Measurement of the gain volume in amplifying and scattering media by a two-beam spatial-correlation method
Light propagation in a multiple-scattering medium has been attracting increasing attention. When the medium has gain, luminescence in the medium experiences strong amplification by stimulated emission. This effect gives rise to the spectral collapse and random or diffusive laser action in the medium. Such an amplifying process was first discussed by Letokhov as an analogy of a nuclear reactor [1]. Lawandy et al. reported the observation of a spectral collapse and shortening of the lifetime in the luminescence from colloidal suspensions containing rhodamine 640 perchlorate dye in methanol and TiO$_2$ particles, which resembled multimode laser behavior [2]. Random laser action depends on various factors [3–6]. When the mean-free-path becomes of the order of the wavelength, the luminescence light would coherently be feedbacked by the recurrent scattering processes [7,8]. Recently, Cao et al. reported discrete and angular dependent lasing modes in semiconductor powder and argued the possibility of coherent random laser [9].

It has been suggested that absorption saturation in the excitation process and the resultant increment in the gain volume is necessary to realize sufficient gain-path length for the random laser action [10]. The presence of the absorption saturation was experimentally well confirmed by the nonlinear reflectivity [11] and the excitation power dependent spectral redshifts and blueshifts in a strongly scattering medium [12]. The size of the gain volume could be examined from the excitation beam size dependence of the threshold energy of the spectral collapse [13,14]. It was found that there is a critical mean-free-path below which the threshold energy is almost independent of the mean-free-path for every excitation beam size. This behavior suggests that the diffusion volume of the luminescence light obtained with the critical mean-free-path is just the same order of the gain volume in the medium [13].

In this paper, we present experiments on the size of the gain volume in amplifying and scattering media using a spatial correlation method, which gives direct information on the size of the gain volume. In this method, two excitation light pulses at the threshold energy of the spectral collapse are injected into the sample and the spectral width of the luminescence is measured as a function of the spatial separation between the first and second beams. When two beams are spatially overlapped within the gain volume, two beams work cooperatively to result in the strong spectral collapse. The spectral width as a function of the beam separation reflects the size of the gain volume in an amplifying and scattering medium. It was found that $L_{th}$ is proportional to $L_a$ for different samples of the different transport mean-free-paths and the different absorption lengths. Numerical simulation based on the coupled rate and diffusion equations well recapitulates the experimental results, which are discussed in terms of the stimulated emission cross section $\sigma_s$, and the absorption cross section $\sigma_a$, of dye molecules.

Figure 1 shows the schematic diagram of experimental setup, which is based on the Michelson interferometer. The excitation light source was the second harmonics of a injection seeded Q-switched Nd$^{3+}$ yttrium-aluminum-garnet (YAG) laser. The wavelength was 532 nm, the pulse width was 8 ns, and the repetition rate was 5 Hz. The excitation pulse energy was changed from 0.0007 mJ to 0.56 mJ. The incident light beam was divided into two beams by a beam splitter (BS), and focused on the sample with the focusing lens L3. The beam diameter $w$, was 80 $\mu$m. The lens, L1 and L2, were inserted in order to diverge the incident beam so that the two excitation beams make a separation distance $d$, at the focusing point of L3. One of the corner cube prisms CP1, was translated in order to change the beam separation $d$, at the focusing point of L3 on the surface of sample cell S. Two laser beams, B1 and B2, were orthogonally polarized by a reflection mirror.

![FIG. 1. Experimental setup. L1, L2, L3 and L4 are lenses; CP1 and CP2 are corner cube prisms; BS is the beam splitter; SM is the spectrometer; S is the sample cell. B1 and B2 are the first and second beams.](Image)
and $10^{-2}$ M, $(\triangle) 10^{-2}$ M with the volume fractions of polystyrene spheres 2%, 5%, and 10%, respectively. The solid, dashed, and dotted lines are corresponding calculated curves.

