Dynanmic Nonlinear Effect on Lasing in Random Media

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ABSTRACT

We review our recent work on lasing in active random media. Light scattering, which had been regarded detrimental to lasing action for a long time, actually provided coherent feedback for lasing. We also trapped laser light in micrometer-sized random media. The trapping was caused by disorder-induced scattering and interference. This nontraditional way of light confinement has important application to microlasers. The threshold of ranodm laser can be reduced by incorporating some degree of order into an active random medium. Our calculation result shows that by optimizing the degree of order one can dramatically reduce the threshold of random laser to the values comparable to those of photonic bandgap defect lasers.

Keywords: Random laser, optical scattering, disordered system

1. INTRODUCTION

In 1966 Letokhov et al. invented laser with nonresonant feedback using scattering reflector.[1] He further proposed a new way of generating light in a scattering medium with negative resonance.[2] In the nineteen eighties, Markushev et al. observed lasing in Nd-doped laser crystal powder.[3,4] They found a single particle, whose size is much larger than the optical wavelength, served as a laser resonator. Since then, there has been much work on powder lasers.[5–7] In early nineties, Lawandy et al. reported stimulated emission from laser dye solution containing microparticles.[8] This observation triggered many experimental[9–14]and theoretical[15–18] studies on light amplification in diffusive media. The term "random laser" appeared. It represented a laser whose feedback is due to random scattering mechanism, as opposed to the reflective feedback by the mirrors of a conventional laser. In this sense, the random laser is also called a mirrorless laser. However the feedback provided by light scattering is incoherent, i.e., the feedback is basically *intensity* feedback. Thus, the physics is not dramatically different from the amplification of spontaneous emission (ASE) without feedback. In the late nineties, random lasers with coherent feedback were realized with disordered semiconductor and organic materials.[19–21] The interference of backscattered waves produces the *amplitude* feedback.

In this paper, we review our recent experimental work on random lasers with coherent feedback. In section 2 we describe random laser with coherent feedback. Section 3 is focused on micro random laser. In Section 4 we present our numerical simulation of partically ordered random laser. Section 5 is the conclusion.

2. COHERENT RANDOM LASER

We observed lasing with coherent feedback in highly disordered semiconductor powder and polycrystalline films.[19,20] We synthesized ZnO nanoparticles with the precipitation reaction.[22] The process involves hydrolysis of zinc salt in a polyol medium. ZnO nanoparticles are cold-pressed to form a pellet of thickness $\sim$ 1 mm. The average particle size is about 70 nm. The transport mean free path was measured in the coherent back scattering experiment. ZnO has a direct band gap of 3.3 eV. To avoid absorption, the frequency-doubled output (λ = 410 nm) of a mode-locked Ti:Sapphire laser (76 MHz repetition rate, 200 fs pulse width) was used as the probe light. From the angle of the backscattering cone, we estimated $l_t \approx \lambda$.

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The ZnO powder was optically pumped by the third harmonics ($\lambda = 355$ nm) of a mode-locked Nd:YAG laser (10 Hz repetition rate, 20 ps pulse width). The pump beam was focused to a 20 μm spot on the sample surface. The emission spectrum was measured by a spectrometer with 0.13 nm spectral resolution. Simultaneously the spatial distribution of the emitted light intensity in the powder was imaged by a ultraviolet (UV) microscope onto a UV sensitive CCD camera. The amplification of the microscope was about 100 times. The spatial resolution is ~ 0.3 μm. A bandpass filter was placed in front of the microscope objective to block the pump light.

Figure 1 shows the measured emission spectra and spatial distribution of emission intensity in ZnO powder at different pump intensities. At low pump intensity, the spectrum consists of a single broad spontaneous emission peak. Its full width at half maximum (FWHM) is $\sim$ 12 nm [Fig. 1(a)]. As shown in Fig. 1(b), the spatial distribution of the spontaneous emission intensity is smooth across the excitation area. Due to the pump intensity variation over the excitation spot, the spontaneous emission in the center of the excitation area is stronger. When the pump intensity exceeds a threshold, discrete narrow peaks emerge in the emission spectrum [Fig. 1(c)]. The FWHM of these peaks is less than 0.2 nm. Simultaneously, bright tiny spots appear in the near-field image of the emitted light distribution in the powder [Fig. 1(d)]. The size of the bright spots at the sample surface is between 0.3 and 0.7 μm. When the pump intensity is increased further, additional sharp peaks emerge in the emission spectrum. Correspondingly, more bright spots appear in the image of the emitted light distribution. The frequencies of the sharp peaks depend on the sample position. As we moved the excitation spot across the sample, the frequencies of the sharp peaks changed.

