatially overlap the gain region. Low-threshold, narrow line lasing was observed in samples in which L is not much larger than ξ . Since the mode lifetime is expected to grow exponentially with the number of layers in a 1D sample, greatly improved performance may be anticipated in samples with larger ratio L/ξ , which may be produced, for example, in binary thin films. When the transverse spread of a wave becomes comparable to the transverse coherence length set by inhomogeneity of the layers [16], the sample can no longer be regarded as one dimensional and the role of transverse disorder must be considered.

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