$\lambda/2$ wave plate in order to avoid the interference effect between two beams. The sample was methanol solution of kitson red with colloidal suspensions of polystyrene spheres. The concentration of dye was varied from $1 \times 10^{-4}$ to $10 \times 10^{-4}$ M, which corresponds to absorption length, $l_a = 980$ to $98 \mu m$. The diameter of polystyrene spheres was $0.45 \mu m$ (Sekisui Chemical Co.) [15]. The volume fraction of polystyrene spheres was changed from 2% to 10%. These volume fractions correspond to the transport mean-free-path of polystyrene spheres was $2\%$, $5\%$, and $10\%$, respectively. The solid, dashed, and dotted lines are corresponding calculated curves.

The concentration of dye was varied from $1 \times 10^{-4}$ to $10 \times 10^{-4}$ M, which corresponds to absorption length, $l_a = 980$ to $98 \mu m$. The diameter of polystyrene spheres was $0.45 \mu m$ (Sekisui Chemical Co.) [15]. The volume fraction of polystyrene spheres was changed from 2% to 10%. These volume fractions correspond to the transport mean-free-path of polystyrene spheres 2%, 5%, and 10%, respectively. The solid, dashed, and dotted lines are corresponding calculated curves.

The concentration of dye was varied from $1 \times 10^{-4}$ to $10 \times 10^{-4}$ M, which corresponds to absorption length, $l_a = 980$ to $98 \mu m$. The diameter of polystyrene spheres was $0.45 \mu m$ (Sekisui Chemical Co.) [15]. The volume fraction of polystyrene spheres was changed from 2% to 10%. These volume fractions correspond to the transport mean-free-path of polystyrene spheres was $2\%$, $5\%$, and $10\%$, respectively. The solid, dashed, and dotted lines are corresponding calculated curves.

The concentration of dye was varied from $1 \times 10^{-4}$ to $10 \times 10^{-4}$ M, which corresponds to absorption length, $l_a = 980$ to $98 \mu m$. The diameter of polystyrene spheres was $0.45 \mu m$ (Sekisui Chemical Co.) [15]. The volume fraction of polystyrene spheres was changed from 2% to 10%. These volume fractions correspond to the transport mean-free-path of polystyrene spheres was $2\%$, $5\%$, and $10\%$, respectively. The solid, dashed, and dotted lines are corresponding calculated curves.

The concentration of dye was varied from $1 \times 10^{-4}$ to $10 \times 10^{-4}$ M, which corresponds to absorption length, $l_a = 980$ to $98 \mu m$. The diameter of polystyrene spheres was $0.45 \mu m$ (Sekisui Chemical Co.) [15]. The volume fraction of polystyrene spheres was changed from 2% to 10%. These volume fractions correspond to the transport mean-free-path of polystyrene spheres was $2\%$, $5\%$, and $10\%$, respectively. The solid, dashed, and dotted lines are corresponding calculated curves.

The concentration of dye was varied from $1 \times 10^{-4}$ to $10 \times 10^{-4}$ M, which corresponds to absorption length, $l_a = 980$ to $98 \mu m$. The diameter of polystyrene spheres was $0.45 \mu m$ (Sekisui Chemical Co.) [15]. The volume fraction of polystyrene spheres was changed from 2% to 10%. These volume fractions correspond to the transport mean-free-path of polystyrene spheres was $2\%$, $5\%$, and $10\%$, respectively. The solid, dashed, and dotted lines are corresponding calculated curves.

The concentration of dye was varied from $1 \times 10^{-4}$ to $10 \times 10^{-4}$ M, which corresponds to absorption length, $l_a = 980$ to $98 \mu m$. The diameter of polystyrene spheres was $0.45 \mu m$ (Sekisui Chemical Co.) [15]. The volume fraction of polystyrene spheres was changed from 2% to 10%. These volume fractions correspond to the transport mean-free-path of polystyrene spheres was $2\%$, $5\%$, and $10\%$, respectively. The solid, dashed, and dotted lines are corresponding calculated curves.

The concentration of dye was varied from $1 \times 10^{-4}$ to $10 \times 10^{-4}$ M, which corresponds to absorption length, $l_a = 980$ to $98 \mu m$. The diameter of polystyrene spheres was $0.45 \mu m$ (Sekisui Chemical Co.) [15]. The volume fraction of polystyrene spheres was changed from 2% to 10%. These volume fractions correspond to the transport mean-free-path of polystyrene spheres was $2\%$, $5\%$, and $10\%$, respectively. The solid, dashed, and dotted lines are corresponding calculated curves.

