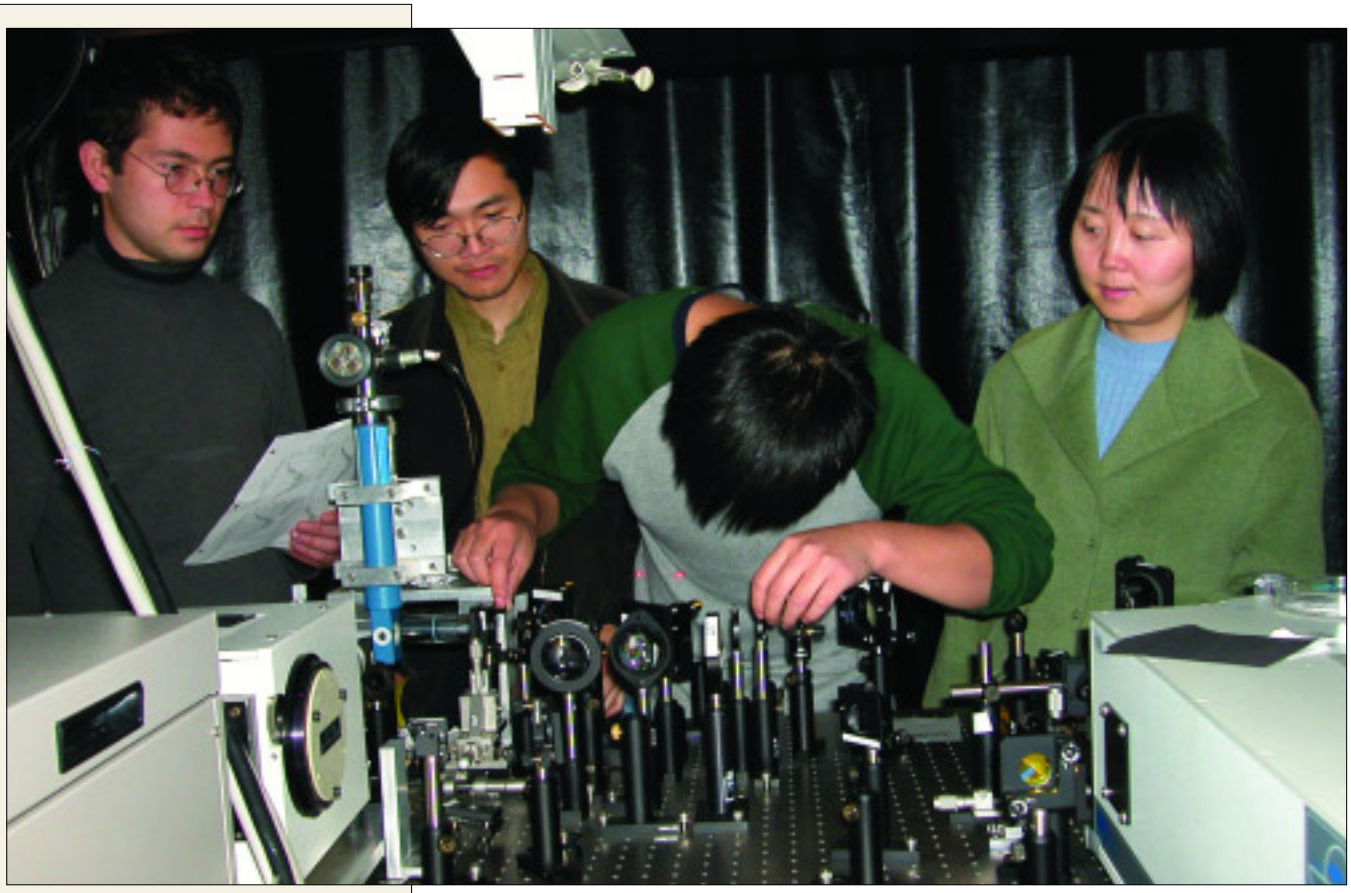

Random LASERS

Development, Features and Applications

Hui Cao

The random laser differs from other types of laser in that its cavity is formed not by mirrors but by multiple scattering in a disordered gain medium. Over the course of the past decade, random lasers have generated significant interest among researchers because their characteristics can lead to unique applications.

(*Above*) Alexey Yamilov (*left*) and graduate students Wei Fang and Xiaohua Wu carry out optical measurements of random lasers in professor Hui Cao's (*right*) laboratory at Northwestern.

