The physics and applications of random lasers

Recent developments in the field of micro and nanophotonics have shown that it is possible to make use of the intrinsic disorder in photonic materials to create useful optical structures. An example is that of a random laser, in which laser action is obtained in disordered structures such as powders and porous glasses. Although these materials are easy to fabricate, it is only recently that researchers have started to fully understand the rich and complex physical processes that take place in amplifying disordered systems. Here, I will give an overview of the various recent results and discuss the physical picture that has now emerged. I will also discuss possible applications of this new type of disorder-based laser light source.

DIEDERIK S. WIERSMA

European Laboratory for Non-linear Spectroscopy (LENS) and INFM-BEC, Via Nello Carrara 1, I-50019 Sesto-Fiorentino (Florence), Italy

e-mail: wiersma@lens.unifi.it

A laser is usually constructed from two basic elements: a material that provides optical gain through stimulated emission and an optical cavity that partially traps the light. When the total gain in the cavity is larger than the losses, the system reaches a threshold and lases. It is the cavity that determines the modes of a laser, that is, it determines the directionality of the output and its frequency. Random lasers work on the same principles, but the modes are determined by multiple scattering and not by a laser cavity.

Multiple scattering is a well-known phenomenon that occurs in nearly all optical materials that appear opaque. It is therefore quite common in daily life and determines the appearance of, for example, clouds, white paint, powders and even human tissue. Light rays that penetrate these materials are scattered often thousands of times in a random fashion before they exit again.

This type of propagation is that of a random walk, just as in the brownian motion of particles suspended in a liquid (Fig. 1). The fundamental parameters describing this process are the mean free path (the average step size in the random walk) and the diffusion constant. Scattering in disordered optical materials is complex yet completely coherent. This means that the phase of each of the optical wavelets undergoing a random walk is well defined and interference effects can occur, even if a material is strongly disordered. The most clear visualization of interference in multiply scattered light is that of laser speckle[1], which is the grainy pattern observed when looking at a laser pointer that is scattered from, for example, a piece of paper. The difference between light diffusion and multiple scattering is that diffusion refers to a simplified picture of multiple scattering in which interference effects are neglected.

Multiple scattering due to randomness not only occurs in natural materials, but is also intrinsically present in photonic materials, such as photonic crystals, intended for the realization of optical devices. In those materials, multiple scattering has always been considered an unwanted property arising from structural artefacts. It has now become clear that such artefacts are difficult to avoid and form a major industrial bottleneck[2]. Using multiple scattering to introduce new functionalities therefore opens up a completely new perspective on disorder in photonic materials.

EMISSION PROPERTIES OF A RANDOM LASER

Light diffusion with gain was already discussed theoretically by Letokhov in the 1960s. He argued that for a diffusion process with amplification, a situation where the total gain is proportional to the volume is obtained, whereas the losses will be proportional to the total surface[3]. It is then easy to see that there exists a critical volume above which gain becomes larger than loss, and the intensity diverges. If the gain depends on wavelength, this model also predicts that the emission spectrum narrows down above threshold with a maximum intensity at the wavelength of maximum gain. In addition, relaxation oscillations as well as laser spiking can be found in such a diffusive model[4].

Several of these features have indeed been observed in experiments. Briskina and co-workers[5] and later Migus and co-workers[6] made a disordered amplifying material by grinding laser crystal into a fine powder. They managed to excite this powder optically and achieve optical gain through stimulated emission together with multiple scattering. Above a certain threshold gain level, the emission spectrum was observed to narrow down and the peak intensity to increase (Fig. 2). A different strategy to achieve multiple scattering with gain was followed by Lawandy *et al.*[7], who suspended microparticles in laser dye. The advantages of such a material are that the amount of scattering can easily be varied by changing the particle concentration and that the material is a suspension and therefore fluid[8]. Lawandy called this material laser paint, as it constitutes a laser that can be painted directly on a surface. In early work, Lawandy was not always in the regime of random lasing, and also observed similar effects at zero or very small particle concentration[9,10]. He corrected this in later work[11]. To call a material a random laser, the multiple-scattering process has to play a role in determining the lasing process. The term random lasing was introduced in 1995 (refs 4,9).

Later theoretical approaches addressed the interesting issue of photon statistics in random-laser emission. It was found that the

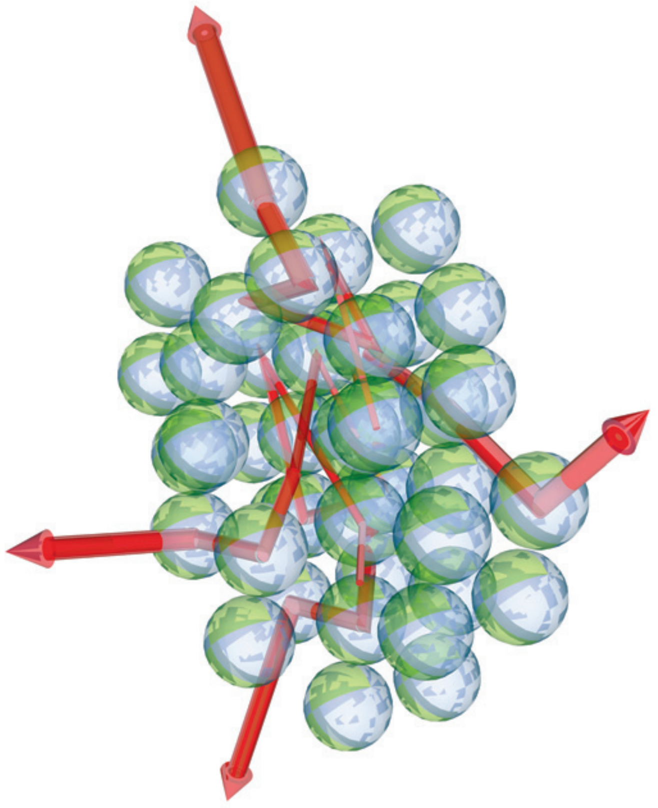

Figure 1 Multiple light scattering with gain. A random collection of microspheres containing laser dye is excited (for example, by an external light source) to obtain population inversion. The microspheres then scatter light and amplify it in the process. The propagation of the light waves becomes that of an amplified random walk.

