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SUPPLEMENTARY INFORMATION

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Supplementary Information

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Statistical analysis

The statistical analysis reported in this paper was based on a set of 4000 single shot random laser spectra acquired with a camera synchronized with the pump laser pulses. The selection of the peaks is based on the camera images, where a single resolution-limited random laser peak appears as a circle. Each circle is the image of the optical aperture at the focal plane of the spectrometer dispersed by the monochromator. When the apertures of the monochromator are fully open, the spectral resolution (represented by the diameter of the circles) is determined uniquely by the entrance optics. The observed diameter of 150 μm (27 pixels), corresponds to a spectral resolution of 0.83 THz. The spectral resolution was measured by replacing the random laser source with a passive scattering medium and illuminating it with a monochromatic pump beam (wavelength 532 nm).

An algorithm selects the brighter disks for each shot in the reference beam, calculating its relative geometric center with sub-pixel accuracy. Using the reference signal instead of the probe to determine the frequencies of the peaks allows us to avoid any apparent frequency shift that might be induced by steep modulations of the transmission function. The intensity of each peak is obtained by integration over the circled area. Occasional pairs of geometric centers occurring at a relative distance smaller than the disk size are discarded to avoid overlapped modes. Using the geometric center rather than its maximum intensity position to determine the

estimated peak frequency allows to reduce errors associated to speckle fluctuations introduced by the multimode fiber.

The integrated intensity of the selected disks in the reference beam is used to normalize the intensity of the corresponding signal disks. The target transfer function is retrieved by taking the average normalized intensity of all modes and binning them within a pre-defined frequency grid (in our case we defined the width of each bin equal to that of the equivalent frequency width of individual camera pixels).

Random Laser sample

To optimize the efficiency of the spectral reconstruction method, it is important to consider the average number of modes that reach the lasing threshold (and hence generate a sharp peak) in each shot. According to the model developed by Karen et al. [30], the average number of lasing modes $\overline{\mathrm{N}}$ should be written as:

$$\overline{\mathrm{N}} = p \cdot \mathrm{N},$$

[eq.2]

where p is the probability of lasing and N the total number of modes in the system. By exploiting the usual balance between spontaneous and stimulated emission, and assuming that all modes contribute equally, this model predicts the beta factor of a random laser to be $\beta=\overline{\mathrm{N}}/\mathrm{N}$, where β is equal to p. For picosecond excitation, p is in the range 0.05-0.1[31]. The average number $\overline{\mathrm{N}}$ of lasing modes in our sample is 5, while the total number of cavity modes can be estimated with the general relation $\mathrm{N}=8\pi n^3 \mathrm{V}\Delta\lambda/\lambda^4$. Under the condition of complete gain saturation and weak scattering, the cavity volume can be estimated by a cylinder with a radius equal to the diameter of the pump beam and length equal to the penetration length of the pump light[32] which is about 300 μm for our sample. This results in $\mathrm{N}\sim$ 1800 and p equal to 0.003, which is one order of magnitude lower than the typical value measured in literature. The intensity fluctuations of the random laser output can be further controlled by the density of dye molecules and the excitation area.

There are several aspects playing a role in the optimization of a liquid sample of an optically pumped RL. The relevant parameters that concur to obtain narrow peaks are the mean free path of the scattering solution and the optical gain provided by the dye. The interplay between these parameters can be tuned to obtain a

suitable condition for sparse narrow peaks with high prominence with respect to the gain medium (due to gain depletion effects).

We have characterized spectrally an array of nine different RL mixtures where we have varied the dye and nanoparticle concentration independently. At very low concentration, the mean free path is inversely proportional to the scatterer concentration, while the optical gain is proportional to the dye molecule concentration. Figure 1 shows an illustrative single shot emission for each RL mixtures. Different samples are arranged in columns representing increasing dye concentration of 5 mM, 15 mM, 45 mM from left to right, while the rows from top to bottom are characterized by increasing scatterer concentration of 10^{12}, $3 \cdot 10^{12}$, $6 \cdot 10^{12}$ particles/cm^3. Even if the number of peaks for each mixture is subject to fluctuations, on average the emissions of samples in the first row (i.e., lowest nanoparticle concentration) contains fewer narrow peaks as compared to the other mixtures, as also expressed by the density of peaks per nanometer, *n*, calculated as the average number of peaks in one spectrum divided by the FWHM of the gain curve. The optimal degree of spectral separation between lasing modes will depend on the properties of the spectrometer used, and in general should be set to be larger than its nominal resolution. In our case (instrumental resolution: 0.8 nm), we varied the physical properties of the random laser used and its pumping conditions so to set an average number of 0.5 modes per nm, corresponding to an average separation of 2.5 times the instrumental resolution.