Lasers have found widespread use in industry, medicine and other areas of contemporary life. The essential ingredients of a laser are a material that amplifies light through stimulated emission (the gain medium) and a cavity that traps light in the gain medium to enable more efficient amplification. The gain medium is pumped electrically, by a source current, or optically, by another laser source or a lamp.

The most common laser cavity is the Fabry-Pérot, made of two mirrors (one of which is partially transmitting) that face each other on either side of the gain medium [Fig. 1(a)]. Light that is bounced back and forth between the mirrors is amplified each time it passes through the gain medium. Light can leave the cavity through the partially transmitting mirror. Light that remains in the cavity interferes constructively after traveling a round trip between the mirrors and returning to its original position. This means that the phase delay of a round trip must be equal to an integer multiple of 2π, a requirement that can be satisfied only at certain frequencies. At such cavity resonant frequencies, light experiences minimum leakage through the partially transmitting mirror and remains in the cavity for a period of time. When the optical amplification is large enough to compensate the loss caused by mirror leakage and material absorption, lasing oscillations occur at cavity resonant frequencies. The laser light transmitted through the partially reflecting mirror has well-defined frequency, good directionality and a high degree of coherence. If, however, there are scatterers inside the cavity, the light can be scattered in other directions, introducing additional loss and increasing the lasing threshold. This is why optical scattering is considered detrimental and why laser engineers work

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hard to minimize the amount of scattering in laser cavities.

What happens when the scattering is very strong? Surprisingly enough, strong light scattering can facilitate lasing action. Let us consider a gain medium that contains a number of scattering centers [Fig. 1(b)]: light is scattered many times before it escapes the gain medium. The multiple scattering increases the dwell time, or path length, of light inside the gain medium, enhancing light amplification. In this scenario, we no longer need mirrors to trap the light in the gain medium since scattering can do the job on its own. Since strong light scattering usually occurs in highly disordered media, the word "random" has been used to describe lasers that operate on the basis of these properties. In the rest of this article, I describe how random lasers operate, the characteristics that differentiate them from conventional lasers and how their unique features can be applied.

"The photonic bomb"

By replacing one mirror of the Fabry-Pérot cavity with a scattering surface, in 1966 a group led by laser pioneer and Nobel laureate N. G. Basov realized a new type of laser cavity that provided nonresonant feedback.[1] Light in the cavity undergoes multiple scattering. Since every time it is scattered its direction is changed, it does not return to its original position after one round trip. In such a cavity, the spatial resonances of the electromagnetic field are absent and the dwell time of the light is not sensitive to frequency. In such a laser, feedback is used simply to return part of the energy, or photons, to the gain medium, i.e., it is energy, or intensity, feedback. Since in a laser with nonresonant feedback the only resonant element is the amplification line of the gain medium, the mean frequency of emission does not depend on the dimensions of the laser cavity but only on the center frequency of the amplification line. If the frequency is sufficiently stable, the emission is characterized by stable mean frequency that can be used to produce an optical standard for length and frequency.

In 1968, Letokhov took the research further when he proposed self-generation of light in an active medium filled with scatterers.[1] The photons spontaneously emitted by the excited atoms or molecules in the gain medium experience multiple scattering and undergo random walk before leaving the medium. As photons travel in the gain medium, they can induce the stimulated emission of additional photons.