Above the threshold where discrete spectral peaks appear, the emission intensity increases much more rapidly with the pump intensity. Simultaneously, the emission pulse is shortened drastically from $\sim$ 200 ps below the threshold to less than 30 ps well above the threshold.[23] Our photocounting experiment reveals that the photon number distribution in a single mode changes continuously from the Bose-Einstein distribution at the threshold to Poisson distribution well above the threshold.[24] As shown in Fig. 2, the normalized second-order correlation coefficient G_2 decreases gradually from 2 to 1 when the pump intensity increases. This result indicates photon number fluctuation in each lasing mode is quenched by gain saturation well above the threshold. These data show that lasing occurs in the ZnO powder.

We investigated the dynamics of individual lasing modes in ZnO powder.[23] Lasing oscillation in different modes starts at different time, and it lasts for different period of time. Their unsynchronized behavior suggests that these lasing modes originate from different cavities. As shown in Fig. 1(d), above the lasing threshold, the emission pattern exhibits spatially separated regions of intense laser radiation. Due to the local variation of particle density and spatial configuration, there exist small regions of stronger scattering. Light can be confined in these regions through the process of multiple scattering and interference. For a particular configuration of scatterers, only light at certain wavelengths can be confined, because the interference effect is wavelength sensitive. In a different part of the sample, the configuration of the scatterers is different, thus light at different wavelengths is confined. In other words, there are many small resonant cavities in the powder. In a finite-sized sample, the trapping of light is incomplete because photons can escape through the boundaries. This gives rise to the cavity loss. When the optical gain reaches the loss, lasing occurs in these cavities.

Unlike the conventional lasers with directional output, laser emission from the random media could be observed in all directions. The laser emission spectra vary with the observation angle. Since different random cavities have different output directions, lasing modes observed at different angles are different.

3. MICRO RANDOM LASER

The results of last section illustrate that the disorder-induced optical scattering not only provides coherent feedback for lasing, but also leads to spatial confinement of laser light in micrometer-sized volume. Utilizing this new mechanism of optical confinement, we fabricated microlasers with disordered media.[25]

To fabricate micrometer-sized random medium, we agglomerated ZnO nanoparticles to form clusters whose size varies from half a micron to a few micron.[26] The inset of Fig. 3 is the scanning electron microscope (SEM) image of a typical ZnO cluster. The size of the cluster is about 1.7 μm. It contains roughly 20000 ZnO nanocrystallites. The ZnO cluster is optically pumped by the third harmonics of a pulsed Nd:YAG laser. The

pump light is focused by a microscope objective onto a single cluster. We simultaneously measured the spectrum of emission from the cluster and imaged the spatial distribution of the emitted light intensity in the cluster.

At low pump intensity, the emission spectrum consists of a single broad spontaneous emission speak [Fig. 4(a)]. Its FWHM is 12 nm. The spatial distribution of the spontaneous emission intensity is uniform across the cluster [Fig. 4(b)]. When the pump intensity exceeds a threshold, a sharp peak emerges in the emission spectrum [Fig. 4(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 [Fig. 4(d)]. When the pump intensity is increased further, a second sharp peak emerges in the emission spectrum [Fig. 4(e)]. Correspondingly, additional bright spots appear in the image of the emitted light distribution [Fig. 4(f)].

As shown in Fig. 3, above the pump intensity at which sharp spectral peaks and bright spots appear, the emission intensity increases almost linearly with the pump intensity. These data suggest that lasing occurs in the micrometer-sized cluster. The incident pump pulse energy at the lasing threshold is $\sim$ 0.3 nJ. Note that less than 1 % of the incident pump light is absorbed. The rest is scattered.

Since the cluster is very small, optical reflection from the boundary of the cluster might have some contribution to light confinement in the cluster. However, the laser cavity is not formed by total internal reflection at the boundary. Otherwise the spatial pattern of laser light would be a bright ring near the edge of the cluster.[27] We believe the 3D optical confinement in a micrometer-sized ZnO cluster is achieved through disorder-induced scattering and interference. Since interference effect is wavelength sensitive, only light at certain wavelengths can be confined in the cluster. In another cluster of different particle configuration, light at different wavelengths is confined. Hence, the lasing frequencies are fingerprints of the clusters. Because optical confinement is not caused by light reflection at the surface of a cluster, but by scattering inside the cluster, we can achieve lasing in clusters with irregular shapes and rough surfaces.

