Tunable degree of localization in random lasers with controlled interaction

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We show that the degree of localization for the modes of a random laser (RL) is affected by the inter mode interaction that is controlled by shaping the spot of the pump laser. By experimentally investigating the spatial properties of the lasing emission we infer that strongly localized modes are activated in the low interacting regime while in the strongly interacting one extended modes are found lasing. Thus we demonstrate that the degree of localization may be finely tuned at the micrometer level.

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RL are among the most complex systems in photonics, encompassing structural disorder and nonlinearity,1 and ranging from micron sized optical cavities2 to kilometer-long fibers3. Attention on this systems has been constantly growing as the number of potential applications, ranging from object coding4 to speckle-free illumination5.

First-principle time domain simulations show that modes of a RL arise from electromagnetic states6–8, that may appear in localized or extended fashion. Several experiments confirmed this view2,9,10, and tried to address the connection between the structure pumping condition, or gain and the degree of localization12–14. The presence of many modes may give rise to unique phenomena: in a linear system extended necklace states spread over the sample via multiple (localized) resonances15 while in a system with gain the inter-mode coupling affects the whole spectrum generating mode repulsion16 directly connected to the nonlinear interaction17.

On the other hand it has been recently demonstrated that the inter-mode coupling plays a critical role in the onset of two fundamentally different RL regimes, distinguished by the shape of the emission: a “resonant feedback random laser” (RFRL)18, which appears as a set of sharp peaks oscillating independently at fixed spectral positions, and the “intensity feedback random laser” (IFRL)19, characterized by a smooth single line narrowed spectrum. By using a tailored spatial shape of the pump area20,21, a switching between the two can be achieved. In fact a RFRL is observed when activating a set of weakly interacting resonances while IFRL is produced under strong interaction that leads to a mode-locked synchronized regime.

In this Letter, we present the experimental evidence that the mode extent is affected by the inter mode interaction. We demonstrate that the degree of localization may be finely tuned by controlling the pump shape.
when the number of modes activated in a single isolated disordered structure is increased, the extent of a single mode is also augmented. Our set up comprises a spatial light modulator (SLM) that is imaged as a pumped area in a rhodamine solution pool where a single cluster of titanium dioxide nanoparticles is embedded. The illuminated area is composed by a circular spot centered on the cluster that prepares the system barely below the lasing threshold and by two wedges of angular aperture Θ that pump the rhodamine bath surrounding the cluster, so that amplified spontaneous emission of the dye pumps the region in a directional and controlled manner (see Fig. 1 for a scheme of the setup and Fig. 2a for an image of the pump shape). The control on the degree of mode interaction is exerted by varying the angular span of the illuminated wedges thus increasing the number of excited cluster modes which, through their mutual interaction, mediate the involvement of more modes in the lasing action.

A recently developed approach (details in 21) allows to fix the amount of lasing modes brought over the threshold by controlling the Θ parameter. We show in the following that the spatial shape of the intensity on the sample at a fixed wavelength is strongly affected by the pumping condition and by the strength of the interaction. In figure 2b-e an image of the cluster (sample C1) is shown for various values of Θ. While at low Θ the image pattern displays hotspots embedded in low intensity regions, when Θ is larger the intensity shows a smoother profile where no dark areas are evident.

In a RFRL, modes have sub-nanometer FWHM and lie in a limited spectral range (see Fig. 3a), thus simple image analysis does not allow to obtain quantitative information about the spatial extent of every single mode; to this aim both spatial and spectral resolution is needed 22. This is achieved by a suitably designed experimental setup: an image of the sample is formed through a 50× magnification optics (microscopy objective and eyepiece) in an plane that is scanned by a motorized fiber connected to a spectograph. The result of a scan session is a complete spatio-spectral map.

The spatial extent of a mode resonant at λ may be obtained by selecting the intensity emitted at the corresponding wavelength. Examples of the map for different modes (frequencies) are given in Fig. 3. Panel a shows the complete spectrum emitted by (whole 23) cluster C3 (whose shape is reported in figure 4d) for Θ = 6°, panels b, c, and d show the spatial distribution of the intensity for the peaks labeled in the spectrum. Each pixel in the images corresponds to a 1 × 1µm area. The three most intense modes dwell in different areas of the cluster. Figures 4a, b, and c show the spatial extent of the most intense mode of the cluster C3 for Θ = 20°, Θ = 40° and Θ = 120°. The physical shape of the cluster can be seen in panel d, which shows the image of the scattered light obtained by pumping at fluences well below the lasing threshold and summing contribution at all wavelengths. Previous work showed that the Θ parameter governs the RL regime allowing the transition from RFRL to a IFRL 20. Fig. 4 shows that this parameter also affects the spatial properties of single modes.