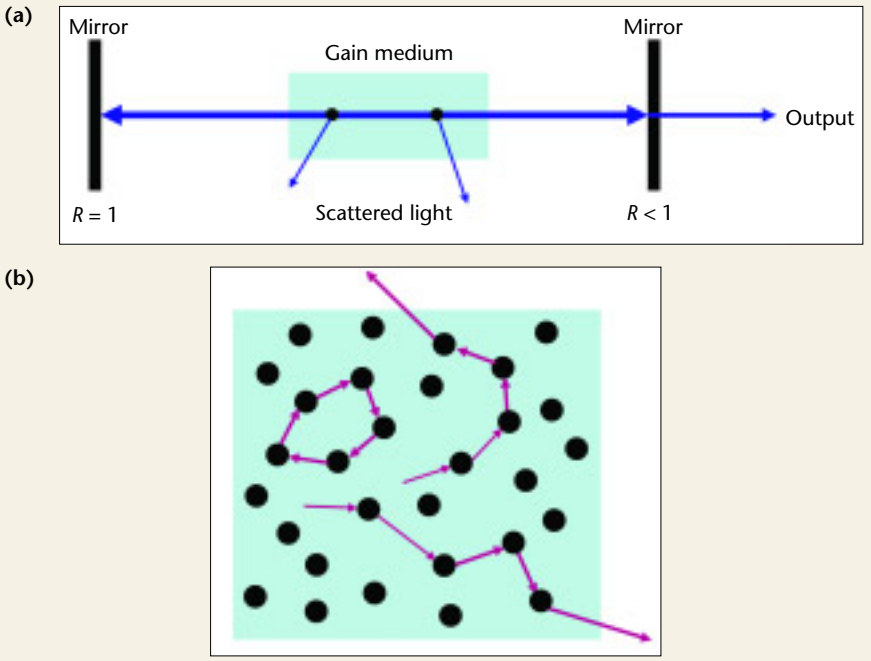


Figure 1. (a) Schematic of a Fabry-Pérot laser cavity made of two mirrors with a gain medium between them. The right mirror is partially transmitting, i.e., its reflectivity $R<1$. The black dots represent the centers that scatter laser light out of the cavity. (b) Multiple scattering increases the path length, or dwell time, of light inside the gain medium. Recurrent light scattering leads to the formation of a closed loop path for light.

There are two characteristic length scales in the scenario first proposed by Letokhov. One is the generation length, L_{gen}, the average distance a photon travels before generating a second photon by stimulated emission. The other is the mean path length, L_{pat}, that a photon travels in the gain medium before escaping. The stronger the scattering, the longer L_{pat}. When L_{pat} exceeds L_{gen}, every photon generates another photon before it escapes the medium. This triggers a chain reaction in which one photon generates two photons, which generate four photons, and so on. The photon number increases with time. Thus, $L_{pat} = L_{gen}$ corresponds to the onset of photon self-generation. Since the process of photon generation is analogous to the process of neutron generation in an atomic bomb, the device proposed by Letokhov has been dubbed a "photonic bomb."

Nearly 20 years after Letokhov's pioneering work in this area, intense stimulated radiation was observed in a wide variety of laser crystal powders, e.g., neodymium-doped glass powder and titanium-doped sapphire powder.[2] When the powder was pumped by laser pulses, the emission pulses from the powder were shortened drastically above a pumping threshold, and spectral narrowing of the emission lines occurred. With further increases in pumping, the emission intensity exhibited damped oscillations. Several explanations were proposed.[3] First, the intense stimulated emission was attributed to the diffusion of light in the powder: it was asserted, in other words, that the photons spontaneously emitted by the doped ions of neodymium or titanium bounced from one particle to another many times before leaving the powder. It was further proposed that it was this multiple scattering that increased the chances of photons hitting excited ions and inducing stimulated emission. It was also speculated that, in