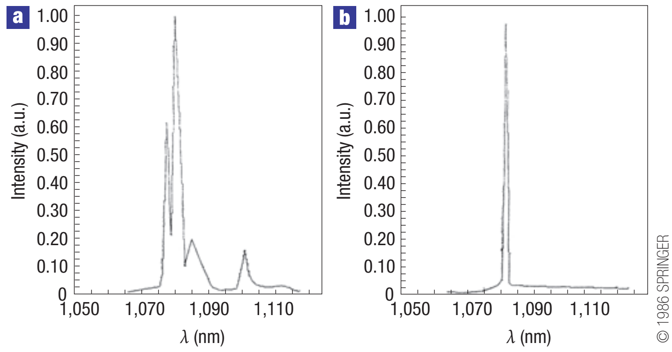

Figure 2 Observation of random lasing. The emission spectrum of a solid random-laser material based on neodinium- (Nd^{3+}) doped lanthanum oxide (La_2O_3). **a**, The fluorescence spectrum of the material below threshold. **b**, The material is above threshold and the spectrum narrows strongly. Reprinted with permission from ref. 5.

photon statistics of a random laser are very similar to those of a regular laser, in several aspects. Regular lasers, for instance, exhibit excess photon noise that originates from the interference between spontaneous and stimulated emission[12]. Beenakker and a co-worker showed, in random matrix calculations, that excess photon noise is also expected to occur in random lasers above threshold[13,14]. Florescu and John calculated the degree of second-order coherence, which is a measure of the fluctuations of the intensity and which characterizes the emission from a laser. The temporal distribution of photons from a chaotic source is bunched (photons have a high probability of arriving together or in other words they travel in 'bunches'), which leads to so-called Bose–Einstein statistics. This is in contrast to the poissonian statistics of the coherent state of a laser, where the photons are more equally distributed in time. Florescu and John predicted that a random laser above threshold should exhibit poissonian statistics, just like a regular laser[15]. This result could be derived by taking into account only diffusive scattering and no coherent feedback. Poissonian photon statistics in random-laser emission were first observed in experiments by Cao *et al.*[16].

Although a simplified model of diffusion with gain, as originally discussed by Letokhov, is very powerful in predicting certain emission properties of a random laser, it also neglects some important aspects. In particular, it neglects the fact that light rays in a random laser, while undergoing a random walk, are subject to interference effects. It is the interference in the multiple-scattering process that determines the mode structure of a random laser. Modes in a regular laser are determined by the laser cavity and usually consist of standing-wave patterns. In a random laser, the spatial profile of the modes is dominated by a speckle pattern with a gradually varying envelope. This interference plays an important role in all random-laser materials that have been studied so far, including the first structures realized by, for example, Briskina, Migus and Lawandy and their respective co-workers (see Box 1).

In this context, the relation between random lasing and amplified spontaneous emission can also be understood. Amplified spontaneous emission is light that originates from spontaneous emission and that is subsequently amplified by stimulated emission. This process takes place without an optical cavity and can therefore occur even in completely transparent active materials where rays can propagate freely. The spectrum of amplified spontaneous emission is determined by the gain curve of the active material. Owing to the stimulated emission process, this light can even become highly coherent under certain conditions (see Box 2). Amplified spontaneous emission is often classified as lasing without mirrors[17]. In a random laser, on the other hand, the multiple-scattering process defines optical modes with a certain central frequency and bandwidth, lifetime and a rich spatial profile. Random lasers are therefore 'mirror-less' but not 'mode-less'.

MODE STRUCTURE OF A RANDOM LASER

To develop a theory that can describe all aspects of a random laser is very difficult. A complete model would have to include the dynamics of the gain mechanism because gain saturation forms an intrinsic aspect of an amplifying system above threshold. Without gain saturation, the intensity would diverge leading to unphysical results. In addition, interference effects have to be included to describe the mode structure. Interference in multiple scattering leads to a granular distribution of the intensity called speckle (Fig. 3). In most random materials, the intensity is spread throughout the sample and the modes are extended. In certain random materials, interference can lead to an effect called light localization[18–20], which is the optical counterpart of Anderson localization of electrons[21]. Owing to interference, the free propagation of waves and thereby the multiple scattering process, comes practically to a halt in that case. This can be understood in terms of the formation of randomly shaped but closed modes with an overall exponentially decaying amplitude (Fig. 3). The average spatial extent of these localized modes defines a length scale called

Box 1: Definition of a random laser

There exists a misconception in the current literature that diffusive and coherent random lasers should be distinguished between. Such a distinction would suggest that in some materials the light scattering process is subject to interference effects, whereas in others it is not. In all random-laser materials, the multiple-scattering process is elastic, so that interference effects are present and part of the physical problem. The question is whether the effect of this interference is observed in a specific experimental configuration. For instance, by using long excitation pulses or by averaging over several laser shots, some interference effects are averaged out. The remaining effects after averaging, such as a smooth narrowing of the spectrum, can be described by a simplified model of diffusion with gain that does not need to take interference into account. It is however possible to observe from the same random-laser material either a smooth spectrum or narrow spikes. The latter can only be modelled by taking into account interference effects.

To model a random laser correctly requires solving Maxwell's equations for a system of randomly varying refractive index with a positive imaginary part. The optical modes that are found this way are complex and can be Anderson localized or otherwise confined in space or extended, depending on the mean free path of the sample. In addition, other properties can determine the nature of the modes, such as the amount of (long-range) correlation in the refractive index or the presence of partial order or strong anisotropy.

Given the broad scale of materials that is studied in the context of random lasing, and the rich physics of amplifying random systems, it is important to provide a clear definition of what is meant by a random laser. A good definition of a laser is an optical structure or material that satisfies the following two criteria: (1) light is multiply scattered owing to randomness and amplified by stimulated emission, and (2) there exists a threshold, due to the multiple scattering, above which total gain is larger than total loss. This definition includes all multiple scattering systems with gain in a broad range of the mean free paths ℓ. Whereas there is no lower bound for ℓ, an upper bound is approximately the system size, otherwise the sample becomes transparent.

