Quasi-Two-Dimensional Diffusive Random Laser Action

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We report on random lasing in a disordered system in which the multiple scattering feedback mechanism can be switched from a three-dimensional random walk to a quasi-two-dimensional type of transport. The emission from this system is anisotropic, extraordinary polarized, and is controlled via an external electric field. The phenomenon is observed in dye-doped polymer dispersed liquid crystals and makes use of the strong scattering anisotropies in these materials.

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If optical gain is added to a diffusive material [1], a unique light source is obtained, now often referred to as a random laser. Random amplifying media can be realized in several ways. For example, one can powder a laser crystal [2], dissolve laser dye in a particle suspension [3], or use a powdered semiconductor such as zinc oxide [4]. The emission properties of random lasers show similarities with a regular laser. The system exhibits a threshold behavior, and when the gain overcomes the losses, the emitted spectrum shows band narrowing and laser spiking [5]. In addition, random lasers can exhibit ultranarrow emission modes [4,6]. The latter can be due to interference effects, such as Anderson localization that provide coherent feedback [4], or due to exponential gain in extended modes in systems with diffusive feedback [6]. Anderson localization effects are expected to play an important role in lower dimensional systems [7]. Even in the absence of interference effects, the photon statistics of a random laser can be Poissonian, indicating a high degree of coherence [8]. Interesting lasing effects have been observed also in related complex systems, such as dye-doped cholesteric liquid crystals [9] and amplifying (organic/semiconductor) films [10,11].

Random lasers use multiple scattering of light as a feedback mechanism to achieve gain that exceeds losses and thereby obtain lasing. This means that if one can control the multiple scattering properties of a sample, one can influence the laser threshold and control its emission properties. Nematic liquid crystals are potentially very suitable for this purpose due to their strong temperature dependent refractive index and coupling with magnetic and electric fields. The nematic phase of a liquid crystal is characterized by an alignment of the molecules along a common axis, known as the nematic director, and an otherwise translational disorder. When heated into the isotropic phase, the liquid crystal behaves as a regular liquid. Nematic liquid crystals have been shown to exhibit anisotropic multiple scattering properties [12] that can even survive interference effects such as weak localization of light [13]. Busch and John proposed to use these liquid crystal properties to tune the density of states of a photonic crystal [14]. In previous experiments, we applied this temperature dependence to tune the light diffusion constant of porous glass [15]. This allowed us to obtain temperature tunable random lasing [16] which could be described theoretically through Monte Carlo simulations [17].

Polymer dispersed liquid crystals (PDLCs) are widely applied in optical devices for their excellent tuning possibilities and extreme form of anisotropic scattering [18]. A PDLC consists of submicron- to micron-sized droplets of liquid crystal dispersed in a polymer matrix and is strongly scattering because of a refractive index mismatch between the polymer matrix and the liquid crystal. When an electric field is applied, the nematic director of the liquid crystal aligns along the field direction and the material appears transparent.

In this Letter, we report on diffusive random laser action in PDLCs, where the light diffusion constant can be controlled over many orders of magnitude through an electric field. We observe a counterintuitive switching behavior: upon applying the electric field, the sample becomes transparent, yet the random laser effect becomes more pronounced as though multiple scattering were increased. The random laser output becomes anisotropic and extraordinary polarized. These observations can be explained by the extreme scattering anisotropies in these systems that lead to random lasing based on a quasi-two-dimensional type of optical transport.

PDLCs are normally used as thin films in the single scattering regime, but one can easily obtain multiple scattering PDLCs by synthesizing very thick samples. Our samples were prepared using photopolymerization. The concentration of the liquid crystal was 85 wt % and that of the polymer was 15 wt %. We chose a concentration of 1 mmol/l of laser dye over the whole sample. The PDLC was prepared using two types of liquid crystals, p-pentyl-p'-cyanobiphenyl (5CB) and E7 (manufactured by Merck), which have similar values for the ordinary (n_o) and the extraordinary (n_e) refractive indices (for E7: $n_o = 1.53$ and $n_e = 1.75$ and for the polymer $n_p = 1.52$). The temperature range of the nematic phase is different

for these two liquid crystals, with E7 having the widest nematic temperature range (from $-10\,^{\circ}$C to $60\,^{\circ}$C), whereas 5CB is nematic between $24\,^{\circ}$C and $33\,^{\circ}$C. The sample had a slab geometry of dimensions 3×2 cm and 1 mm thickness, and was thereby about 2 orders of magnitude thicker than common PDLC systems.