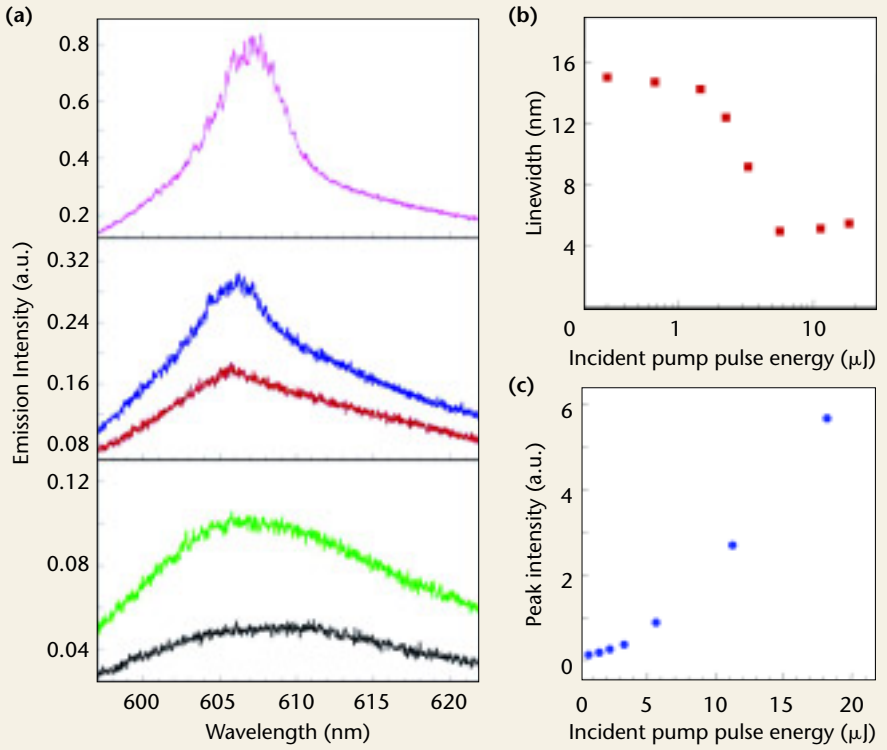


Figure 2. (a) Emission spectra of methanol solution of Rhodamine 640 perchlorate dye and zinc oxide nanoparticles. The suspension is pumped by the second harmonics of a pulsed Nd:YAG laser (λ= 532 nm, pulsewidth = 35 ps, repetition rate = 10 Hz). The incident pump pulse energies are (*top to bottom*): 0.68, 1.5, 2.3, 3.3, 5.6 μJ. The dye concentration is 5 mM per liter. The zinc oxide particle density is 3×10^{11} cm^{-3}. The mean particle diameter is 100 nm. (b) Width of the emission spectrum as a function of the incident pump pulse energy. (c) Intensity at the peak of the emission spectrum versus the incident pump pulse energy.

a powder in which the particle size was much larger than the emission wavelength, light was trapped inside individual particles through surface reflection. In this scenario, the powder was considered an ensemble of optically coupled microcavities and it was theorized that lasing occurred in the modes formed by total internal reflection at the facets of the single grains, or small clusters of them. Finally, because stimulated emission is weak at low doping levels of active ions, the burst of intense radiation was said to be a result of superradiance, or superfluorescence, i.e., the synchronized spontaneous emission of excited ions.

In 1994, Lawandy et al. reported laser-like emission from a methanol solution of Rhodamine 640 perchlorate dye and titanium dioxide microparticles.[4] Unlike the case of the powder samples, the gain medium (dye molecules) and the scattering element (microparticles) were separated in the liquid samples. Optical amplification took place outside the scatterer. The individual particles were about a quarter of a micrometer in size, too small to trap light, and could not serve as laser cavities.

Figure 2 shows the experimental data my group collected to repeat Lawandy's original experiment. The emission spectrum narrows dramatically above a pumping threshold [Fig. 2(a)]. Figure 2(b) illustrates the collapse of the emission linewidth. The input-output plot of the peak emission intensity versus the pump energy in Fig. 2(c) exhibits a slope change at the same pumping threshold. This threshold behavior suggested the existence of feedback. The relatively broad and featureless emission spectrum above the threshold indicated the feedback was frequency-insensitive, or nonresonant. It was found experimentally that the threshold was reduced by more than 2 orders of magnitude when the density of scattering particles was increased from 5×10^{9} to 2.5×10^{12} cm^{-3} at a fixed dye concentration of 2.5×10^{-3} M. The strong dependence of the threshold on the density of scatterers revealed that the feedback was related to scattering.

How could scattering cause the collapse of the emission linewidth? When a spatially broad pump pulse was incident on a dye cell, a disk-shaped amplifying region was formed in suspension near the front window. The photons emitted could easily escape from the amplifying region, either through the front window or deeper into the unpumped region of the suspension. After multiple scattering (or random walk) in the unpumped region, some of the photons returned to the active volume to experience additional amplification. This return process provided energy feedback.

When scattering was stronger, the return probability was higher, so the feedback was stronger. Since the return probability was less than 100 percent, the feedback was incomplete, giving rise to loss. The lasing threshold corresponds to a condition in which the photon loss rate is balanced by the photon generation rate in the amplifying region. The photon generation rate was highest at the peak of the gain spectrum, while the photon loss rate was nearly frequency independent. As the pumping rate increased, the photon generation rate in the frequency region of maximum gain was the first to reach the photon loss rate, while outside this frequency region, the photon generation rate was still below the loss rate. Under these conditions, the photon density around the frequency of gain maximum built up quickly. The sudden increase of photon density near the peak of the gain spectrum resulted in collapse of the emission linewidth.