Box 2: How coherent is a (random) laser?

Coherent feedback is not required to obtain random lasing. The reason is that, like in a regular laser, it is not the cavity itself that is essential for obtaining coherent laser emission. To understand this better, first- and second-order coherence should be distinguished between. First-order coherence is a measure of fluctuations of the field, whereas second-order coherence accounts for fluctuations of the intensity. For a source of sufficiently narrow bandwidth, the first-order coherence is automatically high. Therefore, any mechanism that selects a specific narrow wavelength band (for example, a bandpass filter) creates first-order coherence[65]. Second-order coherence is more difficult to obtain owing to the tendency of photons to 'bunch', which creates large intensity fluctuations. In a laser, second-order coherence is obtained by saturation of the gain. This nonlinear effect limits the fluctuations of the intensity and thereby increases second-order coherence. If light is first- and second-order coherent, the emission can be called 'coherent'.

A laser cavity creates feedback and thereby forms a convenient mechanism that automatically leads to the gain saturation required for second-order coherent light. However, there are other situations in which gain saturation creates light that has second-order coherence. An example is that of the amplification of spontaneously emitted photons by stimulated emission. If the gain is large, the intensity will grow such that it depletes the gain medium completely. This will suppress the fluctuations of the intensity and thereby give rise to second-order coherence, or in other words to the characteristic 'poissonian' photon statistics that characterize the coherent emission of a laser source. This also explains how a random laser can exhibit coherent emission, independently of the degree of localization of the modes and the amount of 'coherent' feedback.

the localization length. Localization can only take place in optical materials that are extremely strongly scattering, the requirement being that the mean free path ℓ becomes smaller than the reciprocal wavevector: $k\ell \leq 1$. This is also known as the Ioffe–Regel criterion[22].

The need for a detailed model of random lasing became clear after an observation by Cao and co-workers, who found that carefully performed experiments revealed narrow spikes in the emission spectrum on top of a global narrowing[23] (Fig. 4). Attempts to understand the origin of these spikes have led to a vivid discussion in the literature. At first it was proposed that Anderson localization could be behind the narrow emission spikes[24–26]. The idea of using localization for lasing had already been introduced by Pradhan and Kumar[27] and was picked up by Jiang and Soukoulis[24] and Vanneste and Sebbah[26]. They calculated the behaviour of lower-dimensional amplifying systems, such as random multilayer stacks and planar waveguides with disorder, and found that localized states can trap light and thereby enhance lasing effects. However, whereas localization is relatively easy to obtain in lower dimensions, it is extremely difficult to obtain in three-dimensional (3D) systems[22]. The reason is that the Ioffe–Regel criterion $k\ell \leq 1$ is very difficult to fulfil in optics. Most random optical materials have $k\ell$ values that are much larger than 1 and care should therefore be taken in extrapolating the theoretical results for 1D and 2D systems to 3D materials.

In a series of recent experiments, it was found that the narrow spikes of a random laser can be observed in nearly the entire range of scattering strengths that is experimentally easily accessible, with $k\ell$ values ranging between 10 and 10^4 (refs 28–30). As these scattering strengths are much too weak to produce Anderson-localization effects, researchers started to look for alternative models. An alternative mechanism that could lead to resonances was suggested, for instance, by Apalkov *et al.*, who discussed

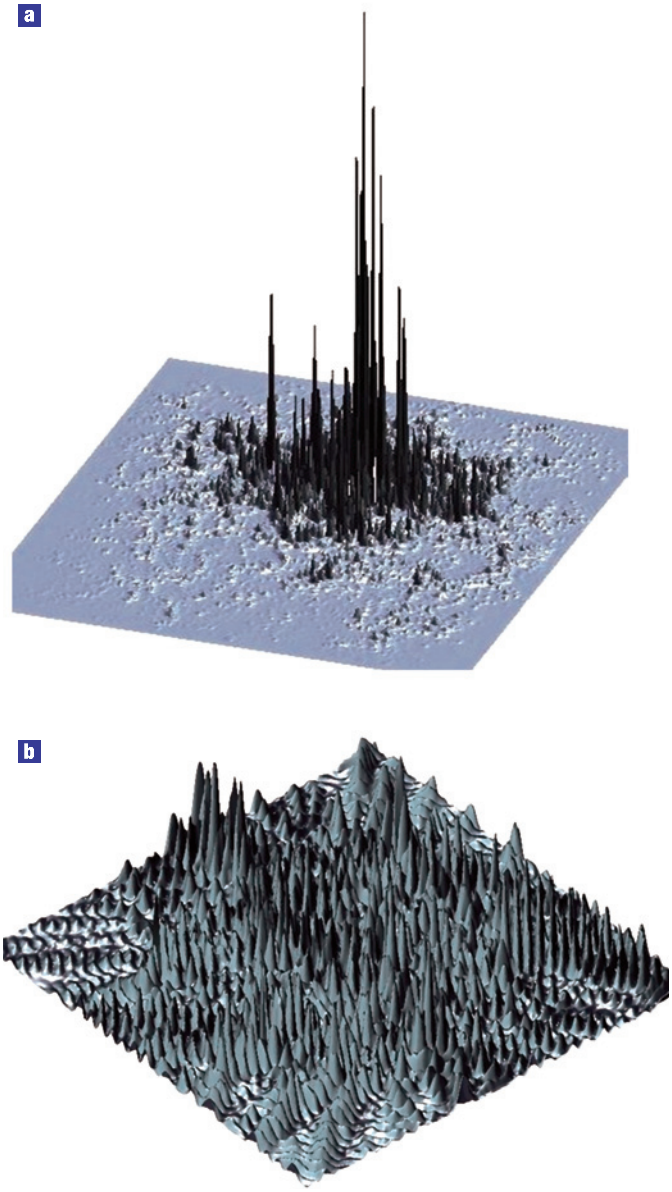

Figure 3 Extended versus localized optical modes. Finite-difference time-domain calculation of the distribution of the electromagnetic-field intensity in a disordered system. **a**, A localized mode. Here, the light is confined to a mode with exponentially decaying tails. **b**, An extended mode. The strongly fluctuating pattern is called speckle. The calculation is carried out for a 2D planar waveguide with random pores. A similar pattern can be expected inside 3D random materials.

