Thermal effects in quasi-continuous-wave $Nd^{3+}:Y_3Al_5O_{12}$ nanocrystalline-powder random laser

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(Received 14 October 2003; accepted 19 December 2003)

We report an experimental investigation on the thermal effects in a $Nd^{3+}:Y_3Al_5O_{12}$ nanocrystalline-powder random laser with a one-mirror structure by quasi-continuous-wave laser diode pumping. Extremely low thermal conductivity in powder and significant temperature dependence of the narrow emission spectrum of $Nd^{3+}:Y_3Al_5O_{12}$ leads to a laser line redshift and gain reduction with a higher heat deposit. Mode drifting induced by the temperature dependence of the refractive index is also discussed. © *2004 American Institute of Physics.*
[DOI: 10.1063/1.1647285]

Random lasers have received much attention in recent years.[1–8] Unlike in conventional lasers, necessary feedback is from strong scattering inside the lasing material. There are two kinds of essentially different phenomena referred to as random lasers: Amplified spontaneous emission and true lasing with coherent feedback.[7] The former had been predicted theoretically more than 30 years ago by Letokhov,[3] where the transport of light intensity can be described by a diffusion formalism. When scattering is stronger and the interference effect cannot be neglected, the system goes toward the regime of photon localization[9,10] and random lasing with coherent feedback is possible. Cao *et al.*[6] demonstrated this experimentally on ZnO powder in 1999. While many fundamental questions remain, possible applications had been proposed, such as improved phosphor, planar display,[11] sensor,[2,12] etc.

Recently, we demonstrated quasi-continuous-wave (QCW) random lasing in $Nd^{3+}:Y_3Al_5O_{12}$ (Nd^{3+}:YAG) nanocrystalline powder with a one-mirror structure by laser diode pumping.[13] The one-mirror structure had been proposed to reduce the laser threshold.[5] In this letter, we report an experimental investigation of the thermal effect in random lasers. Because of using disordered materials as lasing media, heat conduction in random lasers is inevitably low. In a QCW random laser we demonstrated, it was found that the thermal problem has big effect on random laser properties. Up until now, almost all experimental studies are pulsed pump, so the heat deposit is low. Rand *et al.*[11,14] investigated a continuous-wave random laser by electron pump, but they have not discussed the thermal effect. Wiersma *et al.*[2] demonstrated a temperature tunable random laser, which is also different from the present study. In their study,[2] temperature is a measure of active control over a random laser.

In experiments, a Hamamatsu Photonics K. K. (Shizuoka, Japan) QCW laser diode array was used as the pump source, which has four bars with a center wavelength at ~805 nm, and an adjustable pulse duration and repetition rate. The pulse is rectangular with a rising and dropping edge less than 5 μs. The fast axes of the laser output was collimated by a cylindrical lens, and focused by a spherical lens to a spot of about 1 mm diameter. The spot is an image of the laser diode array, so it must contain fine structures. An estimation of pump intensity is not straightforward. The coupling efficiency was found to be only 40% for not optimized optics, and maximum peak power irradiated at sample was 110 W. The sample consists of a 4%-doped Nd^{3+}:YAG powder tablet with a dimension of $\phi 16\times 3\,mm^3$ and a mirror with high reflective coating at 1064 nm (measured to be ~99.5%). The coated face is at the powder side. The powder was provided by Konoshima Chemical Co., Ltd (Osaka, Japan). A scanning electron microscope image showed that the powders have an average diameter of about 250 nm. The dielectric volume fraction was found to be about 50% by comparing the mass density of the power tablet and bulk Nd^{3+}:YAG. The emission from the sample was collected by the same spherical lens of focusing the pump light, separated from the pump light by a dichromatic mirror and an interference filter at 1064 nm, and then focused to detectors. When measuring temporal behavior and intensity, an InGaAs photodiode was used. When measuring emission spectra, a monomode fiber was used to couple some light into an Ando electric Co., Ltd (Kanagawa, Japan) AQ-6315A Optical Spectrum Analyzer.

