Mie resonances to tailor random lasers

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In this paper, we present an optical characterization of photonic glass-based random lasers. We show how the resonant behavior of diffuse light transport through such systems can tailor the lasing emission when a gain medium is added to the glass. A DNA-based organic dye is used as gain medium. The resonances in the transport mean-free path influence the lasing wavelength of the random laser. The laser wavelength is therefore controlled by the sphere diameter. Furthermore, the existence of Mie resonances reduces the necessary pump energy to reach the lasing threshold.

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I. INTRODUCTION

Among all the possible applications of disordered nanophotonics, random lasing [1] is one of the most intriguing and fascinating. Proposed by Letokhov [2] in the 1960s and experimentally observed now in various materials [3], a random laser is basically the combination of a random assembly of elastic scatterers and a gain medium. In such a system, light is multiply scattered and amplified. In standard lasers, light scattering hampers the functionalities and is generally regarded as a parameter to minimize. In these systems, gain saturation is obtained by (resonant) feedback provided by an optical cavity, therefore, scattering induces light losses and increases the lasing threshold. On the contrary, in a random laser, multiple scattering of light is the *sine qua non* condition to reach threshold and lase. It serves to have gain larger than losses and reach gain saturation, which is the fundamental mechanism that leads to coherence [3]. The lasing emission is determined by the interplay between losses and gain. Unlike ordinary lasers, the resulting light emission is multidirectional and not really monochromatic, but the threshold behavior [4], the photon statistics [5,6], and relaxation oscillations [7,8] are very similar to those of standard lasers. This phenomenon is usually observed in random matrices composed by very polydisperse scatterers in shape and size such as colloidal suspensions of oxides powders [5], powdered laser crystals [9], ceramics [10], organic composites [11], and even biological tissue [12]. In all those systems, the scattering parameters, the transport mean-free path or the diffusion constant, which define the diffusive transport, are monotonically dependent on the light wavelength. Thus, the lasing output is selected *a priori* by the gain and loss properties.

In this work, it will be shown how to control light diffusion with the optical resonances of the monodisperse scatterers composing the system. The resonant behavior in the diffuse light through a photonic glass allows controlling the laser emission via the diameter of the spheres and their refractive index. Such a system is a dispersive random device with *a priori* design lasing peak within the gain curve. As a complement to our previous work [13], where the effect on the spectral position of the lasing modes was pointed out, here we show how the threshold value of the random laser also strongly depends on the existence of resonances in the diffusion parameters. This paper is organized as follows. A brief introduction to multiple scattering in a gain medium will be provided in Sec. II. In Sec. III, the gain medium used in this work will be presented. Sections IV and V will deal with conventional and resonant random lasers to emphasize its differences. Finally, Sec. VI will summarize the lasing threshold dependence on Mie resonances.

II. MULTIPLE SCATTERING IN A GAIN MEDIUM

The necessary condition for a random laser is that the material multiply scatters light, which means that the transport mean-free path (the average distance over which the scattered light direction is randomized) is much smaller than L, which is the sample size, $\ell_t \ll L$ [14,15]. The other fundamental quantity is the gain length ℓ_g that 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 $L_{cr}=\pi\sqrt{\ell_g \ell_t/3}$ [14]. Light transport through a multiple-scattering medium can, eventually, be described as a diffusion process characterized by a (transport) mean-free path, ℓ_t, and a diffusion constant, $\mathcal{D}$. Due to the relative low refractive index of the polymer scatterers, diffusion approximation can be accurately applied in our case, as the light propagation regime in our system is far from strong light localization [15]. Diffuse light propagates though such a medium following a diffusion equation as follows:

$$\left[\frac{\partial}{\partial t} - \mathcal{D}\nabla^2 + \frac{v_e}{\ell_i}\right] I(\mathbf{r},t) = S(\mathbf{r},t), \tag{1}$$

where $S(\mathbf{r},t)$ is the light source and v_e is the energy velocity [16,17]. If one solves Eq. (1) for a plane wave incident on a slab of random system, the total transmission is expressed by the Ohm's photonic law [18]