We would like to compare the micro random laser with other types of microlasers. Over the past decade, several types of microlasers have been developed. The key issue for microlaser is to confine light in a small volume with dimensions on the order of optical wavelength. In the vertical cavity surface emitting laser, light is confined by two distributed Bragg reflectors.[28] The microdisk laser utilizes total internal reflection at the edge of a high index disk to form whispering gallery modes.[29] In the two-dimensional photonic band-gap defect mode laser, lateral confinement of light is realized through Bragg scattering in a two-dimensional periodic structure.[30] The fabrication of these microlasers requires expensive crystal growth and nanofabrication facilities. In micro random laser, the optical confinement is achieved through disorder-induced scattering and interference. The fabrication of the micro random laser is much easier and cheaper than that of most microlasers.

4. DYNAMIC NONLINEAR EFFECT

Following the first demonstration of coherent random laser, there have been many experimental[33–38] and theoretical[39–46] studies of this nontraditional laser. Recent studies illustrate that the random laser modes are the long-lived eigenmodes of disordered media.[41–43,46,47] Despite the modes with long lifetime are preferably amplified, their wavefunctions are not modified by the presence of gain. However, nonlinearity can change the eigenmodes of a disordered system. In a random laser nonlinear effect is significant because the nonlinear coefficient is resonantly enhanced at the lasing frequency and the laser intensity is high due to spatial confinement in random media. We studied both experimentally and theoretically the effect of Kerr nonlinearity on lasing in disordered media.

We used two types of random media in our experiments. One is poly(methyl methacrylate) (PMMA) sheets containing rhodamine 640 (Rh640) perchlorate dye and titanium dioxide (TiO_2) microparticles, the other is closely-packed ZnO nanoparticles with average diameter 100 nm. The dye concentration in PMMA is 10 mM. The TiO_2 particles have an average diameter of 0.4 μm. The density of TiO_2 particles is $\sim 1.4 \times 10^{12}$ cm^{-3}. The PMMA sheet is optically excited by the second harmonics of a mode-locked Nd:YAG laser. The pump spot is $\sim$ 100 μm in diameter at the sample surface. When pump intensity exceeds the lasing threshold, discrete lasing modes emerge in the emission spectrum. As the pump intensity increases, the gain spectrum of dye molecules shifts towards longer wavelength. This red shift is caused by excited state absorption,[48] and has been confirmed experimentally in neat dye solution.[49] As shown in Fig. 5, additional lasing modes appear on the long

wavelength side, while some lasing modes on the short wavelength side disappear. However, individual lasing modes shift towards shorter wavelength, as marked in Fig. 1. Typically optical gain pulls the frequencies of lasing modes towards the peak of gain spectrum (frequency pulling effect). In our experiment, the lasing modes move in the opposite direction as the gain spectrum. This suggests the spectral shift of lasing modes is caused by nonlinearity. Nonlinear Kerr effect has been observed in Rh640 solution containing TiO_2 particles.[50] The refractive index n changes linearly with light intensity I: $n = n_0 + n_2 I$, where n_0 is the linear refractive index, n_2 is the nonlinear Kerr coefficient. The nonlinearity is contributed mainly by dye molecules. The value of n_2 in our samples is in the range -10^{-14} to -10^{-15} m^2/W. Above the lasing threshold, the excitation intensity is on the order of 10^{11} W/m^2. Thus the change of refractive index Δn ranges from -10^{-3} to -10^{-4}. The negative sign of n_2 explains the decrease of lasing wavelengths as the pump intensity increases. The wavelength shift is in the range 0.1 – 1.0 nm, which is on the same order of the experimental shift.