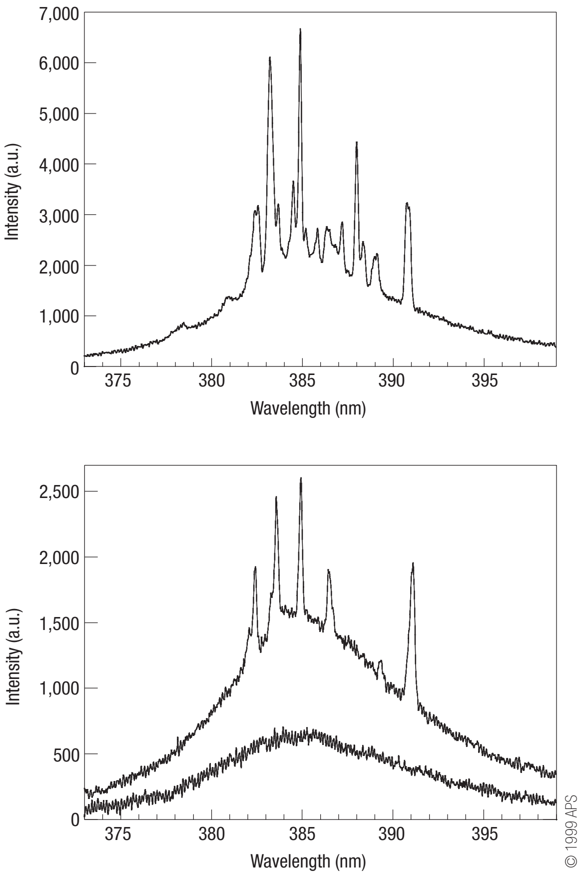

Figure 4 Complex emission spectra exhibiting narrow spikes. Emission spectra of a random laser exhibiting an overall narrowing of the spectrum together with narrow spikes at random frequencies. The sample consists of zinc oxide powder with grain size of the order of 100 nm and is excited by laser pulses of 15 ps pulse duration at 355 nm wavelength (excitation area from bottom to top: 980, 1,350 and 1,870 mm^2). Reprinted with permission from ref. 23.

theoretically the consequences of having a refractive index that varies randomly[31]. They suggest that, in a 2D system, the random variations of the refractive index could cause, by chance, structures that act as waveguides, thereby forming resonant modes.

To better understand the mode structure of a random laser, researchers realized that it was crucial to first understand the modes of passive random materials without gain. To that end, the decay rate statistics of the modes are extremely useful because they provide a clear tool to investigate localization and determine if a material sustains localized or extended modes, or both[32–34]. If gain is then introduced in such materials, it will be the modes with the longest lifetime that have the lowest lasing threshold and that acquire the highest intensity[35]. Chabanov *et al.* studied these decay rate statistics experimentally and suggested that long-lived extended modes, in regular diffusive materials, might be responsible for the observed narrow spikes in random-laser spectra[33]. Mujumdar and co-workers then calculated these modes in numerical simulations and found that indeed in a finite-size random system there exists a subset of rare extended modes with very long lifetime, which become very important when gain is introduced[29]. Mujumdar and co-workers showed that the ‘lucky photons’ that are spontaneously emitted in such long-lived modes can acquire a huge gain and give rise to spikes in the emission spectrum of a random laser.

To determine experimentally the degree of localization of random-laser modes, van der Molen *et al.* have analysed experimentally their spatial extent by confocal microscopy[36]. In this experiment, the material was both analysed and excited through the same microscope objective, which meant that the excited region was small. The modes that they observed this way were of the same size as the diameter of the excited region, so that they could not discriminate between localized or extended modes. Recently, the modes of a random laser were calculated and visualized by Sebbah *et al.* in direct finite-difference time-domain calculations (Fig. 5). They observed that extended modes that cover the entire

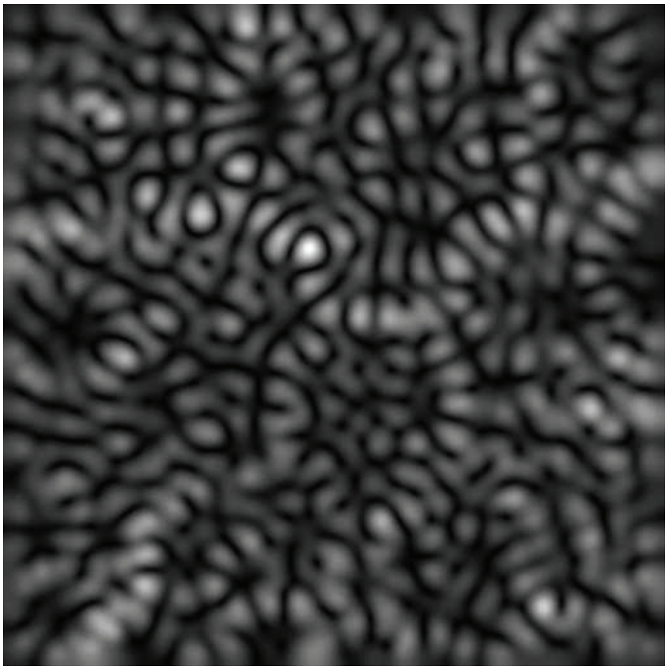

Figure 5 Numerical calculation of mode profile. Numerical calculation of the distribution of the electromagnetic-field intensity in a random laser. The plotted mode is above threshold and lases. The spatial extent of the mode is large and it covers a significant part the sample. Reprinted with permission from ref. 37.

sample can form lasing modes and lead to spectrally narrow emission[37], even if their quality factor is quite low.