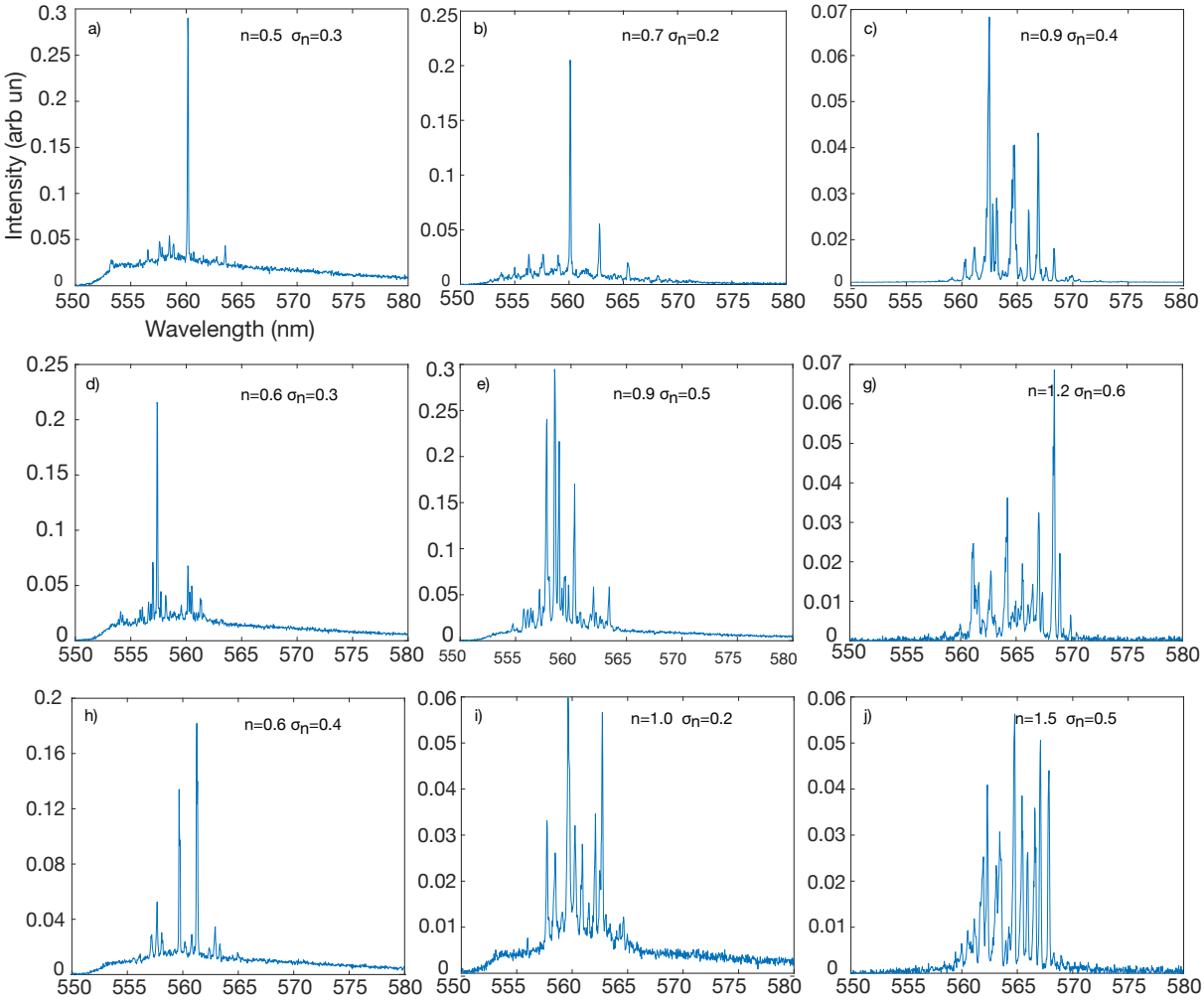

Figure S1: Single shot emission spectra for different RL mixtures. The columns from left to right present an increasing dye concentration of 5 mM, 15 mM, 45 mM, respectively, while the rows from top to bottom show spectra with increasing scatterer concentration of 10^{12}, $3{\cdot}10^{12}$, $6{\cdot}10^{12}$ particles/cm^3.