Experimental study of instability in a random laser with immobile scatterers

G. Zhu, Lei Gu, and M. A. Noginov

Phys. Rev. A **85**, 043801 — Published 2 April 2012

DOI: 10.1103/PhysRevA.85.043801

Experimental study of instability in a random laser with immobile scatterers

G. Zhu, Lei Gu, M. A. Noginov
Center for Materials Research, Norfolk State University, Norfolk, VA

Abstract: Pulse-to-pulse instability in the emission from a random laser based on $Nd:Sc_3(BO_3)_4$ ceramic with immobile scatterers was studied below, at, and above the stimulated emission threshold P_{th}. The correlation between pumping and emission intensities was found to be surprisingly low, especially in the vicinity of the lasing threshold. When the sample was excited by reasonably stable (±5%) pumping pulses, a strong fluctuation of output intensities was observed in the range of pumping energies $P_{th}/2<P<2P_{th}$ – the range in which the Lévy statistics of emission fluctuations has been predicted in random lasers. We, thus, report on the first experimental evidence of the Lévy statistics in a random laser with firmly fixed scatterers.

Random lasers are sources of stimulated emission, in which the feedback is provided by scattering in a gain medium [1-3] rather than by mirrors, as in conventional lasers. Random lasers have been first theoretically proposed in 1967 [4] and experimentally demonstrated in 1986 [5]. Since then, this was a topic of intense theoretical and experimental research [6-10] revealing a broad spectrum of random lasers with coherent and incoherent feedback based on solid [1, 5-7] and liquid [8] gain media as well as biological soft matter [9, 10].

Although scatterers are usually randomly distributed and oriented in space, their positions often do not change in time. This happens in *e.g.* mechanically rigid solid-state systems.

Does this mean that equivalent pumping pulses produce the same emission outputs? The studies of the random laser effect in two systems with fixed disorder – compressed glass powder infiltrated with dye [11] and ZnO nanoparticles embedded into a polymeric host [12] – have shown that this is not always the case. Thus, in the former work, the positions of the narrow spectral lines in the stimulated emission of dye were surprisingly unstable and exhibited strong pulse-to-pulse chaotic behavior [11]. In Ref. [12], the observed fluctuations of the random laser output were much stronger at nanosecond pumping than at picosecond pumping.

As photons propagate in a random laser medium over a path-length l, light intensity, I, increases exponentially, $I \propto \exp(l/l_g)$, where l_g is the gain length. In a diffusion approximation, path lengths, l, have exponential probability distribution $p(l) = \frac{\exp(-l/\langle l \rangle)}{\langle l \rangle}$. With an increase of pumping, l_g becomes shorter and the emission gets dominated by exponentially rare and exponentially strong events of light amplification in very long paths [13] (known as "larger than rare" [14] "lucky photons" [15]). Mathematically, when the parameter $\alpha \equiv l_g/\langle l \rangle$ becomes smaller than 2, the Gaussian statistics of intensity fluctuations changes to the Lévy-stable (or Lévy) distribution, for which the average intensity exists but the variance diverges [13]. In scattering gain media with significant inhomogeneous broadening, this transition results in highly irreproducible pulse-to-pulse spectral measurements with random positions of narrow emission lines [13]. The threshold, defined as "gain equal to loss", corresponds to α=1 [13]. When the (instantly created) gain gets sufficiently large and the parameter α becomes smaller than 1/2, the stimulated emission grows so strong that it depletes the population inversion and makes the effective gain length long again [13]. This returns the system to the regime of Gaussian fluctuations, and pulse-to-pulse instabilities become smaller.

Lévy statistics in random amplifying media has been experimentally realized in liquid rhodamine 6G dye with polysterene microspheres and submicron TiO_2 particles [14] and in ensembles of active fiber segments embedded in a bulk of passive point-like scatterers [16]. In both cases, distributions of scatterers changed from pulse to pulse. In a related study, random laser intensity fluctuations in sulforhodamine B dye solution with suspended TiO_2 particles have been studied at different pulse-to-pulse realizations of disorder [17]. The experimental results [17] have been explained with the model describing the laser output in terms of the effective number of photonic modes in the pumped volume and coupling of spontaneous emission to these modes.