Although it is currently unknown with certainty whether lasing in zinc oxide, as studied first by Cao and co-workers, occurs in localized or extended modes, an overall agreement of possible random-lasing mechanisms has come forward. Both extended and localized modes in random systems can lase and lead to spectrally narrow and coherent output. Depending on the experimental geometry, this can be observed as narrow spikes, or as global narrowing of the spectrum if the narrow spikes are averaged out. Differences between the localized and extended mode picture include the degree of mode coupling, which is obviously much lower for the localized case. Localized modes will also suffer more from gain saturation because they cover a smaller gain volume. The intensity contained in the extended modes can therefore grow much higher before the gain is depleted. The debate that has evolved in the literature on this issue shows how rich and complex the physical processes behind multiple scattering in random systems are, especially when gain is introduced into the problem.

CHAOTIC BEHAVIOUR AND LEVY STATISTICS

A laser that sustains a large number of modes that are strongly coupled can become very sensitive to the boundary conditions. In a pulsed configuration, such a laser can show chaotic behaviour in its temporal and spectral response. If there is no specific frequency that dominates the others, the laser can have a different spectrum each time it is excited. The reason for this is that lasing starts from spontaneous emission which is different at every pulse.

In a random laser, a similar situation can be expected owing to the large number of random modes that compete for the available gain. Conti and co-workers[38] showed that this can lead in a random laser to a behaviour similar to that of a glass transition, in which a certain configuration is 'frozen in'. Mode coupling is particularly strong for extended modes that cover a large volume of the sample. Several modes can partially occupy the same volume and therefore compete for the same gain molecules. The mode coupling therefore takes place through the gain mechanism, which means that modes of different wavelengths can still be coupled, even if they are not spectrally overlapping.

Under the right conditions, a random laser can therefore become very sensitive to tiny intensity fluctuations at the start of each laser pulse. Such effects can be observed in the emission spectra of random lasers[39]. It was found that the narrow emission spikes in the emission spectrum can change frequency in a random fashion from one excitation pulse to another. The emission spectra are then completely uncorrelated from shot to shot (Fig. 6a). As the scattering particles are not moving in these experiments, and all other conditions are kept perfectly constant, these differences between spectra can only be due to the spontaneous emission from which the random laser starts at each shot. This chaotic behaviour is only observed under specific conditions, using, for example, a fast excitation source and by collecting single-shot emission spectra.

Mode coupling in random lasers was shown to lead to level repulsion, meaning that resonances that are very close in wavelength repel each other, thereby enhancing their wavelength difference[36,40]. In addition, the threshold of a random-laser system can show chaotic behaviour, in the sense that under repeatable experimental conditions the system jumps above and below threshold as shown by Anglos and co-workers[41] (Fig. 7). This can again be understood as being due to a very high sensitivity of the system to small intensity fluctuations[42].

An interesting consequence of these strong fluctuations is that the intensity distribution does not obey regular gaussian statistics, but rather becomes of the Levy type. Levy distributions have an infinite variance, owing to the occurrence of rare but very large values, and are characterized by a slowly decaying (power-law)

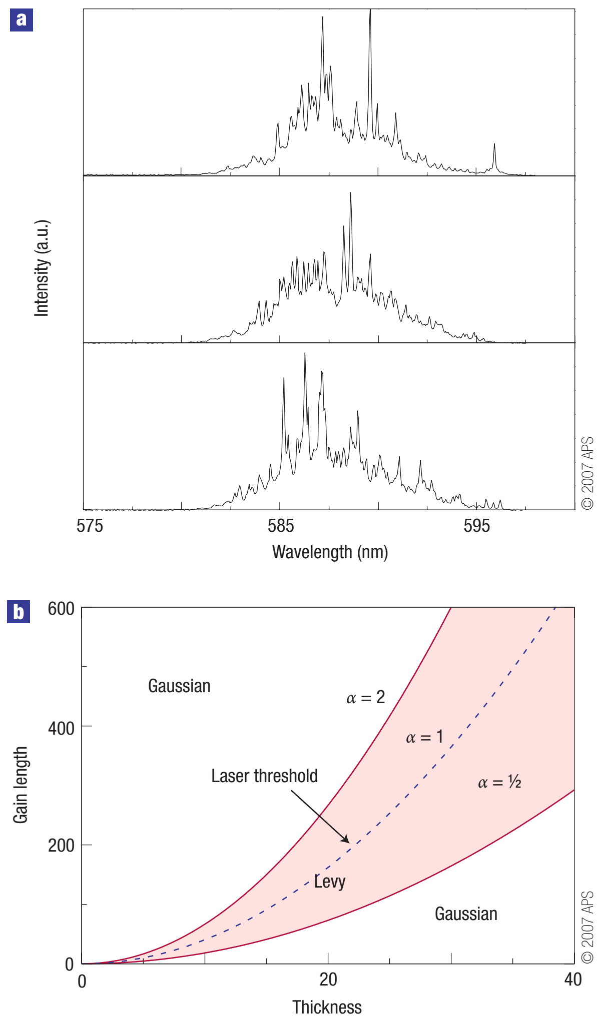

Figure 6 Chaotic behaviour and Levy statistics. **a**, Series of emission spectra taken at successive excitation shots. The single-shot spectra are clearly very different, indicating chaotic behaviour. The sample consists of porous glass doped with laser dye and is excited by single-shot laser pulses of 15 ps pulse duration and 532 nm wavelength. **b**, Phase diagram of random-laser fluctuations (gain length versus sample size). In the red regime, the statistics of the intensity fluctuations are expected to be of the Levy type. The blue line indicates the threshold. α is the ratio between the gain length and the average path length of the light inside the sample. Note that $\alpha = 1$ corresponds, therefore, to the laser threshold. Reprinted with permission from ref. 39 (**a**) and ref. 45 (**b**).