Note that the feedback caused by multiple scattering is nonresonant and incoherent, i.e., it returns light only to the gain volume instead of to its original position; for this reason, the phase of the scattered light can be ignored. This type of laser is called a random laser with nonresonant, or incoherent, feedback. It has also been dubbed a painted-on laser or a photonic paint laser.

The work of Lawandy et al. has triggered many experimental and theoretical studies of random lasers with nonresonant feedback. Random lasers have been realized in a number of material systems, e.g., polymers, liquid crystals and even biological tissues. R. R. Alfano's group reported random laser action in dye-treated chicken legs and pig hearts.[5] Since there is already strong scattering in animal tissue, the researchers needed only to inject dye molecules into the tissues and pump them to provide optical gain.

Coherent random lasers

A few years ago, I reported a different kind of lasing process in highly disordered semiconductor nanostructures[3]: in contrast to the random laser described in the preceding section, the feedback is resonant and coherent. Z. V. Vardeny's group observed similar phenomena in disordered polymers and organic materials.[3] This kind of random laser, with resonant—or coherent—feedback, has also been dubbed a "coherent random laser." To illustrate how it differs from a random laser with nonresonant—or incoherent—feedback, let's consider the case of zinc oxide nanorods.

Zinc oxide (ZnO) is a wide bandgap semiconductor that emits light in the near-ultraviolet (near-UV) regime. Figure 3(a) is a scanning electron microscope image of ZnO nanorods grown on a sapphire substrate by metalorganic chemical vapor deposition. The nanorods are uniform in height and randomly located on the substrate. The average rod diameter is 50 nm. To induce optical gain, the frequency-tripled output of a pulsed Nd:YAG laser (λ = 355 nm, pulsewidth = 30 ps, repetition rate = 10 Hz) was focused from above by a lens onto the nanorods. The nanorods served as both gain medium and scattering element. Figure 3(c) shows the emission spectra of the ZnO nanorod array. At low pumping, the spectrum exhibits a single broad spontaneous 12 nm-wide emission peak. When the pumping intensity exceeds a certain threshold, discrete narrow peaks emerge on top of the broad spontaneous emission peak. The linewidth of these peaks is less than 0.2 nm, much smaller than the width of the spontaneous emission peak. When the pump intensity is increased further, additional narrow peaks appear. The frequencies of the discrete peaks depend on the sample position. When a different part of the sample is pumped, however, the peak frequencies change. This suggests that the discrete spectral peaks result from spatial resonances for light determined by the local configurations of ZnO nanorods.

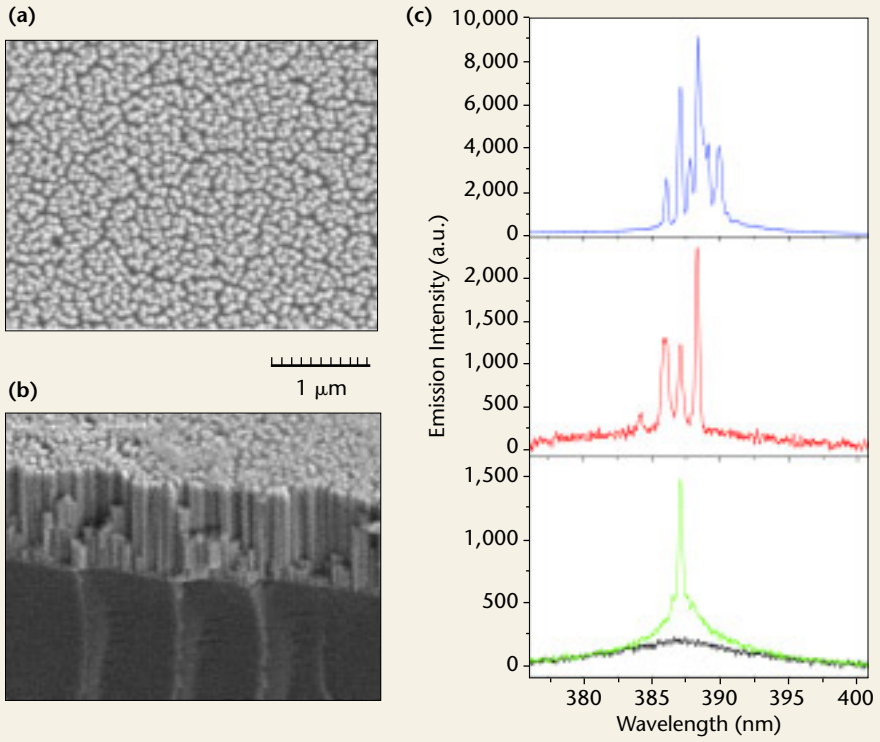


Figure 3. Top view (a) and tilt view (b) scanning electron micrographs of ZnO nanorods grown on a sapphire substrate. (c) Emission spectra of the ZnO nanorods when the incident pump power is (*bottom to top*) 3.2, 4.5, 6.1, 7.0 and 11.1 μJ/cm^2.