We controlled the scattering strength of our sample by applying an external electric field (maximum field 120 kV/cm at 500 Hz). Without the field, the liquid crystal droplets are randomly aligned and the system strongly scatters light. In the diffusion approximation this multiple scattering process is characterized by a diffusion constant D. This diffusion constant can be measured in a time-resolved experiment in which the transmitted diffuse intensity is monitored with a streak camera [15]. This allowed us to measure the diffusion constant in the direction perpendicular to the sample plane (defined as the z direction), which we have reported in function of the applied electric field in Fig. 1. The measurements show a doubling of the diffusion constant when the field reaches 80 kV/cm. Above this value the diffusion constant diverges and the limit of the time resolution of the streak camera is reached. The electric field aligns the optical axis of liquid crystal droplets in the z direction. A light wave propagating in the z direction therefore experiences the ordinary refractive index of the liquid crystal for any polarization vector and is almost not scattered.

The time-integrated transmission coefficient versus the applied field is reported in the inset of Fig. 1. The strong increase of the transmission at the highest field strength confirms that our samples can reach high transparency levels, despite their unusual large thickness. In the inset, we also show the residual transmission at high temperature (62 °C) when the liquid crystal (E7) is in the isotropic phase (black circles). Also, at high temperatures the sample appears almost transparent due to a near refractive index matching in all polarization channels. This suggests that the random lasing in these materials should respond in a similar way to high electric fields and to high temperatures. Nevertheless, we will see below that the random laser behavior is opposite.

To characterize the random laser action in our PDLC samples, we have performed measurements on their emission spectra under various conditions. The excitation source was a frequency doubled Q-switched Nd:YAG laser, with 10 Hz repetition rate. The diffuse emission was monitored with a spectrometer with a resolution of 0.5 nm. The bandwidth of the emission in function of the excitation energy is plotted in Fig. 2 for three cases: the isotropic case ($T = 37\,^{\circ}$C, black triangles), the nematic phase without electric field (0 kV/cm, black circles), and for the case of maximum field (120 kV/cm, open squares). For the measurements in the isotropic phase, we substituted the liquid crystal E7 by 5CB, which already becomes isotropic at 33 °C, to avoid high temperatures that lead to degradation of the laser dye. In this case,

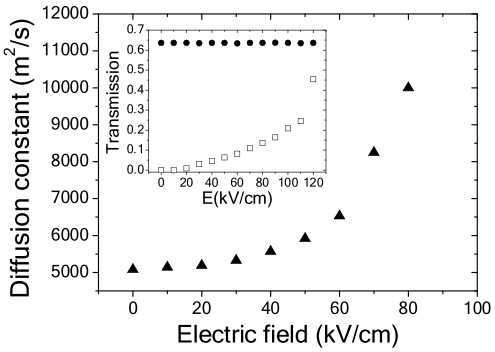

FIG. 1. The diffusion constant in function of the applied electric field measured with a time-resolved technique. In our setup a short (1 ps) laser pulse (Ti:sapphire laser, $\lambda = 750$ nm) is incident on the sample, and the diffuse transmitted light is collected by a streak camera, with a resolution of 3.5 ps. The inset shows how the time-integrated transmission coefficient changes with the electric field, both for the nematic (open squares) and the isotropic phase (black circles).

we observe almost no narrowing upon increasing the excitation energy, which is consistent with the absence of multiple light scattering: the system remains below threshold. For the nematic case without field, we have strong scattering and we observe a significant narrowing of the emission since the diffusive feedback mechanism allows the random laser to go above threshold. This difference between the isotropic and the nematic phase is the known temperature tuning of a random laser [16,17], due to the temperature dependence of the scattering strength [15].

Contrary to the temperature tuning case, we observe in Fig. 2 that the random laser effect becomes stronger upon application of the maximum electric field. The bandwidth at 10 mJ excitation energy decreases from 30 nm at zero field to 20 nm at 120 kV/cm. Although the sample appears nearly transparent at 120 kV/cm, the random laser