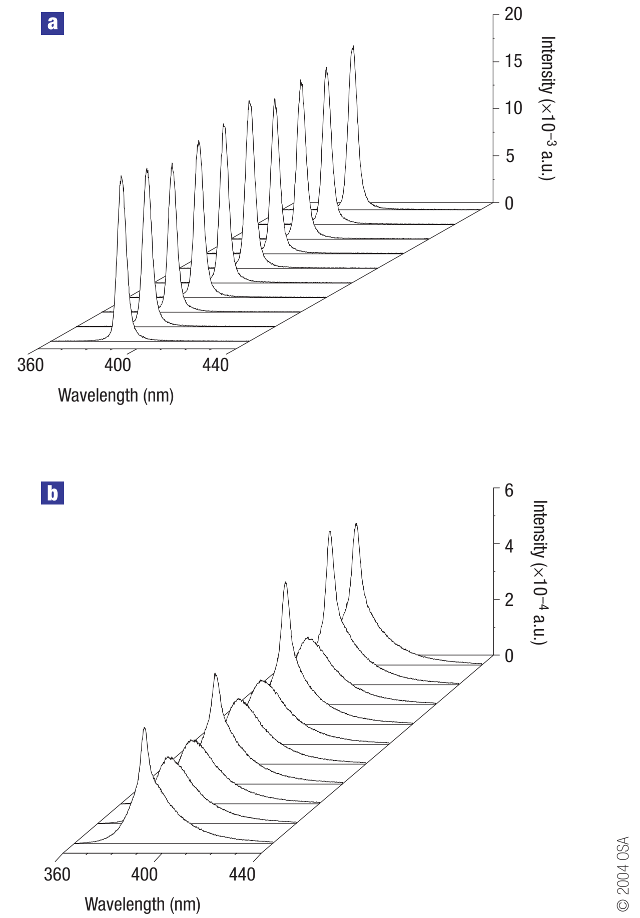

Figure 7 Threshold fluctuations. Series of emission spectra from a random laser based on a zinc oxide polymer composite. **a**, Picosecond, high peak intensity excitation. **b**, Nanosecond, modest peak intensity excitation close to threshold. The series corresponds to emission spectra observed in successive excitation pulses. Although the excitation energy is constant, the system randomly jumps above and below threshold at successive excitation events, as is clearly visible in **b**. These strong fluctuations again indicate a chaotic behaviour of the system. Reprinted with permission from ref. 41.

tail. Such distributions appear in various processes in nature. The food-search pattern of albatrosses were, for instance, found to obey Levy statistics[43]. Although it is very rare to find such behaviour in optics, the groups of Kumar and Ramachandran managed to identify Levy statistics in the intensity distribution of a random laser[44]. Later such behaviour was also found in molecular-dynamics-type simulations[45] (Fig. 6b). In particular, it was found that there exists a regime around the laser threshold where Levy statistics apply, whereas both far below and far above threshold the statistics remain gaussian. This Levy regime is most likely also the range in which the spectra behave chaotically.

The shot-to-shot fluctuations are probably the reason that some initial experimental studies have missed the observation of narrow spikes[46]. If several emission shots are averaged over, with the random laser being in the chaotic regime, the spikes will be averaged out. The same could possibly occur even within a single excitation pulse, if the pulse duration is long enough. Therefore, ideal conditions for the observation of spikes seem to be short (picosecond) excitation and single-shot observation. The details of this chaotic behaviour are, however, still not fully understood and future research is required to better understand the rich physics involved.

MATERIALS AND GAIN MECHANISMS

A wide range of random-laser materials that can be relatively easily produced on an industrial scale has become available. When realizing a random-laser material, it is important to create strong enough scattering for the material to become optically thick. That is, the mean free path ℓ should become at least smaller than the sample thickness. This way the material will appear opaque, in contrast to optically thin samples that look nearly transparent. Strong enough scattering can be achieved by grinding a material into a powder, by suspending scattering elements in solution, or, for instance, by etching a porous network of air into a solid glass or semiconductor crystal[47]. The last of these has the advantage that it can lead to very strong scattering while the pore size, and thereby the scattering strength, can be controlled in the etching process.

The disadvantage of ground and etched materials is that the diameter and shape of the scattering elements remain ill-defined. A different approach to achieving strong and controllable scattering has recently been put forward by assembling monodisperse spheres in a random fashion[48] (Fig. 8a). In analogy to photonic crystals, which consist of an ordered assembly of microspheres, researchers have dubbed this new random material a photonic glass. By using monodisperse spheres, a material in which the scattering at certain wavelengths is resonant with the sphere diameter can be obtained. This leads to strong resonant scattering, which favours random-laser modes at the specific wavelengths of the resonances. Hence, the emission bands of the random laser could be tuned this way by varying the particle diameter.

Essential in the realization of a random laser is to obtain enough gain to reach the laser threshold. Fortunately a laser crystal retains its capacity to amplify light through stimulated emission after grinding. A good candidate for this is titanium-doped sapphire, which emits at visible and near-infrared wavelengths. An alternative crystalline material is zinc oxide, which is very suitable for random lasing in the near-ultraviolet region, owing to its high gain and strong potential for scattering. For visible wavelengths, gain can be obtained by incorporating laser dyes, either in solution or incorporated into the scattering elements or the solid surrounding them.

In all cases, the gain material has to be excited to reach a population inversion. Like in many dye and solid-state lasers, this can be achieved through optical pumping. Nearly all random lasers that have been realized so far work in a pulsed regime, and are excited by a high-power pump laser. The excitation mechanism is not as straightforward as in a regular laser, because the pump light is also multiply scattered by the disordered structure. This essentially leads to a diffusive propagation of the excitation light and therefore a, to first-order, linear dependence of the excitation energy with depth.

Although it is relatively easy to synthesize a random-laser material that supports extended modes, it is extremely difficult to realize one in which the modes are localized. This requires very strong scattering and therefore scattering elements with a size comparable to the wavelength and very high refractive index. At the same time, absorption has to be avoided because it would be counterproductive for lasing. One possible approach is to try to induce a population inversion in disordered gallium arsenide or other semiconductor structures[49]. Evidence for Anderson-localized modes of near-infrared light waves has so far been found in fine powders of gallium arsenide, which has a refractive index as high as 3.5 (refs 50–52). For visible light, signatures of localized modes with long lifetime have been found in titanium dioxide powders, which have a refractive index of about 2.7 (refs 53–55).

