Microlaser made of disordered media

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We have fabricated microlasers with disordered semiconductor nanoparticles. The physical mechanism of optical confinement is based on Anderson localization of light in micrometer-sized random media. Pulsed lasing occurs around 380 nm at room temperature under optical pumping.
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The microlaser has important applications to integrated photonic circuits. Over the past decade, several types of microlasers have been developed. The key issue of the microlaser is to confine light in a small volume with dimensions on the order of the optical wavelength. In the vertical-cavity surface-emitting laser, light is confined by two distributed Bragg reflectors.[1] The microdisk laser utilizes total internal reflection at the edge of a high-index disk to form whispering gallery modes.[2] In a recent work of a two-dimensional photonic band-gap defect-mode laser, the lateral confinement of light is achieved by Bragg scattering in a two-dimensional periodic structure.[3] The fabrication of these microlasers requires expensive state-of-the-art crystal growth and microfabrication facilities. In this letter, we demonstrate a type of microlaser made of disordered medium. The physical mechanism of optical confinement is based on the Anderson localization of light in a micrometer-scale random medium. The fabrication of such a microlaser is much easier and cheaper than that of most microlasers.

The micrometer-sized random material is made of ZnO powder. The ZnO nanoparticles are synthesized with precipitation reaction.[4] The process involves hydrolysis of zinc salt in a polyol medium. Specifically, 0.05 mol of zinc acetate dihydrate is added to 300 ml diethylene glycol. The solution is heated to 160 °C. As the solution is heated, more zinc acetate is dissociated. When the Zn^{2+} concentration in the solution exceeds the nucleation threshold, ZnO nanocrystallites precipitate and agglomerate to form clusters. The size of the clusters can be controlled by varying the rate at which the solution is heated. The inset of Fig. 1 is the scanning electron microscope (SEM) image of a typical ZnO cluster. The ZnO nanocrystallites have an average size of 50 nm. The size of the clusters varies from submicron to a few micron.

The ZnO cluster is optically pumped by the fourth harmonics ($\lambda = 266$ nm) of a mode-locked Nd:YAG laser (10 Hz repetition rate, 15 ps pulse width). The pump light is focused by a microscope objective onto a single cluster. The spectrum of emission from the cluster is measured by a spectrometer with 0.13 nm spectral resolution. Meanwhile, the spatial distribution of the emitted light intensity in the cluster is imaged by an ultraviolet (UV) microscope onto a UV-sensitive charge-coupled-device (CCD) camera. The amplification of the microscope is about 100 times. The spatial resolution is ~ 0.2 μm. A bandpass filter is placed in front of the microscope objective to block the pump light.

We have performed optical measurements of the cluster shown in the inset of Fig. 1. The size of the cluster is about 1.7 μm. It contains roughly 20 000 ZnO nanocrystallites. As shown in Fig. 2(a), at low pump power, the emission spectrum consists of a single broad spontaneous emission speak. Its full width at half maximum (FWHM) is 12 nm. The spatial distribution of the spontaneous emission intensity is uniform across the cluster [see Fig. 2(b)]. When the pump power exceeds the threshold, a sharp peak emerges in the emission spectrum shown in Fig. 2(c). Its FWHM is 0.22 nm. Simultaneously, a couple of bright spots appear in the image of the emitted light distribution in the cluster in Fig. 2(d). The size of the bright spot is ~ 0.3 μm. When the pump power is increased further, a second sharp peak emerges in the emission spectrum [see Fig. 2(e)]. Correspondingly, additional bright spots appear in the image of the emitted light distribution in Fig. 2(f).

Figure 1 plots the emission intensity as a function of the incident pump pulse energy. A threshold behavior is clearly

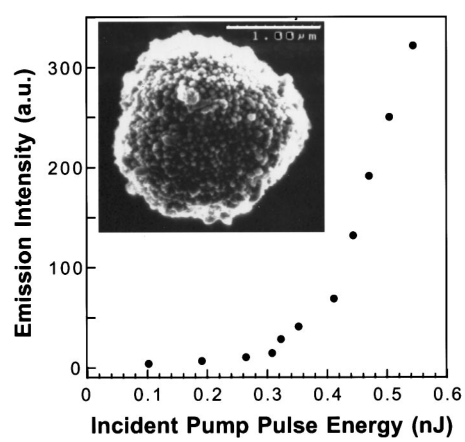

FIG. 1. Spectrally integrated intensity of emission from the ZnO cluster vs the incident pump pulse energy. The inset is the SEM image of the ZnO cluster.