The general observations as proof of random lasing were reported previously.[13] Threshold behavior and substantial spectral narrowing were observed. Besides, spiking behavior was observed when the pump power was higher than a second threshold point. When pump power increases, spiking becomes stronger (larger amplitude and higher repetition rate). With a ''peak hold'' method (for every spectral point, the intensity is monitored for a period of time and the maximum value is recorded), the emission spectrum was found to be very narrow <0.1 nm (limited by instrument resolution 0.05 nm) just above the second threshold, but broadened when the pump power was even higher. This was explained by the appearance of multiple laser lines but irresolvable in the measurement.

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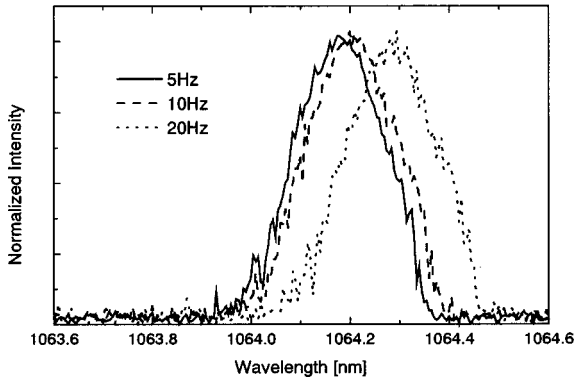

FIG. 1. Emission spectra with different repetition rate 5, 10, and 20 Hz, but same duration 200 μs and pump power 100 W, which is higher than the second threshold 77 W.

Here, experimental observations relating to thermal effects are presented. In Fig. 1, emission spectra with different repetition rates 5, 10, and 20 Hz, but the same duration 200 μs and pump power 100 W, which is higher than the second threshold 77 W, are plotted. One can see that the emission is redshifted with a higher repetition rate. The sharp spikes in the spectra should be viewed as an artifact. It is due to the peak hold method we used. The artificial spikes result from pulse-to-pulse fluctuation.

In Fig. 2, emission wave forms with the same pump power 100 W and repetition rate 50 Hz but different durations, 200, 150, and 100 μs are plotted. As seen in Fig. 2, at the first phase of pulse where emission is fluorescence and intensity grows linearly, there is no difference between different wave forms. This reflects the same pump rate for different pump pulses. At the second phase, where emission grows nonlinearly, that of the shorter pump pulse increases faster. This suggests that the gain is higher with a shorter pump pulse. At the third phase of lasing, stronger pulsing was observed with shorter duration. According to the previous study,[13] stronger pulsing corresponds to a higher gain too.

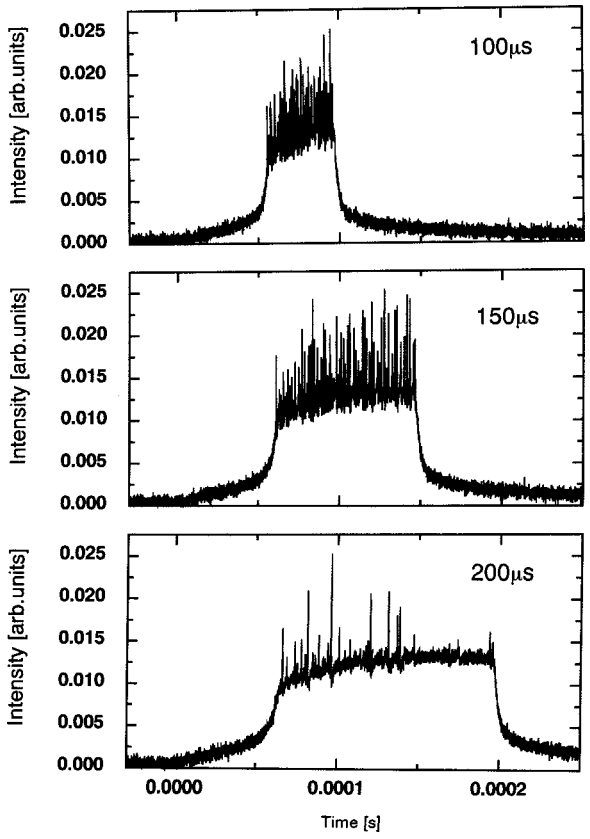

FIG. 2. Emission wave forms with the same pump power 100 W and repetition rate 50 Hz but different duration 200, 150, and 100 μs. Pulse is rectangular.