The degree of localization of a single mode at wavelength λ with spatial profile of intensity I(λ, x, y), is given by the inverse participation ratio $P(\mu m^{-2})$

$$P_\lambda(\Theta) \equiv \frac{\int I(\lambda, x, y)^2 dx dy}{(\int I(\lambda, x, y))^2}$$

and by the localization length $\Omega_\lambda(\mu m)^{-2}$

$$\Omega_\lambda(\Theta) = 1/\sqrt{P_\lambda}.$$  

Fig. 5 shows $\Omega$ as a function of Θ for two different clusters, C3 (Fig. 5a) and C6 (Fig. 5b). Black squares refer to the ten most intense modes, their average for each angular span being represented by the open circles. For C3, $\Omega$ grows from ca. 4 to 6µm (50% increment of the mode extension), while for C6 it grows from ca. 4.5 to 8µm (about 80% increment in the mode extension). In the latter case, the larger $\Omega$ is ascribed to the larger size of C6, in fact when Θ is sufficiently large, modes spread over the whole sample and their shape resembles that of the cluster (see insets of figure 5). These results are valid not only for all the 10 most intense modes for C3 and C6, but also for all the modes observed in four other clusters with comparable spatial dimension (not reported).

It is well known that factors like gain 14 or losses 13 may influence the spatial shape of modes. It is clear that in the present case none of this features may provide an explanation for the measured phenomena. Indeed the pumped area of the cluster is kept constant and the same throughout the measurements. We take care of this fact by exploiting a suitable pumping shape: the disk shaped area centered on the cluster (circular red area in figure...
localized oscillation at frequency different from their natural resonance. Hence a
and force them to oscillate at its own
\( \lambda \)
open cavities be understood by taking into account mode-coupling in
(Fig. 4), for a given realization of disorder, can instead
pumped for all the values of \( \Theta \).

2 (a)) assures that the whole cluster is homogeneously
pumped for all the values of \( \Theta \).

The fact that the mode shape at \( \lambda \) changes with \( \Theta 
(Fig. 4), for a given realization of disorder, can instead
be understood by taking into account mode-coupling in
open cavities, a mode at \( \lambda_1 \) can excite other modes
and force them to oscillate at its own \( \lambda_1 \), i.e. at a frequency
different from their natural resonance. Hence a
localized oscillation at \( \lambda_1 \) spatially broadens subsuming
several other modes in other locations. However, lossy
(or scarcely pumped) modes are not able to retain energy
at frequencies far from their natural resonances (small \( \Theta 
regime). On the contrary, when increasing \( \Theta \), modes in
any point in the structure can sustain oscillations at any
frequency and, as they are all coupled, they synchronize,
resulting in large scale coherent emission.

This particular mechanism in which a single laser mode
alters the oscillation frequencies of its neighbors to extend
its effective localization length has never been measured
previously, nor has, to the best of our knowledge, the
prediction of such behavior been made. The maximum
extent achievable by an individual mode is limited by the
disordered structure: this is confirmed by the insets of
Fig.5 which compare single mode size at high \( \Theta 
with the shape of the corresponding cluster (they turn out to
be very similar) and by the greatest achieved value of the
inverse participation ratio which is larger for bigger
clusters.

Conclusions — We show evidence of the effect inter
mode interaction has on the spatial extent of lasing
modes in a RL. The coupling between modes allows the
spreading of the energy distribution of a mode into the
whole amplifying region. This may be explained by the
fact that a stronger mode forces weaker neighbors to
oscillate at its resonant frequency, increasing numbers of
modes being involved as \( \Theta \) increases. The ability to con-
trol the onset of a collective oscillation and the mode ex-
tent at the micrometer level represents, in our opinion, a
road to many potential applications. Future generations
of light driven photonic chips, for example, may take advan-
tage of similar phenomena needing strongly localized
and weakly interacting resonances when used to store in-
formation while a strong inter-mode interaction could be
exploited at the elaboration step. Other possible appli-
cations for clusters as light sources is in the develop-
ment of tunable optical coherence tomography techniques,
and in speckle-less coherent imaging.

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