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$$T(L,\lambda) = \frac{1}{\alpha z_e}\frac{\sinh[\alpha(z_p+z_e)]\sinh[\ \alpha z_e)]}{\sinh[\alpha(L+2z_e)]},$$

$$z_e = \frac{1}{2\alpha}\ln\left[\frac{1+\alpha z_0}{1-\alpha z_0}\right],$$

$$z_0 = \frac{2}{3}\ell_t(\lambda)\left(\frac{1+R}{1-R}\right). \quad (2)$$

In the solution, $\alpha = 1/\ell_i$ is the inverse absorption length and R is the polarization- and angular-averaged reflectivity of the boundaries [19]. Therefore, a measurement of $T(L,\lambda)$ is a direct probe of the spectral dependence of ℓ_t. Optical amplification can be added to the diffusion picture described so far. Multiple scattering increases the time light remains inside the sample and when the total gain is larger than the losses, the intensity increases until the gain medium saturates, resulting in coherent laser emission. This lasing process can be described, to a first approximation, with the four-level rate equation [20] in the presence of diffusion

$$\frac{dN_1(t)}{dt} = P(t) - \frac{\beta q(t) N_1(t)}{\tau} - \frac{N_1(t)}{\tau},$$

$$\frac{dq(t)}{dt} = \frac{\beta N_1(t)}{\tau}[q(t)+1] - \frac{q(t)}{\tau_c},$$

$$\tau_c = \frac{L^2}{8\mathcal{D}(\lambda)}, \quad (3)$$

where $N_1(t)$ is the number of excited molecules, $P(t)$ is the pump rate, $q(t)$ the number of photons of the laser modes, and τ the spontaneous emission lifetime of the gain medium. The differences between a single mode standard laser and a multimode random laser lie in the cavity decay time τ_c [21,22] that becomes dependent on the diffusion constant (and then on the transport mean-free path) and in the β factor [23] that instead accounts for the average numbers of lasing modes. From Eq. (3) it can be seen that the threshold and lasing process can be controlled modifying $D(\lambda)$ and $\ell_t(\lambda)$.

III. GAIN MEDIUM

A crucial factor in realizing a random laser is the gain medium—the medium which provides light emission and amplification. Most of the materials used to that purpose can be described as a three of four electronic level systems. When the material is optically excited, electrons promote to some high-energy level and decay in a very short time interval to a metastable state. Laser transitions occur between this metastable state and the ground state. Usually, materials which can be optically pumped are considered. Among those gain materials, it has been used mainly semiconductors with photoluminescence properties as ZnO [5] or organic molecules, organic *dyes* [4]. Photoluminescence semiconductors usually present a high refractive index contrast and are used also as the scattering material, as in the case of ZnO. Organic dyes, however, present a low refractive index and should be embedded in a multiple-scattering system. These organic materials present high quantum efficiencies and a wide Stokes shift, which together with their versatility and small size (organic chains with few nanometers) make them very good candidates to be laser materials in a random medium. Among them, the 4-dicyanomethylene-2-methyl-6-p-dimethylaminostyryl-4*H*-pyran (DCM) dye molecule is known as one of the best laser dyes. However, dye molecules present a large quantum efficiency when in dissolution but are affected by quenching and damaging when dry. For random lasing from nanostructured media, this is a hampering factor since, in order to maximize the refractive index contrast, the systems are usually solid in air. The presence of a solvent usually attenuates the multiple scattering of light and makes random lasing hard to achieve. To solve this issue, organic dyes are usually embedded in the scattering medium in solid phase. This highly disturbs the luminescence functionalities of the dyes which easily suffer from quenching due to the proximity of the fluorescent molecules and thermal dissociation and damage due to the high pump energies. In order to avoid these problems, we embedded organic dyes in transparent deoxyribonucleic acid (DNA) helices. We synthetize a gain medium from DCM molecules intercalated into the helices of the biopolymer DNA [24]. This process, which is schematically illustrated in Fig. 1, prevents the quenching of the dye but also provides a way to dissipate the extra heat, accumulated upon optical pumping, to the DNA strands. In such a nanodesigned gain medium, the intensity of the fluorescence is greatly enhanced (highly efficient amplified spontaneous emission has been reported for this dye configurations [25]) and no dye damage occurs for a wide pump energy range. In this work we will use DCM intercalated into salmon DNA helices or strands, DCM@DNA, as gain medium [26].

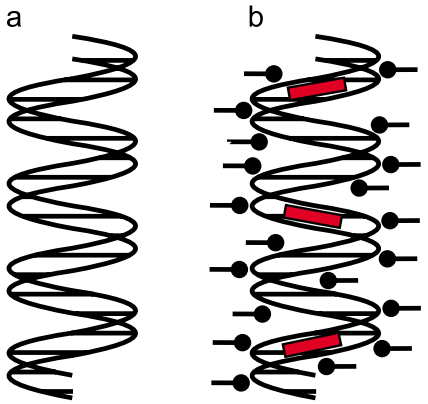

FIG. 1. (Color online) Schematic diagram for the gain medium. (a) Representation of DNA helices. (b) Inclusion of DCM organic dye into the helices of the DNA.