The concentration of dye was varied from $1 \times 10^{-4}$ to $10 \times 10^{-4}$ M, which corresponds to absorption length, $l_a = 980$ to $98 \mu m$. The diameter of polystyrene spheres was $0.45 \mu m$ (Sekisui Chemical Co.) [15]. The volume fraction of polystyrene spheres was changed from 2% to 10%. These volume fractions correspond to the transport mean-free-path of polystyrene spheres was $2\%$, $5\%$, and $10\%$, respectively. The solid, dashed, and dotted lines are corresponding calculated curves.
To explain the experimental results, we performed numerical simulations. The energy states for dyes are the ground state $S_0$ and excited state $S_1$, which consist of many vibrational sublevels. Since the nonradiative decay rates within $S_0$ and $S_1$ manifolds are very fast, we assume that the population of $S_0$ and $S_1$ are in their lowest sublevels. Basic equations used are rate equations for the population in dye molecules

$$\frac{dN_i(r,t)}{dt} = \sigma_s(\lambda_e)N_0(r,t)c\Phi_e(r,t) - \int\sigma_s(\lambda_e)N_i(r,t)c\Phi_i(\lambda_1,r,t)d\lambda_1 - \frac{N_i(r,t)}{\tau},$$

and the diffusion equation for the excitation light density $\Phi_e(r,t)$,

$$\frac{d\Phi_e(r,t)}{dt} = \frac{cI^*}{3}\nabla^2\Phi_e(r,t) - \sigma_s(\lambda_e)N_0(r,t)c\Phi_e(r,t),$$

and luminescence light density $\Phi_i(\lambda_1,r,t)$,

$$\frac{d\Phi_i(\lambda_1,r,t)}{dt} = \frac{cI^*}{3}\nabla^2\Phi_i(\lambda_1,r,t) + \sigma_s(\lambda_e)N_i(r,t)c\Phi_i(\lambda_1,r,t) + \frac{N_i(r,t)}{\tau}L(\lambda_1),$$

where $N_0$ and $N_1$ are ground-state and excited-state population of dye molecules, respectively, $\lambda_e$ and $\lambda_i$ are wavelengths of excitation and luminescence light, respectively, $c$ is the light velocity, $\tau$ is the excited state lifetime, and $L(\lambda_1)$ is the spectral shape of spontaneous emission. Parameters used in our calculation were as follows. The absorption cross section for 532 nm excitation light $\sigma_a$, was $1.7 \times 10^{-20} \text{m}^2$. The stimulated emission cross section $\sigma_s$, was $1.0 \times 10^{-20} \text{m}^2$. The excited-state lifetime $\tau$, was 3.6 ns. In Fig. 2, the solid, dashed, and dotted lines present the numerically calculated spectral width as a function of excitation pulse energy with a single beam excitation. The simulations show good agreement with experimental results for three samples. We developed our simulation model so that we can calculate the luminescence spectral width as a function of the beam separation $d$. The results are shown in Fig. 4 together with the experimental results. The pulse energies indicated by arrows in Fig. 2 were used for the first and the second beams. These results show good agreements with the experimental results. Figure 6 shows the calculated curves for the spatial distribution of the excited-state population of dye molecules in the medium. The lines represent the contour map. We define $L_{th}$ as the full width that the normalized excited-state population becomes 0.5 in the calculation shown in Fig. 6. It is found that the $L_e^*$ obtained from the two-beam spatial-correlation experiment shown in Fig. 4 is in agreement with $L_{th}$ in Fig. 6 for all samples. We can understand that the correlation length $L_e^*$ reflects $L_{th}$.