In this Letter, we report on the experimental studies of pulse-to-pulse instability in the stimulated emission of a neodymium random laser based on sintered $Nd:Sc_3(BO_3)_4$ ceramic with immobile scatterers. We research the correlation between fluctuations of pumping and emission intensities, and find the correlation to be very poor below and at the random laser threshold, slightly improving above the threshold. We also find that the standard deviation of emission pulse energies significantly increases at pumping intensities surrounding the lasing threshold, when the parameter $\alpha \equiv l_g / \langle l \rangle$ corresponds to the predicted range of the Lévy statistics, $1/2<\alpha<2$.

The experimental sample of $Nd:Sc_3(BO_3)_4$ ceramics was kindly provided by J. Paitz at at the Institut für Kristallzuchtung im Forschungsverbund, Berlin, Germany. Its fabrication is described in Ref. [18]. In brief, the starting constituents were mixed in a required stoichiometric ratio and well homogenized. The powder sample of $Nd:Sc_3(BO_3)_4$ was first synthesized by a solid-state reaction. The pellets were fabricated by hydrostatic pressing followed by sintering at 1100^{o}C for 24 hours. The sintered sample had milky-bluish appearance and was as hard and solid as a rock, precluding any motion of constituting it microcrystallines relative to each other.

In the experiments, a piece of $Nd:Sc_3(BO_3)_4$ ceramics was pumped with 5 ns pulses of an optical parametric oscillator at λ_{pump} = 808 nm. The absorption coefficient at this wavelength (determined based on the transmission spectrum of the $Nd:Sc_3(BO_3)_4$ single crystal [19]) was equal to ≈44 cm^{-1}, and the photon transport mean free path was equal to 7μm [20] (at λ= 514.5 nm).

The diameter of the pumped spot, measured using a knife-edge technique, was equal to 0.42 mm. The emission was detected by a photomultiplier attached to the exit slit of a monochromator. In the instability measurements reported in this Letter, its wavelength was set at the maximum of emission in $Nd:Sc_3(BO_3)_4$ (λ = 1061.5 nm) [7]. The signal generated by the photomultiplier was recorded with the 1 GHz oscilloscope (50 ohm input impedance, 4 ns time resolution).

At weak excitation, spontaneous emission at the Nd^{3+} $^4F_{3/2}\rightarrow{}^4I_{11/2}$ transition was characterized by a rather broad spectrum ranging from ≈1050 nm to ≈1080 nm [7] and the decay-time equal to 24 μs [7]. When the pumping energy was increased to a critical threshold value, a typical of neodymium random lasers behavior was observed [7]. Thus, the emission spectrum narrowed to a single line (at 1061.5 nm), the emission kinetics shortened from microseconds to nanoseconds (upper inset of Fig. 1), the peak emission intensity increased dramatically, and the input-output dependence demonstrated a distinct threshold (Fig.1, squares). The latter dependence is fitted with the theoretical formula [21]

$$E = \frac{\tau_{res}\nu_e}{2V_p\nu_p}\left[\left(P-P^*\right)+\sqrt{\left(P-P^*\right)^2+4\zeta PP^*}\right], \qquad (1)$$

where E and P are the emission and pumping energies, P^* is the threshold pumping energy, ζ is the probability with which spontaneously emitted photons populate the lasing mode(s), τ_{res} is the residence time of photons in the pumped medium, V_p is the pumped volume, and ν_e and ν_p are the light frequencies at the emission and pumping wavelengths, respectively (Fig. 1, trace 1).

In order to study pulse-to-pulse fluctuations of emission intensities, we recorded multiple emission kinetics at several nominal pumping energies – below, at, and above the lasing threshold. Their maximal values are plotted versus the pulse number in Fig. 2. [Because of typical to lasers instabilities, pumping energies fluctuated as well. Therefore, for the same pulses, we recorded pumping pulse kinetics (by splitting off a small fraction of the OPO beam) and plotted their peak values along with the emission intensities, Fig. 2.] We have found that the emission fluctuations are much larger at the lasing threshold than below or above the threshold, Fig. 2.