IV. RANDOM LASING

A random system, composed by particles with arbitrary shape and size, has a transport mean-free path that is spectrally flat (nonresonant) at least for $\Delta\lambda \sim 100$ nm. Under this condition, conventional random lasing occurs at a wave-

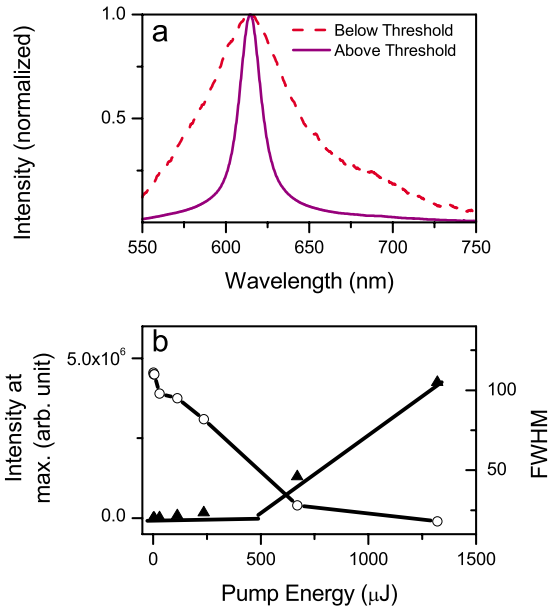

FIG. 2. (Color online) (a) Emission intensities below (dashed curve) and above (continuous curve) the threshold in the case of DCM@DNA embedded in a TiO_2 matrix. The lasing mode is defined by the gain maximum. (b) Emission intensity and its full width at half maximum (FWHM) as a function of pumping energy. A clear threshold appears at a 500 μJ per pulse. Pumping spot size is 2 mm.

length where $\ell_g(\lambda)$ has its minimum, i.e., at the maximum of the gain curve.

Figure 2 shows conventional random lasing from the organic emitter, DCM@DNA, shown in previous section and embedded in a TiO_2 powder matrix. This system is optically pumped by a frequency-doubled Q-switched pulsed Nd:yttrium aluminum garnet (YAG) laser (9 ns pulse duration), with 10 Hz repetition rate. The spot size was fixed at 2 mm diameter to access a large gain volume (much larger than $\ell_t < 2$ μm). The diffuse emission was monitored with a miniature spectrometer with a resolution of 0.5 nm. The scattering strength of such a system is relatively high due to the high refractive index contrast between TiO_2 particles ($n=2.5$) and air ($n=1$), where n is the refractive index. Figure 2(a) shows the normalized output intensity below (red curve) and above threshold (violet curve). As the pump power increases, the emission peak becomes narrower due to the preferential amplification of the frequencies close to the maximum of the gain curve. In such a system, the lasing modes are selected from those at the spectral position of the minimum gain length, ℓ_g. The amount of DCM@DNA was 0.02 wt % for all samples considered here, of which 1% was DCM. Even with such a low concentration of organic emitter, 2×10^{-4} wt %, it is possible to achieve a lasing threshold in the output intensity at 500 μJ, which corresponds to $\sim$60 $\mu J/mm^2$ per pulse [Fig. 2(b)].

As previously mentioned, the wavelength tunability is a great challenge in every lasing device, in particular it is an issue in random lasers. Since gain is fixed and determined by the emitter chemical properties, tunability can only be achieved by tailoring the lasing modes. Our strategy is to exploit the ability of the monodisperse random media to sustain electromagnetic resonances. The idea is to exploit resonances in scattering coefficients [13], called Mie resonances [27], present when the sizes of the scatterers are comparable to the wavelength of the incident light. These resonances are, in principle, also present in the single-scattering properties of the TiO_2 microparticles which compose the powder, but due to the different shape and size of each particle, the resonances are washed out on a macroscopic scale [28]. On the contrary, in a system composed of monodisperse spheres, each scatterer sustains the same resonance at the same energy, giving rise to a macroscopic resonant behavior. A candidate for that purpose is the so-called *photonic glass* [29]; static and dynamic measurements of light diffusion in such systems have shown matching resonances for $\ell_t(\lambda)$ and the diffusion constant of light $\mathcal{D}(\lambda)$ [30,31].

