Unidirectional High Intensity Narrow-Linewidth Lasing from a Planar Random Microcavity Laser

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Lasing was achieved in a new type of random laser: the planar random microcavity laser. The laser consists of a planar microcavity with a random gain layer. Optical confinement by the two-dimensional random cavity and the one-dimensional planar microcavity drastically reduces the number of resonant modes. As a result, the laser output is highly directional (the divergence angle is 1.68°) with an extremely narrow-linewidth and ultralow threshold. All these phenomena are explained in terms of the coupling of random cavity modes and planar microcavity modes. The results demonstrate an important step towards applications of random lasers.

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The control of spontaneous emission using microcavities has attracted a great deal of theoretical and experimental attention in the last two decades for its applications in novel light-emitting diodes, laser diodes, monitors, and almost-threshold-less lasers [1-3]. Suppression and enhancement of spontaneous emission in wavelength-size cavities have been demonstrated in a variety of materials such as semiconductors, organic dye solutions and films, organic semiconductor films, and quantum dots [4-8]. Generally, a planar microcavity laser consists of two mirrors (a distributed Bragg reflector, DBR, is frequently used) and a thin layer that serves as the gain medium. As mirrors only provide one-dimensional optical confinement, the emitted light is very divergent. To take organic microcavities as an example, the divergence angle is 30 to 80 degrees [9–11]. Further optical confinement can reduce the divergence of the light emission from such 1D microcavities. In vertical cavity surface emitting lasers (VCSELs), lasing divergence is reduced by controlling the cavity transverse aperture in a range from several microns to several tens of microns. In photonic crystal microcavity lasers, in-plane confinement is achieved through the photonic band gap generated from twodimensional periodic structures; only light with a vertical wave vector can escape the cavity.

The random laser is a microcavity whose feedback derives from disorder-induced scattering [12]. Extremely narrow linewidth and threshold were observed in disordered ZnO powder with coherent feedback [13]. The smallest size of a random cavity is about 1 μ m. A random laser in a waveguide was reported soon after [14–16]. Random lasers have potential applications due to their small volume, easy fabrication, and low cost compared to sophisticated structures like VCSELs and photonic band gap microcavities. However, isotropic laser emission and high threshold hinder their application.

Efficient optical confinement is necessary to reduce the number of modes in a random laser and thereby improve its

spectral and spatial characteristics. The threshold of a mmthick random laser can be drastically suppressed by placing a mirror close to the random medium [17–19]. Feedback also helps to improve the optical properties of random waveguide lasers [18].

In this Letter, we propose a new structure: the planar random microcavity laser. The structure consists of a traditional planar microcavity and a random gain layer. Therefore, the new structure is a combination of a planar microcavity laser and a random laser. For a random gain layer in a micron-thick film, random modes only form in the film plane; vertical resonances are totally determined by the planar microcavity. Since the random cavity and planar microcavity provide twofold optical confinement, the number of modes with comparable decay rates can be significantly reduced. As a result, the new laser showed narrow divergence, ultralow lasing threshold, and very narrow linewidth.

Experimentally, our planar random microcavity is a 2.5 μ m thick organic/inorganic hybrid film that is sandwiched between two DBRs. The planar microcavity was fabricated according to our previous description but with some modifications [10]. First, a 10-period TiO₂/SiO₂ multilayer DBR was fabricated on a glass substrate. Then an organic-inorganic sol-gel film was dip coated on the DBR. In preparing the sol-gel gain material, Rodamine B (5 wt%) dye, together with silica spheres (150 nm in diameter) of various concentrations were added. After coating the gain layer, the structure was capped with another 8-period TiO₂/SiO₂ multilayer DBR. The refractive indices of the gain film and silica spheres are 1.52 and 1.45, respectively.

The normal-reflectance spectrum of the cavity is plotted in Fig. 1(a). It has a stop band gap between 570 and 680 nm. Two sharp defect resonant modes appear at 591 and 625 nm with 7 nm linewidth (FWHM).

For photoluminescence measurements, the samples were optically pumped by the frequency-doubled output



FIG. 1. (a) The normal reflection spectrum of the microcavity. (b) The photoluminescence spectrum of the same microcavity.

of a mode-locked Nd:YAG laser (532 nm, 10 Hz repetition rate, 35 ps pulse width). The pump beam was focused to a 70-micron spot with an incident angle of 60° away from the surface normal. The emitted light was collected by a fiber bundle in normal direction and coupled to a SPECTROPRO 500i (ACTON RESEARCH, f = 0.5 m) monochromator and detected by a DH 720-18F-03 ICCD (ANDOR Technology).

Figure 1(b) shows a typical photoluminescence spectrum obtained by pumping at 4 μ J/cm², which is higher than the lasing threshold (=2.5 μ J/cm², see below). The peaks at 590 and 624 nm correspond to the resonance modes in Fig. 1(a), but the linewidths of the peaks are obviously much narrower. Looking with higher spectral resolution (See the inset a of Fig. 2), the resonant peaks were actually comprised of three narrower discrete peaks with linewidth $\Delta\lambda$ about 0.12 nm (60 times smaller than the linewidth of the passive defect mode). For comparison, inset b in Fig. 2 shows the emission spectrum when the cavity was pumped below threshold. The resonance peak linewidth is about 7 nm, which is consistent with that of the passive microcavity [10,20,21]. Moreover, peak numbers and their wavelengths depend on the pump position. In other words, pumping a different spot changes the resonant spectral profile and narrow-peak positions. This is clear evidence that the modes have a random nature.