Let’s consider the relation between $L_e^*$ and $L_a$, shown in Fig. 5. The typical path length for the luminescence light to travel in the gain region under the excitation pulse energy of $W_{th}$ is

$$\xi_{th} = \frac{L_{th}^2}{I^*}.$$ (4)

The gain-length product that the luminescence light obtains during the diffusing process may be $\sigma_e N \xi_{th}$. At the thresh-

### Table I. Relation between correlation length and some parameters.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$N$ ($\times 10^{-4}$ M)</th>
<th>$I_a$ (µm)</th>
<th>$I_e$ (µm)</th>
<th>$L_0$ (µm)</th>
<th>$L_e$ (µm)</th>
<th>$L_e^*/L_a$</th>
</tr>
</thead>
<tbody>
<tr>
<td>No. 1</td>
<td>2</td>
<td>490</td>
<td>100</td>
<td>128</td>
<td>684</td>
<td>672</td>
</tr>
<tr>
<td>No. 2</td>
<td>1</td>
<td>980</td>
<td>20</td>
<td>81</td>
<td>570</td>
<td>555</td>
</tr>
<tr>
<td>No. 3</td>
<td>5</td>
<td>196</td>
<td>40</td>
<td>51</td>
<td>372</td>
<td>349</td>
</tr>
<tr>
<td>No. 4</td>
<td>5</td>
<td>196</td>
<td>20</td>
<td>36</td>
<td>290</td>
<td>259</td>
</tr>
<tr>
<td>No. 5</td>
<td>10</td>
<td>98</td>
<td>20</td>
<td>26</td>
<td>208</td>
<td>163</td>
</tr>
</tbody>
</table>

FIG. 5. Correlation length $L_e^*$, as a function of diffusive absorption length $L_a$. The solid line represents fitting line with a proportional factor 5.0.

FIG. 6. The contour map for the normalized excited state population of 0.5 under the excitation pulse energy of $W_{th}$. The concentrations of dye and the volume fractions of polystyrene for solid, dashed, and dotted lines are $2 \times 10^{-4}$ M, 2%; $5 \times 10^{-4}$ M, 5%; $10 \times 10^{-4}$ M, 10%, respectively. The $r$ axis is the distance from the center of the excitation beam on the incident surface. The $z$ axis is the depth from the incident surface.
old, this product should be equal to $G_{th}$, where $G_{th}$ is the threshold gain to overcome the loss in the medium. Therefore,

$$L_{th} = \sqrt[3]{G_{th} \sigma_a / \sigma_s}.$$  \hspace{1cm} (5)

From Eq. (5), it is seen that the ratio of $L_{th}$ to $L_a$ is proportional to the square root of the ratio of the absorption cross section $\sigma_a$, to the stimulated emission cross section $\sigma_s$. Since dye molecules used in our experiments was a single kind of dye, kiton red, the value of $\sqrt[3]{\sigma_a / \sigma_s}$ is constant and $\sqrt[3]{\sigma_a / \sigma_s}=1.3$ [16]. We consider that $L_{th}$ obtained in the two-beam correlation measurement reflects $L_{th}$ as discussed before. Therefore, the relation between $L_{th}$ and $L_a$ shown in Fig. 5 is well explained in this model. From Fig. 5, $L_{th}$ at the threshold is five times longer than $L_a$. This means that absorption saturation and the resultant increment in the gain volume are essential in the spectral collapse of the random laser. The proportional factor of 5.0 in Fig. 5 would reflect $G_{th}$, which is subject to the loss of the luminescence light in the medium due to the reabsorption, scattering, and geometrical configurations. From the value $\sqrt[3]{\sigma_a / \sigma_s}=1.3$ for kiton red and the proportional factor of 5.0, $G_{th}$ is estimated to be 1.2, that means $G_{th}=1$, which is a reasonable value for the gain threshold.

In summary, we measured the size of the gain volume in an amplifying and scattering medium using the two-beam spatial-correlation method. It was found that $L_{th}$ is proportional to $L_a$. Numerical simulation based on the coupled rate and diffusion equations recaptures the experimental results. These results are well explained when we consider the diffusive propagation of light in the medium in the presence of absorption saturation.

This work was supported by Casio Science Foundation and Hamamatsu Science and Technology Foundation. The authors are grateful to Dr. K. Totsuka for his help in writing the program.