V. RESONANT RANDOM LASING

In a photonic glass, the lasing wavelength becomes very sensitive to the diameter of the constituent spheres and follows the resonances of the system, as will be experimentally shown. To that purpose, a set of photonic glasses composed by spheres with different diameters was used as the basis of an amplifying system completed by embedding organic laser dye. In a first work, the dye was inserted by liquid infiltration and subsequent solvent evaporation [13], while here we use DCM@DNA as a gain medium.

The emitter embedded in the different photonic glasses, DCM special couple into the DNA strands, is the same as the one used in the case of TiO_2 powder in Fig. 2, with the same concentration. DNA hosts charge ions in certain strand positions. The whole emitter has, therefore, a net charge due to this fact, about 3×10^{-6} M charge concentration. This extra amount of charge, when added to the colloidal suspension of PS microspheres, is enough to force the flocculation of the colloidal particles [29]. Upon evaporation of the solvent, a solid photonic glass based on PS spheres coated by few nm of a DCM@DNA gel is realized.

The optical pumping and the diffuse emission were monitored as in the case of TiO_2-based random laser. The multiple light-scattering condition was ensured by the sample thickness $L>100$ μm, which is much larger than the typical mean-free path in such photonic glasses, $\ell_t \cong 2-3$ μm, for visible light [30,31]. Figure 3 shows in more detail how the resonances sustained by the scatterers affect lasing action. As a reference, we consider a photonic glass of very small spheres with $d=0.2$ μm [Fig. 3(a), light blue curve] as well as the polydisperse TiO_2-based random laser (violet curve). Both systems, for different reasons, do not show resonances in the static or dynamic measurements [30,31]. In both these cases, lasing occurs nearly at the same wavelength, close to the maximum of the gain curve. In the same figure [Fig. 3(a)], instead, one can observe a controlled overall shift of the lasing wavelength of about 35 nm between different photonic glasses composed with spheres with diameters $d=0.9$ μm (yellow curve) and $d=1.0$ μm (orange curve). Lasing modes from both systems are shifted with respect to the gain maximum which can be explained in terms of Mie resonances.

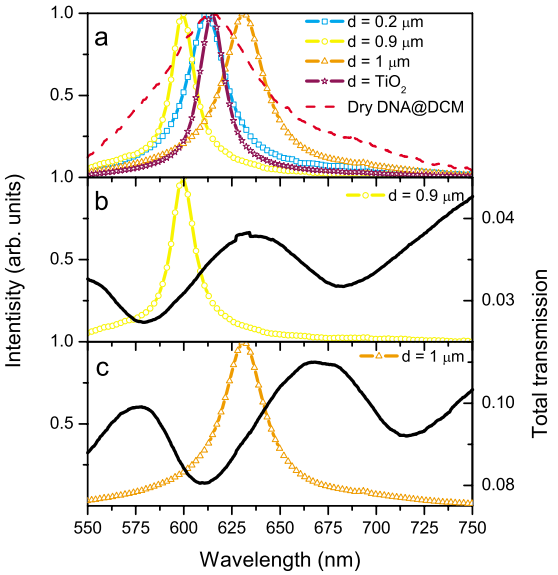

FIG. 3. (Color online) (a) Random laser emission from photonic glasses with different sphere diameter compared to the pure dry dye fluorescence and a reference sample made with TiO_2 powder doped with DCM@DNA. The pump energy for every sample is around 10 mJ. [(b) and (c)] Emission intensity and total transmission for photonic glasses, respectively, with $d=0.9$ μm and $d=1.0$ μm. Lasing occurs close to the transmission minimum.

Figures 3(b) and 3(c) compare the lasing wavelength dependence to the sample total transmission. For these two photonic glasses, the Mie resonances are pronounced and in phase opposition, even if the difference in the diameter is only ~ 0.1 μm. The minimum in transmission corresponds to a minimum both in ℓ_t and $\mathcal{D}$ and with a maximum in the scattering strength. Light at this wavelength populates a Mie mode in the structure and dwells longer in the active media, enhancing the light-dye interaction and therefore the lasing probability. According to that, the system tends to lase where the scattering strength is maximum, even far from the gain maximum. The limited gain puts a limit on the wavelength shift induced by the scattering resonance, which is of 35 nm. The shift is enough to be observed by naked eye. Figure 4 shows the images corresponding to the emission of both systems above threshold which are distinguishable by naked eye by the color of the lasing emission.