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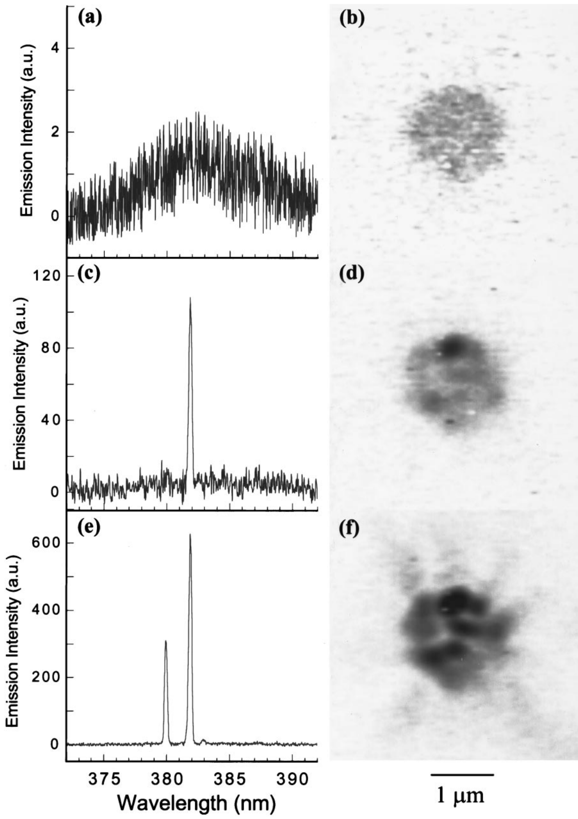

FIG. 2. (a), (c), and (e) are the spectra of emission from the ZnO cluster shown in Fig. 1. (b), (d), and (f) are the corresponding spatial distributions of emission intensity in the cluster. The incident pump pulse energy is 0.26 nJ for (a) and (b), 0.35 nJ for (c) and (d), and 0.50 nJ for (e) and (f).

seen: above the pump power at which sharp spectral peaks and bright spots appear, the emission intensity increases much more rapidly with the pump power. These data suggest that lasing action has occurred in the micrometer-sized cluster. The incident pump pulse energy at the lasing threshold is $\sim$0.3 nJ. Note that around less than 1% of the incident pump light is absorbed. The rest is scattered.

The experimental fact that the bright spots in the emission pattern and the lasing modes in the emission spectra always appear simultaneously suggest that the bright spots are related to the laser light. There seems to be two possible explanations for the bright spots. One is that the laser light at the locations of the bright spots is very strong. The other is that the laser light is not particularly strong at the locations of the bright spots. However, there are some efficient scattering centers at the locations of the bright spots, and thus the laser light is strongly scattered. In the latter case, those scattering centers should also strongly scatter the spontaneous emission below the lasing threshold, because scattering is a linear process. Hence, these bright spots should exist below the lasing threshold. However, there are no bright spots below the lasing threshold. Therefore, these bright spots are caused not by efficient scatterers, but by strong laser light. In other words, the bright spots indicate spatial localization of laser light.

Figure 3 presents the measurement result of a second cluster. The SEM image shown in Fig. 3(a) indicates that this cluster has an irregular shape, and its size is slightly larger than 1 μm. A similar lasing phenomenon is observed in this cluster. The incident pump pulse energy at the lasing threshold is about 0.2 nJ. The FWHM of the emission spectrum narrows dramatically from 12 nm below the lasing threshold to 0.16 nm above the lasing threshold [Fig. 3(b)]. After taking into account the instrumental broadening, the actual linewidth of the lasing mode is only 0.09 nm. Bright spots appear in the image of the laser light distribution in the cluster. By adjusting the microscope objective, the light distribution on different planes inside the cluster is imaged onto the CCD camera. Figures 3(c) and 3(d) are the images of the light distribution on two planes with different depths inside the cluster. Some bright spots appear in one image, but not the other. This suggests that these bright spots are buried with different depths inside the cluster. These images illustrate three-dimensional localization of laser light in the micrometer-sized random medium.