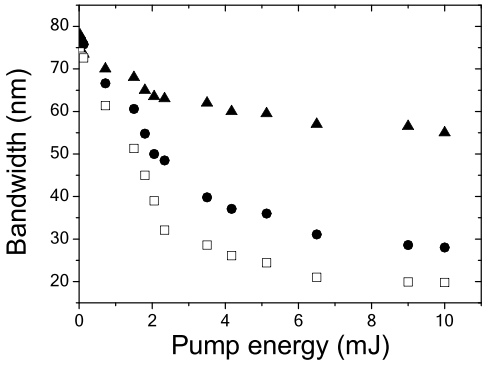

FIG. 2. Bandwidth of the emission spectra in function of the excitation energy. In the isotropic case (black triangles) ($T = 37\,^{\circ}$C, liquid crystal 5CB) the sample is transparent in all directions (no multiple scattering). The other two curves refer to the PDLC in the nematic phase; black circles for $F = 0$ kV/cm and open squares for $F = 120$ kV/cm.

effect increases. This indicates that strong multiple scattering takes place somewhere inside the sample, which is not visible in the z direction.

The explanation lies in the extreme scattering anisotropy of the PDLC, which results in a strongly anisotropic diffusion constant D. With the nematic director of the liquid crystal droplets aligned in the z direction, any light wave propagating in z will experience the ordinary refractive index of the liquid crystal which is nearly index matched to that of the polymer host ($n_o \cong n_p$), and hence is hardly scattered. On the other hand, however, a wave propagating in the x-y plane of the sample with polarization vector parallel to the director probes the extraordinary refractive index of the liquid crystal droplet ($n_e/n_p \cong 1.15$) and is strongly scattered. The reason that this scattered light is not observed along the z axis is that the angular distribution of the scattered light is peaked in the x-y plane. This scattering anisotropy is consistent with the reciprocity condition imposed by Maxwell's equations: light that is incident in the z direction is not scattered and, vice versa, the light that is multiple scattered in the x-y plane is not scattered in the z direction. The measurement in Fig. 1 reveals only the z component of the diffusion constant and is not sensitive to the value of D in the sample plane.

The differential scattering cross sections of aligned liquid crystal droplets dispersed in a host medium have been calculated by, amongst others, Zumer and Doane [19]. In the Rayleigh-Gans approximation, we have

$$\frac{d\sigma/d\Omega}{\Gamma^4\sigma_0} = \frac{u^2}{9\pi}[\cos^2\alpha\cos^2\delta(\zeta+2\eta)^2 + \sin^2\alpha(\zeta+1)^2]. \quad (1)$$

Here $\frac{d\sigma/d\Omega}{\Gamma^4\sigma_0}$ is the total differential cross section normalized by the geometrical cross section σ_0 and $\Gamma = k_i R$, where R is the radius of the droplet and k_i the incident light wave vector. The zoom of the PDLC droplets in Fig. 3(b) shows the typical scattering geometry, where $\mathbf{k}_s$ is the scattered wave vector ($|\mathbf{k}_s| = |\mathbf{k}_i|$), δ is the angle between $\mathbf{k}_s$ and the plane of incidence (defined by $\mathbf{k}_i$ and the nematic director), and α is the incoming polarization angle, defined as the angle between the polarization vector $\mathbf{E}_i$ and the scattering plane (defined by $\mathbf{k}_i$ and $\mathbf{k}_s$). For simplicity, Eq. (1) is referred to the case where the plane of incidence coincides with the scattering plane, and the incoming wave vector is orthogonal to the nematic director. Finally, ζ and η are functions of the refractive indices of the liquid crystal and the polymer, while u depends also on δ:

$$\zeta = \frac{1}{3n_p^2}(2n_o^2 + n_e^2) - 1, \qquad \eta = \frac{1}{3n_p^2}(n_e^2 - n_o^2), \quad (2)$$

$$u = \frac{3\sin[2\Gamma\sin(\delta/2)] - 2\Gamma\sin(\delta/2)\cos[2\Gamma\sin(\delta/2)]}{[2\Gamma\sin(\delta/2)]^3}. \quad (3)$$