To prove our concept and to confirm that indeed the transport resonances determine heavily the lasing wavelength, the spheres of the photonic glass with the gain medium DCM@DNA are dispersed in ethanol solution (spheres concentration 5 vol %). In such a way, the refractive index contrast and, therefore, the Mie resonances are strongly damped. In previous works [14,21], colloidal suspensions of spheres of particles mixed with the laser dye have been used for random lasing. This makes the contrast so low that the Mie resonances nearly disappear. Figure 5(a) shows such an effect. The straight curve represents total light transmission for a photonic glass composed by spheres with $d=1.0$ μm, while the dashed curve represents the total light transmission through the colloidal suspension of the same spheres in ethanol. The figure clearly reveals that the resonances are strongly damped. The corresponding emission spectra are plotted in Fig. 5(b), where the colors represent the same sphere diameters as for the samples in Fig. 3(a). Figure 5 points out the fact that by damping the Mie resonances of the system, the tunability of the lasing emission also disappears as expected.

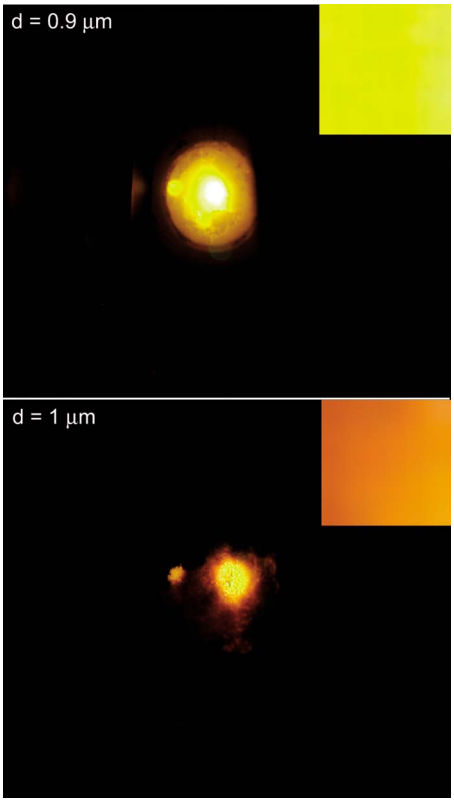

FIG. 4. (Color online) Real image of the lasing mode from $d=0.9$ μm and $d=1.0$ μm photonic glasses doped with DCM@DNA with a pump energy around 10 mJ in both cases. The spectral difference between both lasing emissions (~ 30 nm) is distinguishable by naked eye.

VI. THRESHOLD DEPENDENCE ON MIE RESONANCES

The existence of scattering resonances in the system not only affects the lasing mode tunability but also the threshold value of the photonic glass-based random laser. In general, scattering resonances enhance lasing action and may, eventually, lower the lasing threshold [32,33]. The presence of a maximum in the scattering strength and, correspondingly, a minimum in the transport parameters as ℓ_t and $\mathcal{D}$ corresponds to a maximization of the light scattering. For certain wavelengths for which the multiple scattering of light is optimized, the pump energy necessary to achieve gain larger than losses is minimized and, thus, the lasing becomes easier.

Figure 6 compares the threshold for the photonic glasses used in this work. All the samples measured here were pumped under the same conditions. The threshold value for four different systems is shown in Fig. 6; all these systems are composed with spheres of the same material, PS, but with different diameters and polydispersity. Apart from the