The ZnO nanorod array is a two-dimensional (2D) scattering system, i.e., light is scattered by the nanorods in the plane perpendicular to the rods. After multiple scattering, light can return to a nanorod from which it was scattered, before forming a closed loop. This process is called recurrent light scattering [see loop in Fig. 1(b)]. When the optical amplification along the loop exceeds the loss, lasing oscillation occurs in the loop that serves as the cavity. As in the case of a typical ring laser, the lasing frequencies are determined by the condition that the phase delay along the closed loop is equal to an integer multiple of 2π. This picture of lasing created by recurrent light scattering in a single loop is intuitive but somewhat simplistic: in fact, light can return to its original position through many different paths. Since all the backscattered waves interfere, the feedback is field, or amplitude, feedback. Only at certain frequencies is the interference constructive, making it possible to confine light inside the random system. Lasing occurs at these frequencies, producing discrete peaks in the emission spectrum. Above the lasing threshold, emission intensity increases much more rapidly with pumping intensity, while the emission pulse length is reduced from 200 ps to nearly 20 ps. In the photon counting experiment, we confirmed that the laser emission from the random array of ZnO nanorods is indeed coherent. This result is counter-intuitive, in the sense that it shows coherent light can be generated from an inherently disordered structure.

The behavior of a random laser with coherent feedback can thus be very similar to that of a conventional laser, yet there is a significant difference between the two. A random laser does not have directional output: because the disorder-induced scattering is random in

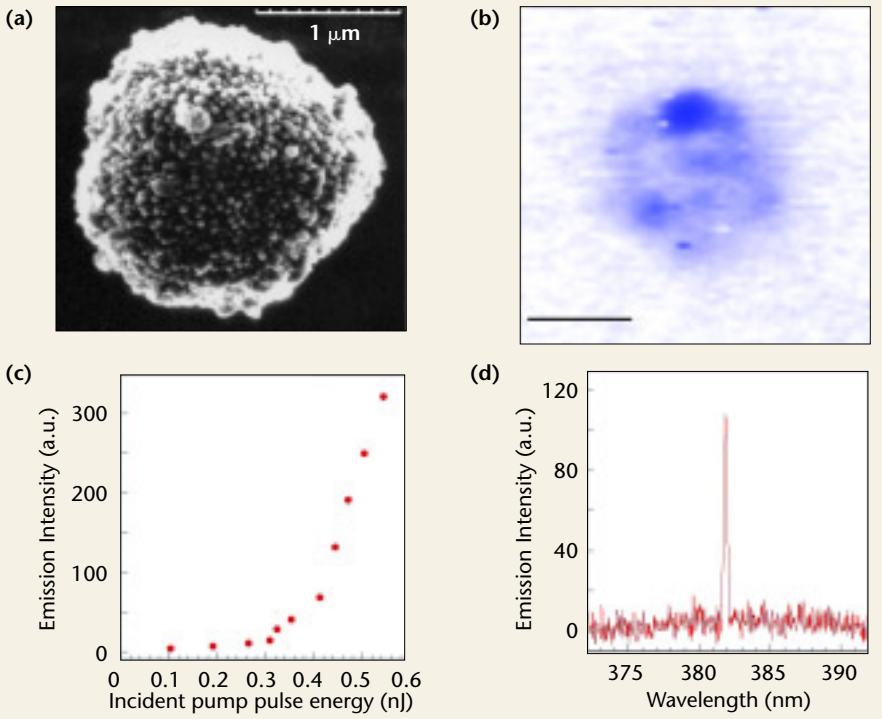


Figure 4. (a) Scanning electron micrograph of a microcluster of ZnO nanocrystallites. (b) Optical image of the emitted light distribution across the cluster. The incident pump pulse energy is 0.35 nJ. (c) Spectrally integrated emission intensity as a function of the incident pump pulse energy. (d) Spectrum of emission from the cluster at the incident pump pulse energy of 0.35 nJ.

direction, the output is multidirectional. In fact, for individual lasing modes, the directions of the output are different.