The degree of correlation between fluctuations of pumping pulse intensities and corresponding to them emission intensities has been analyzed with the aid of the Pearson correlation coefficient defined as

$$C = \frac{\sum_i \left[\left(E_i - \overline{E} \right) \left(P_i - \overline{P} \right) \right]}{\sqrt{\sum_i \left(E_i - \overline{E} \right)^2 \sum_i \left(P_i - \overline{P} \right)^2}}. \qquad (2)$$

Here P_i and E_i are the peak pumping intensity and corresponding to it peak emission intensity recorded for each laser pulse; $\overline{P}$ is the mean pumping intensity calculated for multiple events that correspond to the same nominal pumping energy, $\overline{P} \equiv \left(\sum_i^n P_i \right) / n$; $\overline{E}$ is the analogously calculated mean emission intensity $\overline{E} \equiv \left(\sum_i^n E_i \right) / n$; and n is the number of points in the data series.

The correlation coefficient is maximal, C=1, when deviations of P_i and E_i from their mean values are linearly proportional to each other with a positive proportionality coefficient γ, $\left(P_i - \overline{P} \right) = \gamma \left(E_i - \overline{E} \right)$, and the distribution of (E_i,P_i) points (plotted in *E vs P* coordinates) forms a straight line with a positive slope, see right panel of Fig. 3. A particularly important case of the maximal correlation coefficient corresponds to linear proportionality of P_i and E_i. When positive deviation of E_i from its mean value $\left(E_i - \overline{E}\right)$ is proportional to negative deviation of P_i from its mean value $\left(P_i - \overline{P}\right)$, the correlation coefficient is minimal, C=-1. In this case, (E_i,P_i) points form a straight line with a negative slope. When the quantities $\left(E_i - \overline{E}\right)$ and $\left(P_i - \overline{P}\right)$ do not correlate with each other, the correlation coefficient is equal to 0, and the (E_i,P_i) points form a filled circle. When the correlation or anti-correlation exists but is not perfect, the correlation coefficient corresponds to the range $0<C<1$ or $-1<C<0$, and the (E_i,P_i) points form an ellipse-like shape, which long axis has positive or negative slope, respectively.

The correlation coefficients determined as discussed above are plotted for several pumping energies in the main panel of Fig. 3, and the spreads of the (E_i,P_i) points corresponding to average pumping energies equal to 6 mJ and 11 mJ are plotted in the lower panel of Fig. 3. One can see that below and around the lasing threshold, the correlation between the fluctuations of pumping and emission is very poor, with the correlation coefficient being close to zero. Above the threshold, the correlation coefficient shows the trend of increase, although in a rather random and not monotonous way. [Note that qualitatively similar (although numerically slightly different) results have been obtained when integrals under the kinetics curves have been used in calculations instead of peak intensities.]

When two detectors were set up to measure the intensity of the pumping light scattered by the sample into two different solid angles (≈ 90^{o} apart), the correlation coefficient was found to be equal to C=0.96 as indicated by the horizontal line in the main panel of Fig. 3. The deviation of this correlation coefficient from unity was probably due to electronic noise in the detectors and the oscilloscope.

As pumping energies in our experiment were not very stable, the question arose whether the instability of the emission output was solely determined by the instability of the pumping. To answer this question, we judiciously selected from each data set only those points for which the pumping intensities P_i deviated from the mean pumping intensity $\overline{P}$ by less than ±5%. Such subset of the data points plotted in Fig. 2b is shown in the inset of the figure. As one can see, the emission intensities fluctuate by large margin even if the (pre-selected) pumping energies are fairly stable.