APPLICATIONS

A major advantage of random lasers over regular lasers is that their production is cheap and the required technology relatively simple. The high-precision methods needed to create ultraprecise microcavities, used in for, example, diode lasers, are not required here. In addition, the materials can be produced on a large scale and have a high emission efficiency.

The properties that make a random laser special in respect to regular lasers are its colour and angular dependence, as well as its complex features in emission spectra. The angular distribution of the output of a random laser is very broad and can be distributed

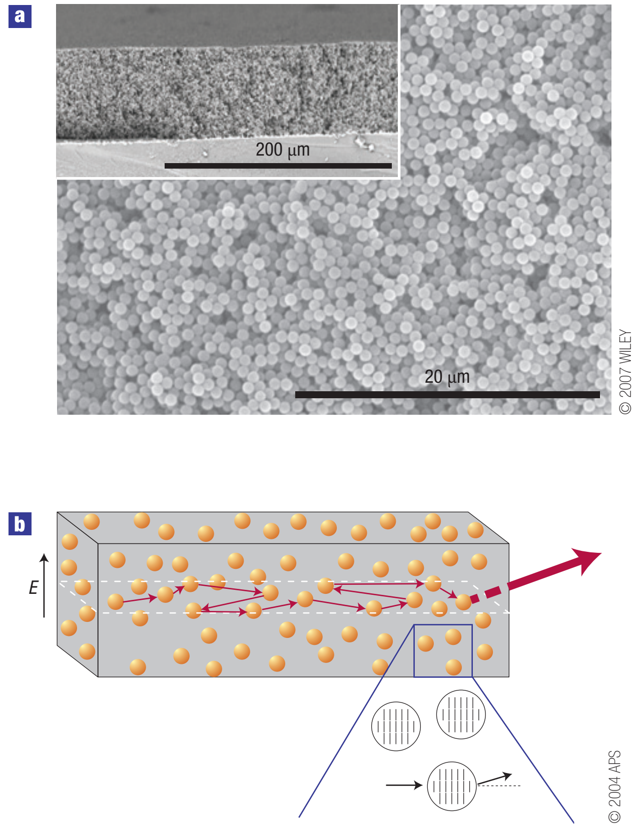

Figure 8 Random-laser materials. **a**, Photonic glass consisting of a random collection of highly monodisperse microspheres. The resonances of the spheres lead to strong scattering at specific wavelengths, which can be used to tune the emission spectrum of a random laser. **b**, Quasi-2D random walk with gain in a random laser based on polymer-dispersed liquid crystals. The multiple scattering in these materials is due to small droplets of liquid crystal (shown as orange spheres) incorporated in a polymer matrix. The droplets contain laser dye to achieve both scattering and optical gain. If an electric field *E* is applied in the vertical direction, the liquid-crystal molecules align along the field (shown in the magnified view) and the scattering becomes highly anisotropic. This anisotropy is so strong that it leads to a quasi-2D diffusion process. The effect enables electrical control over the emission of a random laser: when the electric field is applied, the emission regains (planar) directionality and becomes polarized. Reprinted with permission from ref. 48 (**a**) and ref. 56 (**b**).

over the complete solid angle of 4π. This broad angular emission is in principle ideal for display applications. However, to develop such applications it is crucial to have electrical control over the emission. It was found by Gottardo *et al.*[56] that this directionality can indeed be electrically tuned by realizing a random laser from a polymer-dispersed liquid crystal, a material commonly applied to realize liquid-crystal displays. In these experiments, a new type of active polymer-dispersed liquid crystal was realized using liquid-crystal droplets in a polymer matrix with added laser dye. The liquid-crystal droplets are optically birefringent and therefore give rise to an extremely anisotropic, quasi-2D diffusion process, when all aligned in the same direction by an electric field (Fig. 8b). The result is a random laser that emits mainly in a plane and of which the output is polarized with controllable polarization.

A particular advantage of random-laser materials is that they can be prepared in the form of suspensions of particles that can be applied as coatings on surfaces of arbitrary shape[57]. This has interesting potential for environment lighting, for example, in the form of street lighting that is applied directly on the road. Such a coating can also be applied to road vehicles, ships and aircraft. The same technology has been patented for use in the identification of friendly/enemy vehicles[58].

In addition to electrical control over random-laser emission, temperature tuning has been demonstrated as well[59]. In this case, the random-laser spectrum depends strongly on environment temperature and the random laser can even be brought above and below threshold by temperature changes. This can be applied for remote temperature sensing in hostile environments by inserting a grain of random-laser material in the environment and probing its emission spectrum remotely with a telescope. In addition, this property can be applied to create coatings of which the colour is temperature dependent and, for example, smart windows with different optical properties in summer/winter.

The combination of localization and random lasing is particularly interesting because every individual random-laser source would give a unique emission spectrum defined by the specific localized modes in each sample. Complicating chaotic effects due to mode competition would also be much weaker in this case. This would enable coding of objects and, for example, documents or banknotes, by incorporating a small amount of random-laser material that would manifest itself with a unique emission spectrum, but only when excited by a bright light source.

Localization can be relatively easily obtained in lower-dimensional systems, such as random multilayers and planar waveguides with disorder. Recently, Milner and Genack demonstrated experimentally that localized modes in amplifying random multilayers can indeed be created, and that these 1D systems can be brought to lase[60]. The output is in this case completely directional, as from a regular laser. The spectrum can become very narrow and the output is again coherent. A useful property of this random laser is that the cavity is intrinsically robust against disorder. Small manufacturing errors can shift the frequency of the lasing modes, but will not ruin the efficiency of the system.

Recently, an application was proposed for random lasers in a very different area, namely that of medical diagnostics. Vardeny and co-workers found that the emission spectrum of cancerous human tissue, when doped with laser dye, could be distinguished from healthy tissue[61]. This would enable an alternative strategy for tumour diagnostics. The result links to the very active field of biomedical imaging with diffuse light and makes use of the fact that different tissue and bone types have different optical structures and mean free paths. Techniques such as photon-density wave analysis and diffusing wave spectroscopy, all based on the knowledge of multiple light scattering in random systems, have already proved their value in characterizing tissue and monitoring blood flow during treatment and surgery[62].