In addition to time-integrated spectra, we measured time-resolved lasing spectra to track spectral shift of lasing modes in time. Figure 6 shows a spectral-temporal image of laser emission from closely-packed ZnO powder. The ZnO powder is optically excited by 20 ps pulses from a frequency-tripled mode-locked Nd:YAG laser. The pump beam is focused to a 20 μm spot on the sample surface. Laser emission from ZnO powder is dispersed by a 0.3-meter monochromator, then directed into a Hamamatsu streak camera. Individual lasing modes are separated by the monochromator, and their temporal evolutions are recorded by the streak camera. The lasing modes at shorter wavelength are red-shifted with time, while the lasing modes at longer wavelength are blue-shifted with time. The modes in the center do not shift in wavelength. For example, the mode labeled A shifts from 387.76 nm at t = 40 ps to 387.96 nm at t = 130 ps. The mode labeled B shifts from 390.69 nm at t= 40 ps to 390.58 nm at t= 105 ps. The wavelength of mode C remains nearly constant in time. Temporal shift of lasing frequencies must result from dynamic change of the refractive index of ZnO. It is known that ZnO has a large third-order nonlinearity near its band edge.[51,52] The values of its third-order nonlinear coefficient $\chi^{(3)}$ range from 10^{-16} to 10^{-14} m^2/V^2 at room temperature. The nonlinear response of ZnO is also fast: its nonlinear response time is measured to be 160 fs near band edge.[53]

To understand the nonlinear effect on random lasing, we performed numerical simulation of a model system. The algorithm is based on the finite-difference time-domain (FDTD) solution to Maxwell equations coupled with the rate equations of electronic populations. The third-order nonlinearity with finite response time is included. Our numerical results demonstrate that nonlinearity not only modifies the frequency and size of the eigenmodes of a disordered system, but also changes laser emission intensity and laser pulse width of a random laser. The speed of nonlinear response determines how nonlinearity affects random lasing process. We find two regimes depending on the relative values of two time scales, one is the nonlinear response time, the other is the lifetime of the lasing state. For slow nonlinear response, collective scattering of many particles determines the buildup of lasing modes. Nonlinearity changes the laser output through modification of the spatial size of the lasing state. But when the nonlinear response is faster than the buildup of the lasing mode, the lasing mode cannot response fast enough to the nonlinear refractive index change. Rapid change of the phase of the scattered light undermines the interference effect of many particle scattering. Instead, the nonlinear effect of single particle scattering becomes dominant. Strong nonlinearity could leads to instability.

5. CONCLUSION

We reviewed our experimental studies of coherent random lasers. For a long time light scattering had been regarded detrimental to lasing action. We showed that strong scattering can facilitate lasing by forming cavities and supply coherent feedback. Our experimental results demonstrated that coherent light can be generated in a disordered medium with gain. We illustrated the similarity and difference between random laser and conventional laser.

Disorder-induced scattering not only provides coherent feedback for lasing, but also leads to spatial confinement of laser light in micrometer-sized volume. Utilizing this new mechanism of optical confinement, we fabricated microlasers with disordered media.

We studied both experimentally and numerically the dynamic effect of nonlinearity on lasing in disordered medium. The third-order nonlinearity not only changes the frequency and size of lasing modes, but also modifies

the laser emission intensity and laser pulse width. When the nonlinear response time is comparable to or longer than the lifetime of the lasing mode, the nonlinearity changes the laser output through modifying the size of the lasing mode. When the nonlinear response is much faster than the buildup of the lasing mode, positive nonlinearity always extracts more laser emission from the random medium due to the enhancement of single particle scattering.

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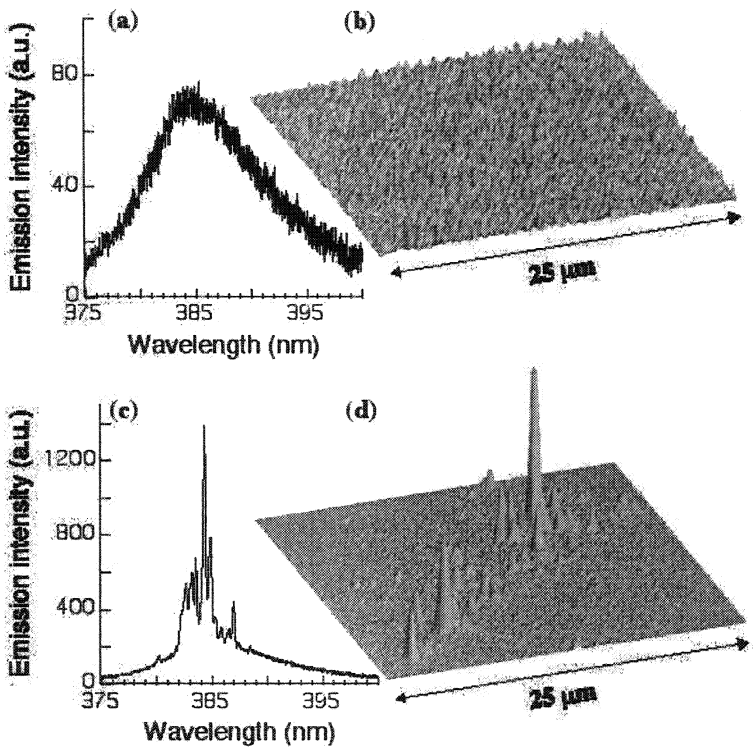

Figure 1. (a) and (c) are the measured spectra of emission from the ZnO powder. (b) and (d) are the measured spatial distribution of emission intensity in the powder. The incident pump pulse energy is 5.2 nJ for (a) and (c), and 12.5 nJ for (b) and (d).