Instabilities of pumping and emission intensities can be quantified in terms of the standard deviation

$$\sigma_I = \sqrt{\frac{\sum_i^n (I_i - \overline{I})^2}{n}} \quad , \tag{3}$$

where I stands for P or E, respectively. The standard deviation calculated for the pumping intensities confined within the ±5% limit was as low as σ_P=0.024. At the same time, around the lasing threshold, $\overline{P}_{th}$, the standard deviation of the corresponding emission intensities was as high as $\sigma_S = 0.24$. One can see that the values σ are reasonably small below and above the lasing threshold ($\overline{P} \le \overline{P}_{th}/2$ and $\overline{P} \ge 2\overline{P}_{th}$) and increase dramatically in the vicinity of the threshold ($\overline{P}_{th}/2 \le \overline{P} \le 2\overline{P}_{th}$), Fig. 1. Note that the latter range exactly corresponds to the range of parameters $\alpha \equiv l_g/\langle l \rangle$, for which fluctuations of random laser intensities are predicted to have the Lévy statistics, $1/2 < \alpha < 2$ [13]. We, thus, report on the first evidence of the Lévy statistics observed in a random laser with stationary scatterers.

According to a simple model, which assumes that laser input and output are firmly related to each other with Eq. (1), instability of emission around the threshold is predicted to follow the (nonlinearly) amplified instability of pumping, with more intense pumping pulses resulting in relatively much stronger emission pulses (low inset of Fig. 1). Using a random number generator, we have simulated an instability of pumping and, based on the input-output curve of Fig. 1, calculated the corresponding to it instability of emission quantified in terms of the standard deviation σ_E and the pumping-emission correlation coefficient C. As one can see in Fig. 1, values σ_E have a peak around the lasing threshold, in a qualitative agreement with the experiment. However, this peak is much higher and much narrower than the experimental one. Even more striking difference between the experiment and the model prediction is found in the behavior of the correlation coefficient C (Fig. 3). We, thus, conclude that the experimentally observed fluctuations of the random laser emission cannot be explained in terms of a simple model accounting for an instability of pumping magnified by nonlinearity of the input-output curve, and the nature of the emission volatility is more complicated. This should become a subject of a separate theoretical study. Strong nonlinear interactions of random laser modes [23, 24] can be a contributing factor to the intensity instabilities reported here.

To summarize, we have studied pulse-to-pulse instability in the emission of a random laser based on sintered $Nd{:}Sc_3(BO_3)_4$ ceramic with immobile scatterers below, at, and above the threshold. We have found a surprisingly low correlation between the pumping and emission intensities, in particular at and below the random laser threshold. When the sample was excited with reasonably stable (±5%) pumping pulses, a strong fluctuation of output intensities was observed in the range of pumping energies $P_{th}/2<P<2P_{th}$ – the range in which the Lévy statistics of emission fluctuations has been predicted in random lasers [13]. We, thus, report on the first experimental evidence of the Lévy statistics in a random laser with scatterers firmly fixed in space and time.

The authors cordially thank J. Paitz and G. Huber for providing experimental samples. The work was partly supported by the NSF PREM grant # DMR 0611430, NSF NCN grant # EEC-

0228390, AFOSR grant # FA9550-09-1-0456, NSF IGERT grant # DGE 0966188, and subcontract from UTC #10-S567-001502C4.

References and Notes

1. M. A. Noginov, Solid-State Random Laser, Springer, printed in the USA, 2005.
2. H. Cao, J. Phys. A: Math. Gen. 38, 10497 (2005).
3. D. S. Wiersma, Nature Physics **4**, 359 (2008).
4. V. S. Letokhov, Sov. Phys. JETP **26**, 835 (1968).
5. V. M. Markushev, V. F. Zolin, and Ch. M. Briskina, Sov. J. Quantum Electron. **26**, 835, 281 (1986).
6. H. Cao, Y. Zhao, S. T. Ho, E. W. Seelig, *et al*., Phys. Rev. Lett. **82**, 2278 (1999).
7. M. A. Noginov, N. E. Noginov, H. J. Caulfield, P. Venkateswarlu, T. Thompson, M. Mahdi, and V. Ostroumov, J. Opt. Soc. Am. B **13**, 2024 (1996).
8. N. M. Lawandy, R. M. Balachandran, A. S. L. Gomes, and E. Sauvain, Nature **368**, 436 (1994).
9. R. C. Polson and Z. V. Vardeny, Phys. Rev. B **71**, 045205 (2005).
10. A. Smuk, E. Lazaro, L. P. Olson and N. M. Lawandy, Optics Communications **284**, 1257 (2011).
11. S. Mujumdar, Volker Türck, R. Torre, and D. S. Wiersma, Phys. Rev. A **76**, 033807 (2007).
12. D. Anglos, A. Stassinopoulos, R. N. Das, G. Zacharakis, et al., J. Opt. Soc. Am. B **21**, 208 (2004).
13. S. Lepri, S. Cavalieri, G.-L. Oppo, and D. S. Wiersma, Phys. Rev. A **75**, 063820 (2007).
14. D. Sharma, H. Ramachandran, and N. Kumar, Fluct. Noise Letter. **6**, L95 (2006).
15. S. Mujumdar, M. Ricci, R. Torre and D. S. Wiersma, Phys. Rev. Lett. **93**, 053903 (2004).
16. D. Sharma, H. Ramachandran, and N. Kumar, Optics Lett. **31**, 1986 (2006).
17. K. L. van der Molen, A. P. Mosk, and Ad Lagendijk, Phys. Rev. A **74**, 053808 (2006).