In these semiconductor nanostructures, the formation of random laser cavities is attributed to recurrent scattering and interference effects. This mechanism applies to random media with discrete scatterers and strong short-range disorder. A different type of laser cavity can be formed by smooth long-range inhomogeneity in weakly disordered media such as π-conjugated polymer or organic-dye-doped gel films. The spatial fluctuation of the film thickness results in long-range inhomogeneity of the refractive index. Light is trapped in a high-refractive-index region by total internal reflection at the boundary of this region. The typical laser cavity is quite large, i.e., on the order of hundreds of micrometers.

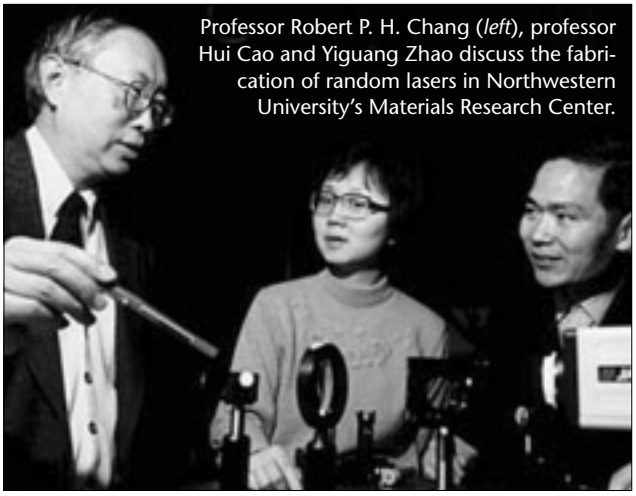
Micro-random lasers

Strong scattering can be used to produce microlasers, which are essential components of integrated photonic circuits. The key issue in microlaser fabrication is the confinement of light in a small volume, with dimensions on the order of an optical wavelength. Several types of microlasers have been developed over the past two decades. In the vertical cavity surface emitting laser, light is bounced between two distributed Bragg reflectors separated by one or two wavelengths. The microdisk laser uses total internal reflection at the edge of a high index disk to confine light in a closed path near the edge of the disk. In the 2D photonic bandgap defect mode laser, light confinement is realized through Bragg scattering in a periodic structure. The fabrication of these carefully configured microlasers requires state-of-the-art crystal growth and microfabrication facilities. In 2000, Cao et al. showed that microlasers can be made of disordered media.[3] Strong scattering in random media is another way to confine light so as to create a microlaser. This approach to fabricating a micro-random laser is easier and cheaper than any of the others: it makes it possible to create a laser even by means of a chemical reaction in a glass beaker on a hot plate.

We recently made a micro-random laser of closely packed ZnO nanoparticles. The fabrication process is very simple: we add a certain amount of zinc salt (zinc acetate dihydrate) to a solvent (diethylene glycol). When the solution is heated to 160 degrees C, ZnO nanocrystallites precipitate and coalesce into clusters measuring 1-2 μm. Millions of clusters are formed simultaneously in the solution. The size of the clusters can be controlled by varying the rate at which the solution is heated. Figure 4(a) shows a 1.7 μm cluster. It contains roughly 20,000 50-nm diameter ZnO nanocrystallites. The clusters are taken from the reaction solution and arranged at intervals on a quartz substrate.

The optical experiment is performed on a single cluster. The ZnO nanoparticles are excited by the fourth harmonic of a pulsed Nd:YAG laser (λ = 266 nm, pulsewidth = 25 ps, repetition rate = 10 Hz). The pump light is focused onto a single cluster by means of a microscope objective lens. We simultaneously measure the spectrum of emission from the cluster with a spectrometer and image the spatial distribution of the emitted light intensity across the cluster with a CCD camera. The pump light is blocked by a bandpass filter placed in front of the camera.

At low pump intensity, the emission spectrum consists of a single broad spontaneous emission peak. The spatial distribution of the spontaneous emission intensity is uniform across the cluster. When the pump intensity exceeds a certain threshold, a sharp peak emerges in the emission spectrum [Fig. 4(d)]. Simultaneously, a few bright spots appear in the image of the emitted light distribution in the cluster [Fig. 4(b)]. When the pump intensity is increased, a second sharp peak emerges in the emission spectrum and additional bright spots appear in the image of the emitted light distribution. The curve of the total emission intensity as a function of pump intensity shows a distinct change in slope at a threshold that corresponds to the onset of lasing in the micrometer-sized cluster [Fig. 4(c)]. At a pumping level significantly above the threshold, the total emission intensity increases almost linearly with the pump intensity.