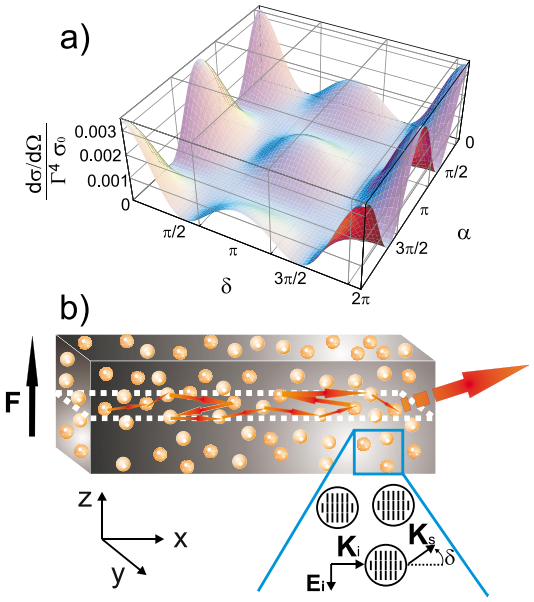

FIG. 3 (color). (a) Scattering cross section of an aligned nematic droplet in function of the scattering angle δ and the incoming polarization angle α. (b) Simplified representation of the random laser action in an active PDLC system, with a strong electric field F applied in the z direction. Multiple light scattering takes place for the extraordinary polarization channel and is strongly peaked in the x-y plane, resulting in a quasi-2D type of light transport.

A 3D plot of the differential cross section as function of δ and α is depicted in Fig. 3(a). We see that for $\alpha = 0$ (extraordinary polarization) the cross section has maxima at $\delta = 0$ and $\delta = \pi$. In addition, the probability for an ordinary ray ($\alpha = \pi/2$) to be scattered is almost zero. Changing the angle between the scattering and the incidence plane, one finds that the angular distribution of the scattered light remains peaked in directions in the x-y plane and nearly zero in the z direction. Also if one recalculates the differential cross section of Eq. (1) for other angles of incidence, one finds that the scattered light remains peaked in the x-y plane. This provides an efficient multiple scattering mechanism and basically confines the light to an anisotropic random walk in the sample plane. Because of internal reflection at the sample interface, all wave vectors under an angle between $-48°$ and $48°$ with respect to the x-y plane will be trapped inside the sample. If we integrate the complete differential cross section of the liquid crystal droplets over the corresponding range of angles, we obtain that this amounts to $>93\%$ of the light at each scattering event. A maximum of $<7\%$ of the light can therefore escape out of the sample plane at each scattering event, which determines the system loss factor.

The resulting transport is schematically depicted in Fig. 3(b), where a random walk of extraordinary light is shown for the high electric field case. A spontaneously emitted photon with extraordinary polarization and appropriate wave vector will perform a very anisotropic

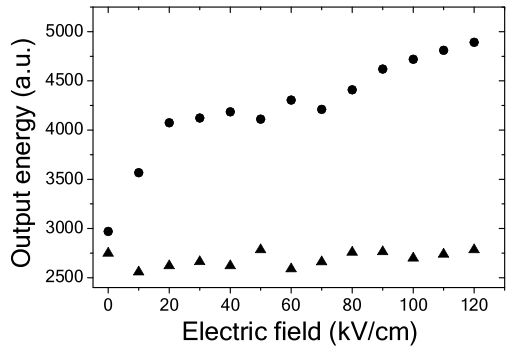

FIG. 4. Output energy of the emission in the x direction for the two polarization channels (circles: extraordinary polarization; triangles: ordinary polarization), in function of the applied field. The excitation energy is 3.5 mJ. The emitted light becomes gradually extraordinarily polarized when the field is increased.

random walk which retains the light long enough inside the samples for gain to become larger than losses. The electric field allows one to switch from a standard 3D diffusive process to an extraordinary quasi-2D type of transport. This means that, if we collect the light emitted along the x or y axis at high fields, we expect to measure extraordinary polarized emission. Figure 4 shows the spectrally integrated output energy in the x direction in function of the applied field for the two polarization channels, extraordinary (parallel to z axis) and ordinary (parallel to y axis). The extraordinary channel grows with the field, and a polarization ratio of two is reached at maximum field. This means that the PDLC random laser emits polarized light with controllable degree of polarization. The increase of the total intensity (sum of both polarization channels) is consistent with the increase of excitation efficiency caused by the transparency of the sample in the pump direction. The strong anisotropy provides a clear advantage for efficient optical excitation, since the pump beam is not scattered and hence can be absorbed for almost 100%. In regular 3D random lasers a maximum pump efficiency of typically 30% is obtained, which is limited by the strong scattering losses of the excitation beam [2]. These scattering losses are absent in the PDLC at high electric field. Polarized emission was visible only in the x-y plane and not in the z direction. Moreover, the emitted light is observed to be spatially anisotropic. We have measured that at maximum field $F = 120$ kV/cm the total emission in the x direction increases by 60% with respect to $F = 0$ kV/cm, while in the z direction the total emission increases only by 16%.

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