18. M. A. Noginov, S. U. Egarievwe, H. J. Caulfield, N. E. Noginova, M. Curley, P. Venkateswarlu, A. Williams, and J. Paitz, Optical Material **10**, 1 (1998).

19. A Czochralski grown single crystal of Nd:$Sc_3(BO_3)_4$ was kindly provided by G. Huber at University of Hamburg.

20. M. Bahoura and M. A. Noginov, J. Opt. Soc. Am. B **20**, 2389 (2003).

21. M. A. Noginov, I. N. Fowlkes, G. Zhu, and J. Novak, Phys. Rev. A **70**, 043811 (2004).

22. "Correlation and dependence", in *Wikipedia*, Retrieved August 17, 2011, from http://en.wikipedia.org/wiki/Correlation_and_dependence.

23. H. E. Türeci, Li Ge, S. Rotter, and A. D. Stone, Science **320**, 643 (2008)

24. M. Leonetti and C. Conti, J. Opt. Soc. Am. B **27**, 1446 (2010)

Figures Captions

Figure 1 (color online). Squares and trace 1 – input-output dependence of the $Nd{:}Sc_3(BO_3)_4$ ceramic random laser and its fit with Eq. 1. Circles – experimental standard deviation of emission intensities σ_E calculated for pre-selected pumping pulses, which instability did not exceed ±5% (σ_P=0.024). Triangles and trace 2 – values σ_E calculated (using a simple model accounting for strong nonlinearity of the input-output curve around the threshold) for simulated pumping pulses with σ_P=0.024. Shaded area corresponds to the range of pumping energies, $P_{th}/2<P<2P_{th}$, in which the Lévy statistics is predicted [13]. Upper inset: typical stimulated emission kinetics above the threshold. Lower inset: Zoomed input-output curve of the main panel. Relatively small deviations of pumping energies from their nominal values Δ_P (as pumping changes from A to B) correspond to relatively large changes in emission intensities Δ_E.

Figure 2 (color online). Intensities of pumping pulses (squares and solid line) and emission pulses (diamonds and dashed line) plotted against the pulse number. Nominal pumping energies: 1mJ (a), 6mJ (b), and 10 mJ (c). Standard deviation of pumping intensities $\sigma_P \approx 0.12$. Inset: Same as in the main frame of figure 2 for pre-selected pumping pulses, which intensity fluctuations did not exceed ±5% (σ_P=0.024).

Figure 3 (color online). Main panel: Pumping-emission correlation coefficient C; diamonds – experiment, squares – calculations based on a simple model accounting for a strong nonlinearity of the input-output curve. Solid line – correlation coefficient C calculated for two scattered pumping light intensities detected by two photodetectors. In experiments and calculations, the instability of pumping corresponded to σ_P=0.12. Low panel (left): The spread of experimental 'emission vs pumping' (P_i,E_i) points at average pumping energy 6 mJ (C=0.008). Lower panel (right): Same as above at average pumping energy 11 mJ (C=0.5). Right panel: spreads of the

(P_i,E_i) points corresponding to C=1 (top), C=0 (middle), and C=-1 (bottom). (Adopted from [22].)