The dependence of the emitted light power on pumpenergy fluence (Fig. 2) indicates a threshold at $E_{\rm th} = 2.5 \ \mu J/{\rm cm}^2$. This threshold is five to 6 orders of magnitude smaller than the threshold of a bare random gain layer without planar DBR feedback. Strong feedback and enhancement of the spontaneous emission in the planar microcavity and the overlap of the pump/lasing region are the key factors resulting in the threshold reduction.

On the other hand, the dependence of the lasing threshold on the density of silica spheres gives direct evidence of threshold reduction due to transverse optical trapping.



FIG. 2 (color online). Dependence of the output energy on the input pump energy near threshold for the microcavity. Insets: (a) high-resolution spectrum of the 590 nm resonant peak in Fig. 1(b) at a sample pump fluence of 4 μ J/cm². (b) The emission spectrum from the microcavity pumped at 1 μ J/cm², i.e., below the lasing threshold.

This is shown in Fig. 3. The laser threshold is almost constant when the density is larger than $7.5 \times 10^{12}/\text{cm}^3$ (about 500 nm sphere-sphere spacing). When the density of spheres decreases from $1.5 \times 10^{13}/\text{cm}^3$ to $1.875 \times 10^{12}/\text{cm}^3$, the threshold increases from about $13 \ \mu\text{J/cm}^2$ to $1.97 \times 10^4 \ \mu\text{J/cm}^2$. Meanwhile the divergence remains at about 1.7° . No directional random lasing occurs when the density of silica spheres is decreased further. The result clearly reveals that, when the scattering length is close to the emitted wavelength, coherent scattering will be strong enough to efficiently trap the light inside the random cavity. Consequently, the lasing threshold drops drastically.

Near-field and far-field emission patterns also provide information about optical confinement. The near-field emission patterns from the cavity were recorded by imaging the luminescent spot with a $40 \times$ objective lens onto a



FIG. 3. The dependence of the lasing threshold on the density of silica spheres.



FIG. 4 (color online). The near-field images of the spatial distribution of the emission intensity in the microcavity with pump fluence of (a) 2.5 and (b) 5.0 μ J/cm². (c) The near-field image of the spatial distribution of the emission intensity in the common planar microcavity at a pump intensity of 6000 μ J/cm².

CCD camera. At 2.5 μ J/cm² pump power (lasing threshold), a hot spot appears [Fig. 4(a)]. When the pump intensity increases to 5 μ J/cm², additional hot spots appear [Fig. 4(b)]. Figure 4(c) shows the near-field spatial distribution of a planar microcavity without silica spheres in the gain layer and pumped at 6000 μ J/cm². It has a much more uniform pattern across the whole detected area. This pattern is similar to that when the planar random microcavity is pumped below E_{th} . Thus we come to the conclusion that for a planar cavity with scattering spheres, random lasing occurs at very low threshold, and different regions have different thresholds due to the randomness of the scattering.

When the sample was pumped above E_{th} , light was emitted from the surface normal as a well-collimated laser beam. The far-field emission pattern was recorded by placing a screen 13 cm away from the cavity in the normal direction. Figure 5(b) shows the far-field image at the pump fluence $E = 276 \ \mu\text{J/cm}^2$. The diameter of the



FIG. 5 (color online). (a) Schematic illustration of the mechanism of the planar random microcavity laser, (b) the far-field image of the laser beam detected at 13 cm away from the planar random microcavity laser.

bright center spot was about 0.38 cm, which means the emission from our devices is strongly forward directed and the beam divergence amounts to only 1.68°. The sample was also pumped with a 4 mm pump laser spot, the laser still emits directionally from the cavity. This means that the directional emission is not caused by the small pump area.

The well-collimated laser beam emission was considered to come from the coupling of the two types of cavity modes (planar microcavity and random laser cavity). In a conventional planar microcavity, which consists of two DBR mirrors and a uniform gain layer, top-emitting mode, leakage mode, and transverse waveguide modes exist simultaneously. In the planar random microcavity laser, however, the random gain layer can be considered as a two-dimensional (2D) random laser; random cavities can be envisaged as rings with dielectric constant larger than the average value [22]. Then the whole structure can be simplified to 2D random ring cavities covered with two DBR mirrors. Each composition of random cavity and planar microcavity mode can be considered naturally as a mini-VCSEL laser. When in resonance, the optical wave vector can be decomposed to K_z and K_t , where K_z is the vertical component determined by the planar microvaity mode and K_t is the transverse component determined by the random ring cavity modes. As the planar microcavity length is much smaller than the random cavity length, K_{7} should be much larger than K_t when fundamental modes are considered [23]. Therefore light can only emit in the vertical direction with a very small divergence angle.

Our experimental results yield a physical explanation of V. Bulovic's work on directional emission from a planar organic microcavity [24]. Considering the technique for the synthesis of their active medium and their ASE laser spectrum [25], some density fluctuations were considered to resemble the scattering by silica spheres as in our experiment.

In summary, we proposed and fabricated a new type of random laser structure: a planar random microcavity laser. The optically pumped hybrid cavity produced a very narrow linewidth with ultralow threshold and unidirectional laser emission. The threshold was lowered to 2.5 μ W/cm², and a bright, collimated laser beam with 1.68° divergence was observed. All the phenomena were concluded to come from the coupling of the random laser microcavity and the planar microcavity.

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