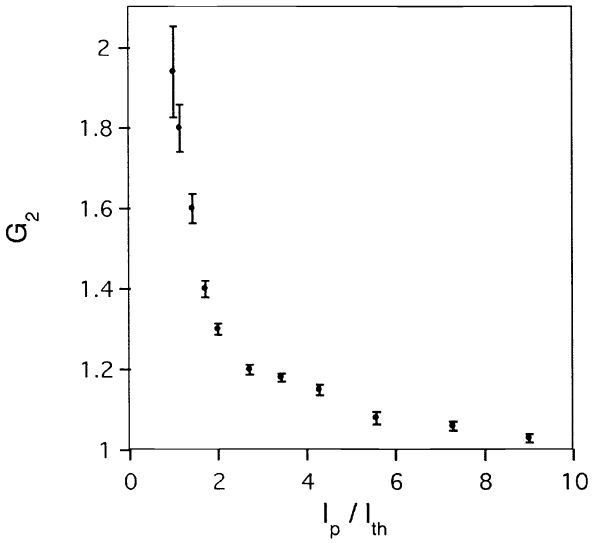

Figure 2. Normalized second-order correlation coefficient G_2 as a function of the ratio of the incident pump intensity I_p to the threshold intensity I_{th}.

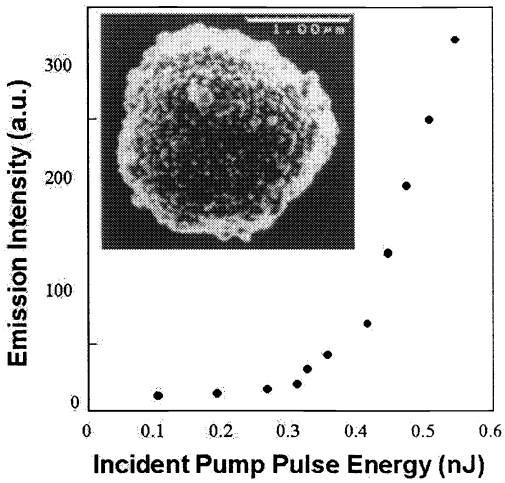

Figure 3. Spectrally integrated intensity of emission from the ZnO cluster versus the incident pump pulse energy. The inset is the SEM image of the ZnO cluster.