All of these findings can be attributed to heating inside the powder. The absorbed pump power is partly converted to light by radiative transitions, but the rest becomes heat deposited because of nonradiative relaxations. For the low thermal conductivity in the powder sample, the temperature rise could be significant in high repetition rate pumping. One can get information on the temperature rise inside the powder by monitoring the emission spectrum. In the following, a qualitative analysis on the effect of heating is presented.

Near room temperature, the ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition (1064 nm) of Nd^{3+}:YAG is homogeneously broadened with a Lorentzian line shape,[15]

$$g = \frac{\frac{\Delta \nu_H}{2\pi}}{(\nu - \nu_0)^2 + \left(\frac{\Delta \nu_H}{2}\right)^2}, \tag{1}$$

where $\Delta \nu_H$, ν_0, and ν are the full width at half maximum linewidth, center frequency, and frequency, respectively. Due to coupling between the Nd^{3+} ion and lattice vibration, the center frequency and linewidth will change with temperature. At 300 K, $\Delta \nu_H \approx 0.45$ nm, and $\nu_0 \approx 1064.15$ nm.[16,17] According to the data in literature,[15,16] near room temperature (300 K), $d\lambda_0/dT \approx 0.004$ nm/K, and $d\Delta\lambda_H/dT \approx 0.0045$ nm/K.

It should be noted that the above description of the temperature dependence of the emission spectrum is a simplified version. For real systems, the thermal population of an upper Stark level may change the emission spectrum. Besides, if there are two or more transitions with nearly equal frequencies, as in the case of Nd^{3+}:YAG transitions at 1064 nm,[16] the evolution of the emission spectrum with temperature will become more complicated. But for a clear presentation of the effect, such a simplified model is enough. In addition, we directly use the emission spectrum as the gain spectrum.

So, for the temperature distribution inside the pumped powder, the gain spectra at different position are different. To get the effective gain coefficient, one needs to know all of the information concerning pump and temperature distribution, which is impossible. One may assume a uniform distribution of excited ions with temperature for a qualitative study. Thus, the effective gain coefficient is written as

$$g_e \propto \frac{\int g(T)dT}{\Delta T}, \tag{2}$$

where ΔT is the temperature variation span, which is larger when the heat deposit power is higher, and the integration is over the whole temperature range. Figure 3 shows numerical results of the gain spectra for several ΔT values (the lower limit of temperature is 300 K). The effective gain spectrum is broadened, the center wavelength is redshifted, and the maximum gain decreases. Therefore, the observations shown

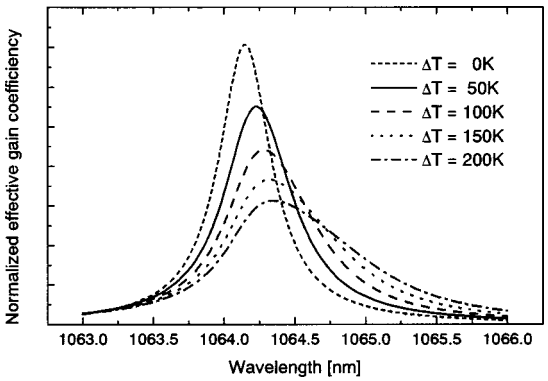

FIG. 3. Numerical results of gain spectra for several temperature span values ΔT.

in Figs. 1 and 2 can be explained. When fitting the experimental spectral data with this simple model, the average temperature rise ($\Delta T/2$) inside the pumped volume is calculated to be 8, 18, and 57 K for a pump repetition rate of 5, 10, and 20 Hz, respectively.