Professor Robert P. H. Chang (*left*), professor Hui Cao and Yiguang Zhao discuss the fabrication of random lasers in Northwestern University's Materials Research Center.

Three-dimensional confinement of light in the micrometer-sized cluster is realized through a process of multiple scattering and wave interference. The photons spontaneously emitted inside the cluster by ZnO cannot easily get out: they bounce from one nanocrystallite to another thousands of times before finally escaping through the boundary of the cluster. During the process of multiple scattering, light is amplified by stimulated emission and may return to a position it has visited before. The interference of all the returned light, when it is constructive, can minimize light leakage out of the cluster. But constructive interference takes place only at certain frequencies determined by the spatial configuration of the scatterers. It is at these frequencies that lasing occurs. In a sample whose structure is different, the spatial arrangement of the ZnO nanocrystallites changes, as does the phase of the scattered light. Hence, the interference of the scattered light is constructive at different frequencies at which lasing occurs. This means that the lasing frequencies are the fingerprint of the unique random structure of a specific cluster. Because optical confinement is not caused by light reflection at the surface of a cluster but by scattering inside it, we can achieve lasing in clusters with irregular shapes and rough surfaces.

Applications

The light-scattering-based feedback mechanism of a random laser has applications in the fabrication of lasers in spectral regimes in which efficient reflective elements are not available, such as X-ray lasers and γ-ray lasers. Other applications are enabled by characteristics of random lasers that include low fabrication costs, sample-specific wavelength of operation, small size, flexible shape and substrate compatibility.

An example is document encoding and material labeling, where the lasing frequencies represent the "signature" of a random structure. Micro-random lasers can be used to provide optical tags in biological and medical studies. When the nanoparticle clusters are attached to biological targets, the position of the targets can be traced by detecting the lasing emission from the clusters. We can differentiate the targets because each nanoparticle cluster has its own unique set of lasing frequencies.

The multidirectional output of a random laser makes it suitable for use in displays. A thin layer of a random medium doped with emitters can, for example, be used to coat an arbitrarily shaped display panel. Random lasers can be switched on and off much faster than light-emitting diodes and can thus be used to create high speed displays. A multicolored display can be assembled by incorporating emitters of different frequencies into a single random medium. The shape, flexibility and substrate compatibility of random lasers also enable applications in areas that include assembly lines (machine vision to verify manufactured parts) and search and rescue of downed aircraft or satellites.

Most random lasers realized to date have been optically pumped. Some applications, such as flat-panel, automotive and cockpit displays, require electrical pumping. Recently, electrically pumped continuous-wave laser action was reported in rare-earth-metal-doped dielectric nanophosphors.[6] Electrical pumping is much more efficient than optical pumping, in which most of the pump light is scattered instead of being absorbed by the random medium.

In the medical arena, random lasers can be used for tumor detection and photodynamic therapy. Z. V. Vardeny's group has shown that human tissues have strong scattering and, when infiltrated with a concentrated laser dye solution, can support random lasing.[7] Since cancerous cells grow much faster than normal cells, they generate significantly more waste, in the form of fragments. The excess disorder in the malignant tissue leads to stronger scattering and more efficient lasing. One can envisage a new method of probing tumors by scanning a focused laser beam across the tissue: the laser light pumps the infiltrated dye molecules in a local region; when the pumping is not very strong, lasing can occur only at the location of the tumor, where scattering is stronger. Since lasing can occur in a region much smaller than 1 mm, it would be possible to detect a tumor in the very early stages.

Random lasers could also serve as the active elements in photonic devices and circuits. For example, a micro-random laser could function as the crucial miniature light source in a photonic crystal.[8] By infiltrating liquid crystal into macroporous glass, Wiersma and Cavalieri made a temperature-tunable random laser.[9] Such a light source exhibits a temperature-tunable color spectrum and is expected to find applications in photonics, temperature-sensitive displays and screens, and remote temperature sensing. Study of the random laser could also help us understand the galaxy masers and stellar lasers, the feedback of which is also caused by scattering.[10]

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OSA Member

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