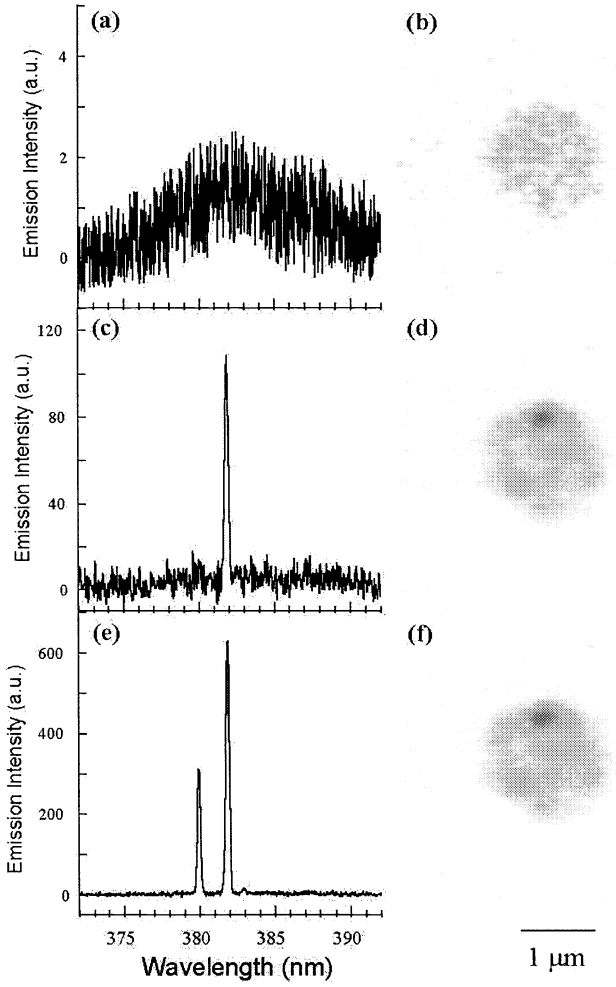

Figure 4. (a), (c), and (e) are the spectra of emission from the ZnO cluster shown in Fig. 3. (b), (d), and (f) are the corresponding spatial distribution of emission intensity in the cluster. The incident pump pulse energy is 0.26 nJ for (a) and (b), 0.35 nJ for (c) and (d), and 0.50 nJ for (e) and (f).

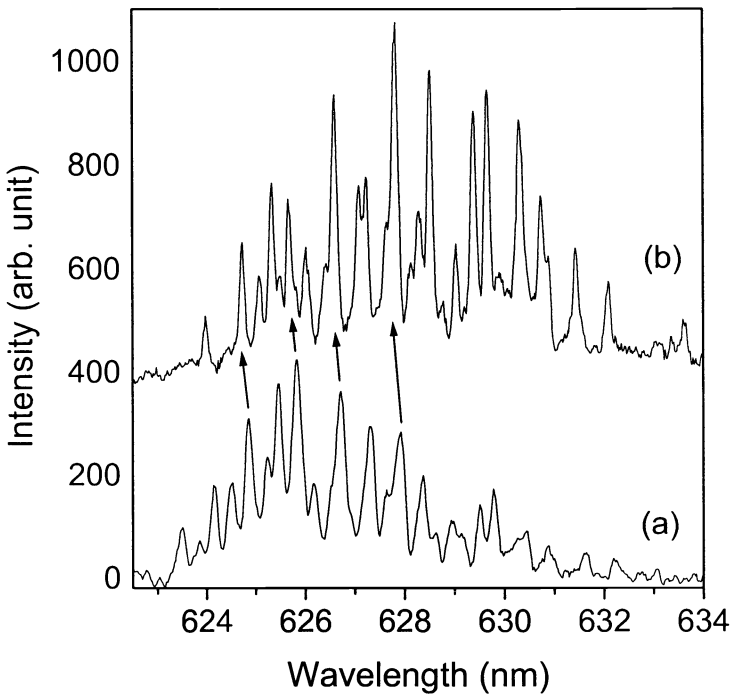

Figure 5. Time-integrated spectra of laser emission from a PMMA sheet with dye and microparticles. The incident pump intensities are 2.0×10^{12} W/m^2 (a), and 4.0×10^{12} W/m^2 (b). The top spectrum is shifted vertically for clarity.

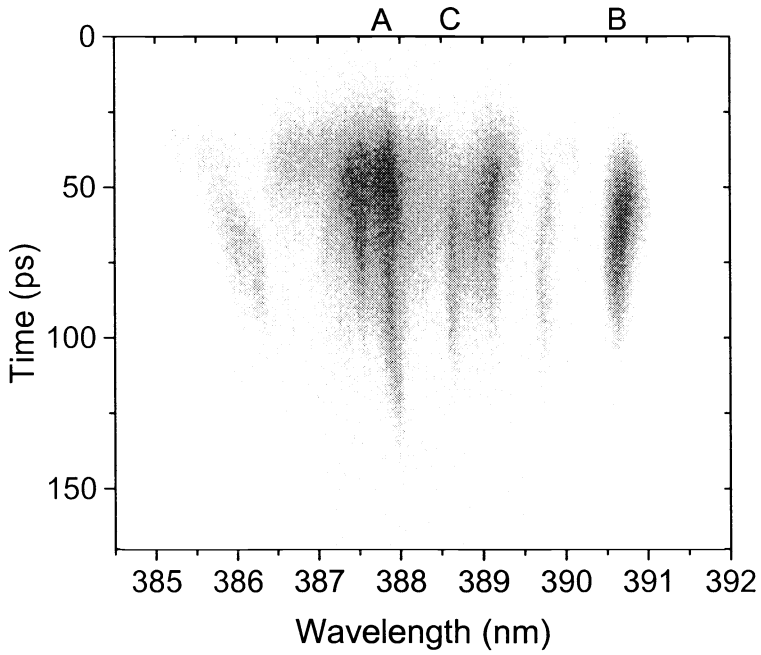

Figure 6. A spectral-temporal image of laser emission from ZnO powder.