This kind of thermal effect is significant in a Nd^{3+}:YAG random laser, because the gain spectrum is narrow and temperature dependence is relatively high. In systems with broad gain spectra, such as ZnO powder film, dye solution with TiO_2 scatterers, etc, the effect is negligible.

To evaluate thermal processes in the sample, one at least needs to know the thermal conductivity in the powder and how much power was absorbed. But both are difficult to obtain. We tried to calculate the thermal conductivity by models described in literature,[18] but the results were different from model to model. However, with the aid of those calculations, it was concluded that the thermal conductivity in the powder may be taken as one-tenth of that of YAG crystal ($0.11\ \mathrm{W\,cm^{-1}\,K^{-1}}$), which will not be too far from the real value and good for a rough estimation. The thermal time constant can be written as $\tau = D^2/\alpha$ where $D = 0.1$ cm is the diameter of heated spot, $\alpha = \lambda/(X\rho C)$ is diffusivity, $\lambda = 0.01\ \mathrm{W\,cm^{-1}\,K^{-1}}$ and $X = 0.5$ are the thermal conductivity and volume fraction of the powder, $\rho = 4.56\ \mathrm{g\,cm^{-3}}$ and $C = 0.59\ \mathrm{W\,s\,g^{-1}\,K^{-1}}$ are the density and specific heat of a YAG crystal, respectively.[17] Then, τ is calculated to be about 1.4 s.

Because the thermal time constant is much longer than the duration of one pulse, the temperature rise by one pump pulse can be approximated by $\delta T = E_h/(X\rho VC)$, where E_k is the deposited energy and V is the heated volume. E_k depends on how much light is absorbed and the fraction of absorbed energy finally converted to heat. We assume E_k equals 10% of the pump energy, and effective heated volume V is $0.1 \times 0.1 \times 0.04\ \mathrm{cm^3}$. Then, δT is calculated to be about 3.7 K. By taking into account the heat accumulation, the consistence with temperature rise deduced by above model of emission shift is plausible.

In the following, we discuss the thermal effect on the mode structure of a Nd^{3+}:YAG random medium. By using appropriate pump power level, just above the second threshold, wave forms with sparse pulses can be obtained. The sparse pulse train is not stable, but can be traced in proceeding triggers. We observed a slow evolution of the pulse train with a time scale larger than 1 s, which is believed to be due to the thermal effect.

A slight modification of dielectric index can result in a significant change of mode structure, both mode frequency and spatial distribution. In terms of ray tracing, the modes correspond to closed light paths. Intuitively, if the resulting variation in light path length is comparable with a half wavelength, the change of mode structure is large. For a YAG crystal, the temperature dependence of refractive index at room temperature is $dn/dT = 7.3 \times 10^{-6}\ \mathrm{K^{-1}}$,[17] which is small. However, in our experiments, lasing modes correspond to light paths which contribute to the center region of a coherent backscattering cone, which are known to be due to a large number of scattering events N, $>10^4$ for example.[19] Taking into account the average diameter of crystalline particles $d = 250$ nm and assuming a mean temperature rise of $\Delta T = 10$ K and number of scattering events $N = 10^4$, one may estimate the change in light path length as $\Delta L = dn/dT \cdot \Delta T \cdot N \cdot d \approx 0.18\ \mu$m, which is comparable with half of the laser wavelength of 0.53 μm. Therefore, temperature evolution-induced mode structure change is large in the present experimental system.

In summary, thermal effects in a QCW Nd^{3+}:YAG nanocrystalline-powder random laser are investigated. The thermal effects are due to temperature dependence of the gain spectrum and index of Nd^{3+}:YAG nanocrystalline-powder. The former leads to a redshift of the emission line and reduction of effective gain, which is significant only for systems with a narrow gain spectrum. The latter leads to mode drifting with temperature evolution of the disordered medium, which should be general in QCW or continuous-wave random lasers.

This work was supported by the 21st Century COE program of Ministry of Education, Science, and Culture of Japan.

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