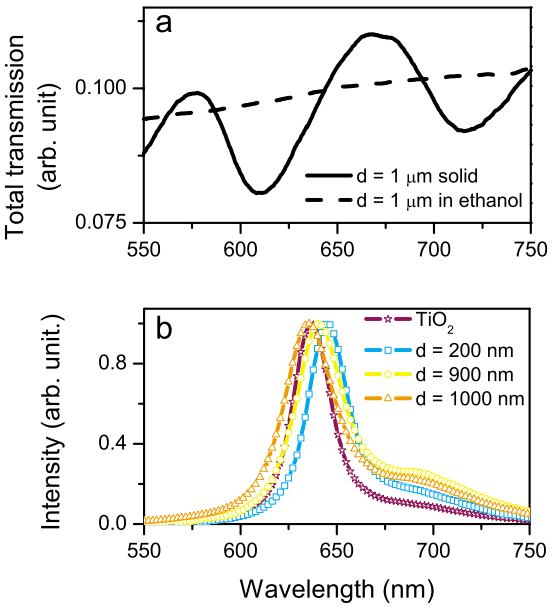

FIG. 5. (Color online) (a) Total integrated transmission for a photonic glass with $d=1$ μm (continuous curve) and from a colloidal suspension of the same spheres (5 vol % of concentration) in ethanol (dashed curve). The refractive index of the host medium increases, respectively, from 1 (air) to 1.36 (ethanol). (b) Random laser emission for different spheres suspension in ethanol with DCM@DNA as optical active medium, with an external pump energy fixed at 10 mJ. In this case, the total spectral separation of the emission maxima for the different suspensions is only 7 nm.

diameter, a very important parameter to take into account is the monodispersity of the scatterers. A high polydispersity value washes out the macroscopic resonant response [31]. In this measurement, we have used three photonic glasses composed by very monodisperse microspheres ($<2\%$) and one with a high polydispersity (represented by the bar). This last sample is grown by mixing spheres of the same material with slightly different diameters. We intentionally introduce the polydispersity in the system to obtain a very polydisperse reference sample composed by the same material as the other photonic glasses. The diameter of the spheres is normalized to the lasing wavelength emission (λ_{lasing}) and the polydispersity is represented by the bar on the diameter variable. In the particular case of the photonic glass composed by spheres with $d=200$ nm ($d/\lambda_{lasing}=0.3$), no resonances can be sustained in the visible due to the small size of the spheres. Therefore, those spheres behave, upon this particular light energy illumination, almost as pointlike scatters giving rise to Rayleigh scattering (where scatterer structure is negligible) instead of Mie scattering (where resonances are featured). Resonances are expected in other energy ranges (in the UV) for this particular small sphere diameter. With these considerations, it is easily observed how random lasing has a lower threshold for those photonic glasses where Mie resonances are present close to the gain curve, as in the case of $d=0.9$ μm ($d/\lambda_{lasing}=1.45$) and $d=1.0$ μm ($d/\lambda_{lasing}=1.6$). These photonic glass-based random lasers need about half the pumping energy than the two reference samples: the photonic glass composed by small microspheres (with $d=0.2$ μm) and a very polydisperse arrangement of the same kind of spheres with a main diameter about $d=0.8$ μm ($d/\lambda_{lasing}=1.3$). It is also important to mention that these two reference samples, due to the lack of scattering resonances, lase at the same spectral position of the random lasing emission of TiO_2 conventional random laser. Their emission is controlled only by the gain curve and their threshold is higher than Mie resonances-based random lasing.

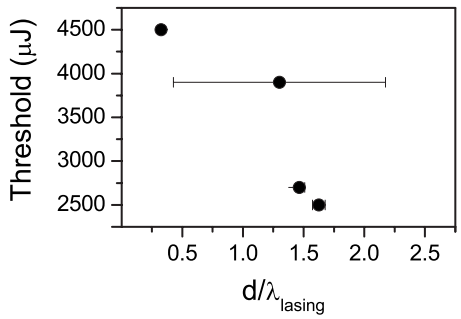

FIG. 6. Threshold for various photonic glasses: a standard sample with $d=0.2$ μm ($d/\lambda_{lasing}=0.3$) which does not sustain Mie resonances in the visible, two photonic glasses with $d=0.9$ μm ($d/\lambda_{lasing}=1.45$) and $d=1.0$ μm ($d/\lambda_{lasing}=1.6$) which sustain Mie resonances within the gain curve and a polydisperse arrangement ($\sim 42\%$) of PS spheres for which a threshold appears at 3.9 mJ. This plot points out how the existence of scattering resonances for light within the gain spectral range reduces the pump energy necessary to achieve gain larger than losses.

VII. CONCLUSIONS

In this work, random laser action from a three-dimensional system with resonant transport features has been presented. The experiments shown here demonstrate that Mie resonances of photonic glasses can strongly effect random lasing and determine both the lasing emission wavelength and threshold. The concurrence of disorder with scattering strength, gain, and monodispersity is what allows these novel functionalities. The high contrast of the dielectric spheres in air secures strong light-matter interaction, the dye hosted in DNA helices leads to amplification, finally the resonances spectrally select the modes and minimize the lasing threshold of the laser. This work shows that it is possible to control spectrally light diffusion and use it for self-tuning of random lasing, opening a unique route to active-disorder-based photonic devices [34].

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