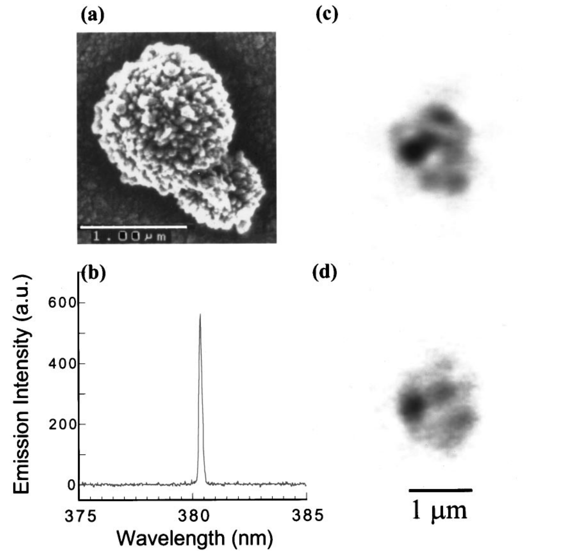

FIG. 3. (a) is the SEM image of a second ZnO cluster. (b) is the spectrum of emission from this cluster above the lasing threshold. The incident pump pulse energy is 0.27 nJ. (c) and (d) are the spatial distribution of emission intensity in the cluster at the same pump power.

The highly disordered structure of the cluster leads to strong light scattering. We characterize the scattering mean-free path in a macroscopic-scale film made of ZnO powder with the coherent backscattering measurement.[5–7] To avoid absorption, the probe photon energy is slightly less than the ZnO band-gap energy (3.3 eV). From the angular width of the coherent backscattering cone, we estimate that the scattering mean-free path is about half of the optical wavelength, after taking into account internal reflection.[8–10] Because of the short scattering length, light can be trapped inside the random medium through the process of multiple scattering and wave interference. This phenomenon is called the Anderson localization of light.[11–14]

For the particular configuration of scatterers in a cluster, photon localization occurs only at certain wavelengths. This is because photon localization is based on the interference effect which is wavelength sensitive. In another cluster, the configuration of the scatterers is different, and thus photon localization occurs at different wavelengths. Because of the finite size of the random medium, photon localization is not complete. Namely, some photons escape through the surface of the cluster. This gives rise to the loss of a localization cavity. When the optical gain exceeds the loss of a localization cavity, laser oscillation occurs at the localization frequency. The spatial distribution of the laser light intensity exhibits the spatial profile of the localization mode. There may be several localization cavities in a cluster. Since different localization cavities have different loss, their lasing thresholds are different. Thus, with an increase of optical gain, lasing occurs in a second localization cavity, leading to an additional peak in the emission spectrum. The simultaneous addition of bright spots in the spatial light pattern not only confirms that lasing occurs in a different localization cavity, but also reveals the spatial profile of the cavity mode.

To confirm that laser cavities are formed by optical scattering, we vary the amount of scattering to see whether the lasing threshold is changed. A simple way of changing the scattering strength is to change the size of the ZnO nanocrystallites, which can be done by annealing. After annealing at 800 °C in ambient argon gas, the average size of the ZnO nanocrystallites is increased from 50 to 150 nm by annealing. We have measured the lasing threshold of the same cluster before and after annealing. For most clusters, the lasing threshold pump power after annealing is more than ten times higher than that before annealing. Some clusters stop lasing after annealing. The increase of the lasing threshold is attributed to a decrease of the localization cavity quality factor. After annealing, the number of nanocrystallites in a cluster is decreased. Thus, the number of scattering events photons undergo is reduced. It is easier for photons to escape through the surface of the cluster. This is equivalent to an increase of the loss of the localization cavities. Hence, the lasing threshold pump power is increased.

In summary, we have demonstrated that light can be localized in micrometer-sized random medium through the process of multiple scattering and wave interference. Based on this discovery, we have fabricated an ultraviolet microlaser with disordered semiconductor nanoparticles. Our result shows that the Anderson localization of light provides a physical mechanism of optical confinement for the microlaser.

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