Resonance-driven random lasing

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A random laser is a system formed by a random assembly of elastic scatterers dispersed into an optical gain medium[1]. The multiple light scattering replaces the standard optical cavity of traditional lasers and the interplay between gain and scattering determines the lasing properties. All random lasers studied to date have consisted of irregularly shaped or polydisperse scatterers, with a certain average scattering strength that was constant over the frequency window of the laser[2–4]. In this letter we consider the case where the scattering is resonant. We demonstrate that randomly assembled monodisperse spheres can sustain scattering resonances over the gain frequency window, and that the lasing wavelength can therefore be controlled by means of the diameter and refractive index of the spheres. The system is therefore a random laser with an *a priori* designed lasing peak within the gain curve.

In recent years the interest in random lasing has grown very rapidly, particularly following the observation of this phenomenon in powdered laser crystals[5], ceramics[6], organic composites[7], and even biological tissue[8]. The necessary condition for a random laser is that the material is multiply scattering light, which means that the transport mean free path (the average distance over which the scattered light direction is randomized) $\ell_t \ll L$, where L is the sample size. The other fundamental quantity is the gain length ℓ_g, which represents the path length over which the intensity is amplified by a factor e^{+1}. The interaction between gain and scattering determines the unique properties of the random laser and, in particular, defines the critical thickness for the sample (in slab geometry) to lase, $L_{cr} = \pi\sqrt{\ell_g \ell_t/3}$ (ref. 9). Unlike in ordinary lasers, the resulting light emission is multidirectional, but the threshold behaviour[3], the photon statistics[10,11] and relaxation oscillations[12,13] are very similar to those of standard lasers. The spectral output of a random laser system contains narrow emission spikes[4], which for large spectral width can merge into a smooth peak with an overall narrowing of the spectrum in most experimental configurations[3,14], like the one considered in this paper.

Wavelength tunability is a crucial property of lasing devices. In regular lasers this is easily achieved by tuning the resonance frequency of the resonator. The same principle has also been applied in more complex cavity structures, such as distributed feedback lasers and photonic crystals lasers, in which the cavity modes are the Bloch modes associated with the periodic structure. Tuning the lattice constant then provides a simple tool to tune the laser for high-quality photonic crystals[15,16] or with localized periodicity[17,18]. These tricks do not work in random structures due to the absence of periodicity. Here we will show, however, that even in a completely random system with no periodicity, resonant tunability can be achieved based on single-particle resonances.

A random system, composed of particles of arbitrary shape and size, has a transport mean free path that is nearly spectrally flat (nonresonant), at least over a wavelength range of ~100 nm. Under this condition, conventional random lasing occurs at a wavelength where $\ell_g(\lambda)$ has its minimum; that is, at the maximum of the gain curve. To obtain a random system where lasing modes can be selected at specific wavelengths, we propose to make use of a medium with resonances in the transport parameters. Our idea is to exploit the resonances in scattering coefficients, called Mie resonances[19], that are present when the sizes are comparable with the wavelength of the incident light, which can be easily achieved for regularly shaped resonators such as disks, cylinders or spheres. In a random assembly of such identical scatterers, the resonances survive as peaks in the transport parameters, and in particular in $\ell_t(\lambda)$. We chose monodisperse polystyrene spheres as constituents of a three-dimensional, solid random system that we have dubbed 'photonic glass'[20] (see Methods). Here we report on the random lasing action from such photonic glasses. The resonant nature of $\ell_t(\lambda)$ selects the lasing energy. We experimentally demonstrate that the lasing wavelength becomes very sensitive to the diameter, d, of the constituent spheres and follows the resonances of the system. In a system with a broad gain curve we managed to observe mode competition this way, because we were able to access more than one resonance with comparable gain.

Self-assembly of monodisperse spheres is the technique most commonly used to grow direct opals[21]. To obtain a disordered packing of spheres a modified self-assembly method has been developed (see Methods). The photonic glass obtained in this way was used as the basis of an amplifying system by embedding dry organic laser dye. To that end, 4-dicyanomethylene-2-methyl-6-p-dimethylaminostyryl-4H-pyran (DCM) special was dissolved in pure ethanol and the solution was infiltrated into the photonic glass. Complete ethanol evaporation was assured by gently heating the sample. The amount of DCM was 0.3 wt% for all samples considered here; this was verified by comparative weight measurements.

A scanning electron microscope (SEM) image of such photonic glass is depicted in Fig. 1a. Autocorrelation of the SEM image of the surface and cleaved edge revealed a completely disordered system without positional correlation[20]. Static and dynamic measurements of the photonic glass light transport have shown matching resonances for $\ell_t(\lambda)$ and the diffusion constant of light $D(\lambda)$ (ref. 22). In Fig. 1b–d, transmission measurements are

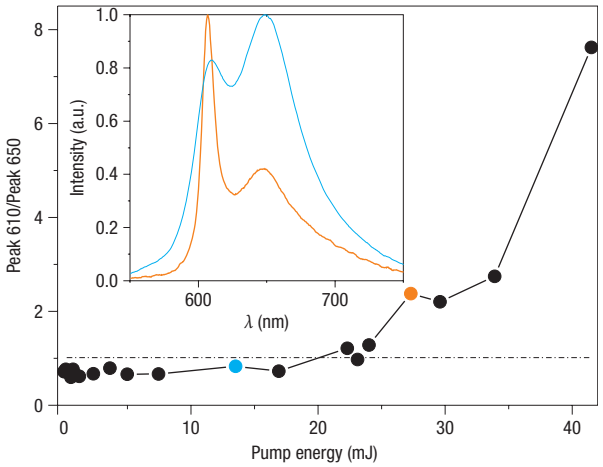

Figure 4 Mode competition between Mie resonances. Intensity ratio for the two lasing peaks of a photonic glass ($d = 1.22$ μm) doped with two laser dyes. The highest energy peak starts to dominate above threshold (20 mJ), reaching a value eight times larger than the low-energy one. The inset shows the emission spectra below (blue) and above (orange) threshold.

the lasing curve for photonic glasses. It is possible, this way, to select specific modes of the random laser and study mode-coupling mechanisms. In the experiments the laser dye was always distributed on the surface of the spheres. The random laser action could be even more pronounced and the lasing threshold lower if the dye was placed inside the spheres, close to the maximum of the resonant internal mode. This work shows that it is possible to control spectrally both light diffusion and random lasing emission, opening a novel route to active disorder based photonic devices.

METHODS

Polystyrene spheres were synthesized by free emulsion polymerization[28] with different diameters (0.2, 0.9, 1.0 and 1.2 μm) with a polydispersity around 2% measured with transmission electron microscopy. The surface potential of the polystyrene spheres ($d = 1.2$ μm) was −39.8 mV, established from a measurement of their electrophoretic mobility. In order to provoke the flocculation of the colloidal suspension, a 1 per cent volume of 0.01 M aqueous suspension of $CaCl_2$ or HCl had to be added to the initial polymeric colloidal suspension. The total solution was stirred under ultrasound for 5 min to force the flocculation of the spheres. To grow the photonic glass in a clean microscope glass slide previously hydrophilized, a volume of the charged colloidal suspension with a concentration 2 vol% was confined in a circular area (typically 1 cm radius) by applying adhesive tape of known thickness (millimetres). The system was kept at 50 °C for 3 h to evaporate the water. The filling fraction of the samples could be estimated as 0.55 by precisely weighting different samples with very well known geometry.

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