Figure 1

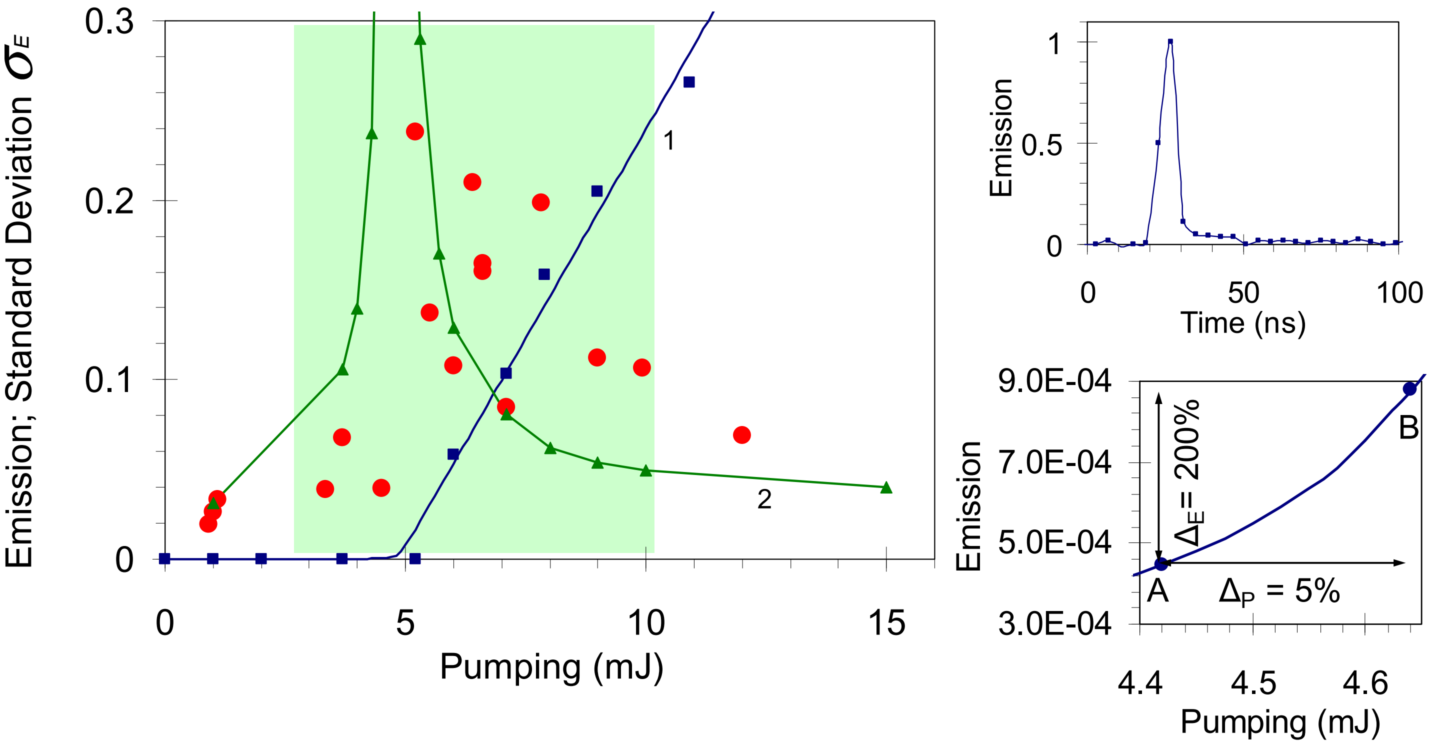
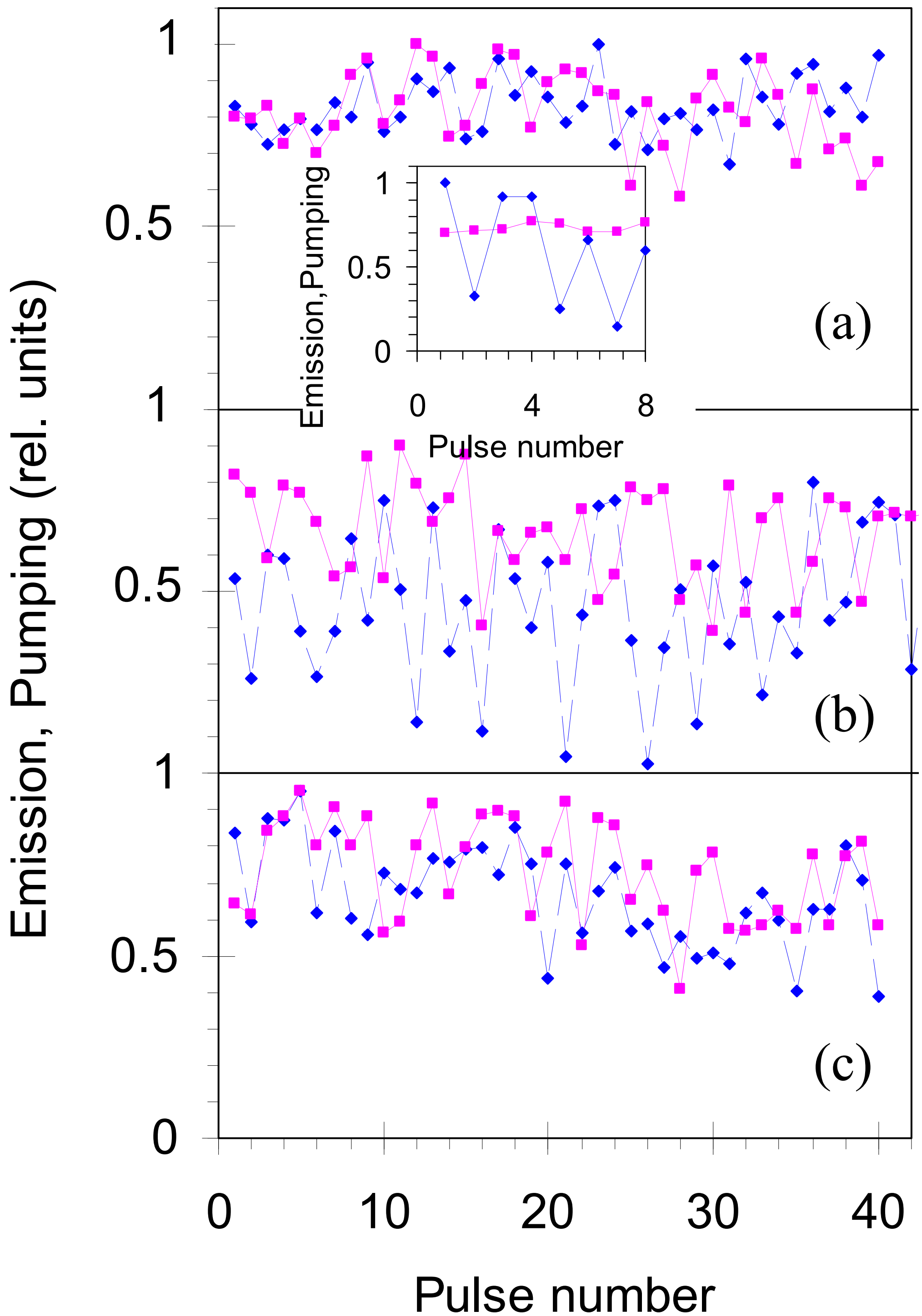
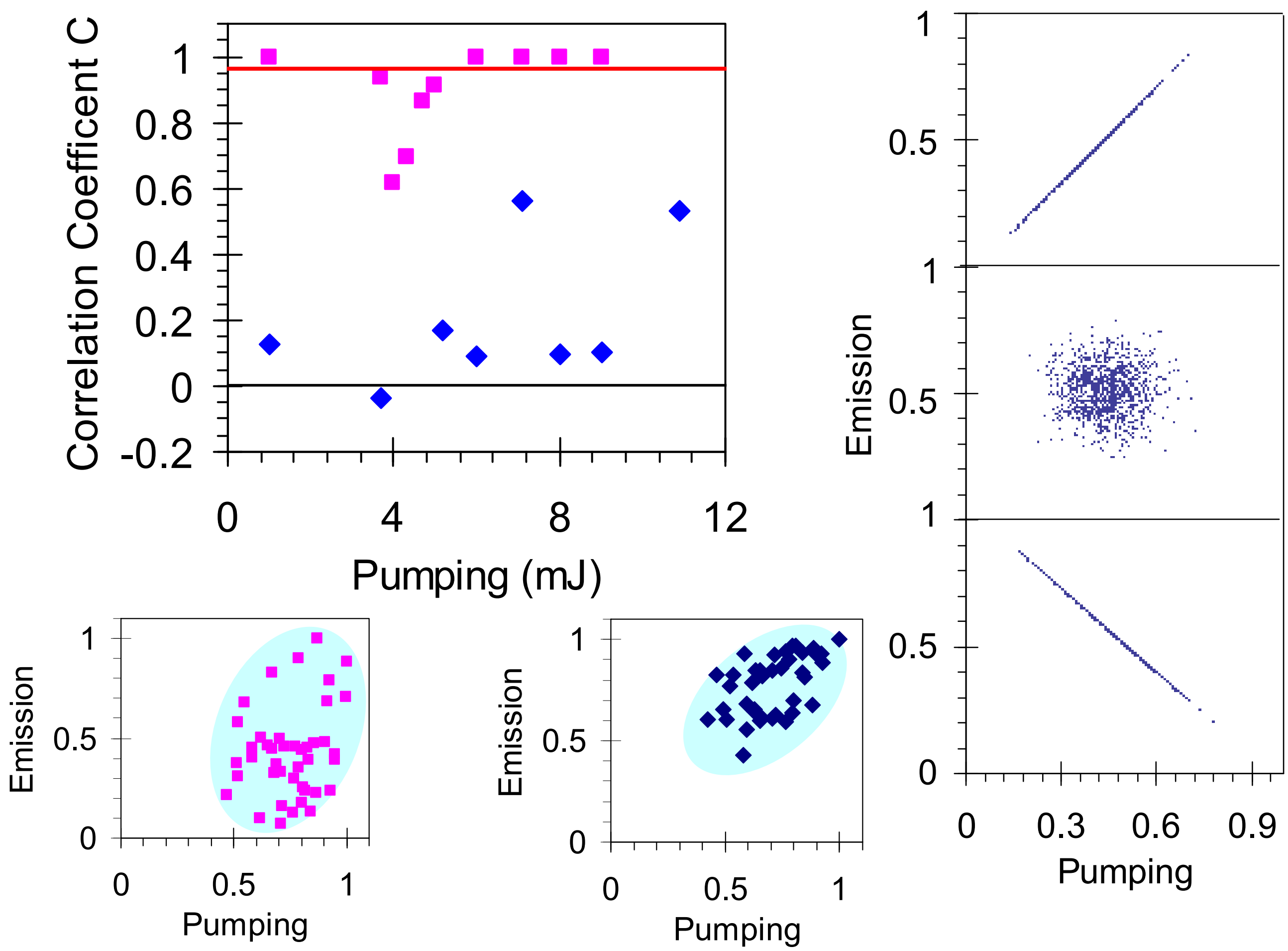

0.3
0.2
0.1
0
Emission; Standard Deviation σ_E
1
2
0
5
10
15
Pumping (mJ)
1
0.5
0
Emission
0
50
100
Time (ns)
9.0E-04
7.0E-04
5.0E-04
3.0E-04
Emission
Δ_E= 200%
B
A
Δ_P = 5%
4.4
4.5
4.6
Pumping (mJ)

Figure 2



Emission, Pumping (rel. units)
1
0.5
1
0.5
1
0.5
0
(a)
(b)
(c)
0
10
20
30
40
Pulse number
Emission,Pumping
1
0.5
0
0
4
8
Pulse number

Figure 3



Correlation Coefficent C
1
0.8
0.6
0.4
0.2
0
-0.2
0
4
8
12
Pumping (mJ)
Emission
0.5
1
0
0.5
1
Pumping
Emission
0.5
1
0
0.5
1
Pumping
1
0.5
1
Emission
0.5
1
0.5
0
0
0.3
0.6
0.9
Pumping