OUTLOOK

In future studies, it would be interesting to combine strong scattering with gain and try to observe direct signatures of Anderson localization on one side and random lasing on the other, from the same material. A possible approach to reach this could be the introduction of high enough gain in strongly scattering GaAs powders, for example, by electrical pumping of the GaAs itself or the introduction of quantum-dot structures. A first step in this direction was done by Noginov, who managed to achieve gain and lasing in random GaAs structures[49]. Alternatively, approaches could involve composite structures based on strongly scattering titanium dioxide combined with efficient gain materials such as zinc oxide or possibly colloidal quantum dots.

Another topic for future research could be the study of interference effects in the multiply scattered excitation light and to look for phase correlations between the excited molecules arising from the mode pattern of the pump light. Such phase correlations could, for example, lead to effects such as super-radiance, in which the well-defined phase between the emitting molecules leads to coherent emission effects. In particular, constructive interference leads, in that case, to a high and rapidly decaying emission peak that depends on the square of the number of molecules.

The main challenge for the development of future applications is that of electrical excitation of a random-laser material, which is crucial for display and lighting technology. Electrical conductance is going to be an important issue here, owing to the disordered and often porous character of the materials under study. Initial studies of rare-earth-doped oxide powders that can be excited electrically were carried out by Rand and co-workers[63]. Interestingly, random lasing by electrical pumping has recently been achieved in the terahertz regime by Mahler and Tredicucci[64]. They managed to observe random lasing of localized modes in a quasi-periodic 1D structure, pumping the device electrically with very high efficiency. Although development of applications of random lasers is now on its way, it has also become clear that the physics of these materials is very rich and is bound to provide us with more surprises in the near future.

doi:10.1038/nphys971

References

1. Feng, S., Kane, C., Lee, P. A. & Stone, A. D. Correlations and fluctuations of coherent wave transmission through disordered media. *Phys. Rev. Lett.* **61**, 834–838 (1988).
2. Koenderink, F., Lagendijk, A. & Vos, W. L. Optical extinction due to intrinsic structural variations of photonic crystals. *Phys. Rev. B* **72**, 153102 (2005).
3. Letokhov, V.S. Generation of light by a scattering medium with negative resonance absorption. *Zh. Eksp. Teor. Fiz.* **53**, 1442–1447 (1967); *Sov. Phys. JETP* **26**, 835–840 (1968).
4. Wiersma, D. S. & Lagendijk, A. Light diffusion with gain and random lasers. *Phys. Rev. E* **54**, 4256–4265 (1996).
5. Markushev, V. M., Zolin, V. F. & Briskina, Ch. M. Powder laser. *Zh. Prikl. Spektrosk.* **45**, 847–850 (1986).
6. Gouedard, C., Husson, D., Sauteret, C., Auzel, F. & Migus, A. Generation of spatially incoherent short pulses in laser-pumped neodymiun stoichiometric crystals and powders. *J. Opt. Soc. Am. B* **10**, 2358–2362 (1993).
7. Lawandy, N. M., Balachandran, R. M., Gomes, A. S. L. & Sauvain, E. Laser action in strongly scattering media. *Nature* **368**, 436–438 (1994).
8. Sha, W. L., Liu, C. H. & Alfano, R. R. Spectral and temporal measurements of laser action of Rhodamine-640 dye in strongly scattering media. *Opt. Lett.* **19**, 1922–1924 (1994).
9. Wiersma, D. S., van Albada, M. P. & Lagendijk, A. Random laser? *Nature* **373**, 203–204 (1995).
10. Lawandy, N. M. & Balachandran, R. M. Random laser?—Reply. *Nature* **373**, 204 (1995).
11. Martorell, J., Balachandran, R. M. & Lawandy, N. M. Radiative coupling between photonic paint layers. *Opt. Lett.* **21**, 239–241 (1996).
12. Petermann, K. Calculated spontaneous emission factor for double-heterostructure injection lasers with gain-induced waveguiding. *IEEE J. Quantum. Electron.* **15**, 566–570 (1979).
13. Beenakker, C. W. J. Thermal radiation and amplified spontaneous emission from a random medium. *Phys. Rev. Lett.* **81**, 1829–1832 (1998).
14. Mishchenko, E. G. & Beenakker, C. W. J. Radiative transfer theory for vacuum fluctuations. *Phys. Rev. Lett.* **83**, 5475–5478 (1999).
15. Florescu, L. & John, S. Photon statistics and coherence in light emission from a random laser. *Phys. Rev. Lett.* **93**, 013602 (2004).
16. Cao, H., Ling, Y., Xu, J. Y., Cao, C. Q. & Kumar, P. Photon statistics of random lasers with resonant feedback. *Phys. Rev. Lett.* **86**, 4524–4527 (2001).
17. Siegman, A. E. *Lasers* (Univ. Science Books, Mill Valley, 1986).
18. John, S. Electromagnetic absorption in a disordered medium near a photon mobility edge. *Phys. Rev. Lett.* **53**, 2169–2172 (1984).
19. Anderson, P. W. The question of classical localization: A theory of white paint? *Phil. Mag. B* **52**, 505–508 (1985).
20. Lagendijk, A., van Albada, M. P. & van der Mark, M. B. Localization of light: The quest for the white hole. *Physica A* **140**, 183–190 (1986).
21. Lee, P. A. & Ramakrishnan, T. V. Disordered electronic systems. *Rev. Mod. Phys.* **57**, 287–337 (1985).
22. Abrahams, E., Anderson, P. W., Licciardello, D. C. & Ramakrishnan, T. V. Scaling theory of localization: Absence of quantum diffusion in two dimensions. *Phys. Rev. Lett.* **42**, 673–676 (1979).
23. Cao, H. *et al.* Random laser action in semiconductor powder. *Phys. Rev. Lett.* **82**, 2278–2281 (1999).
24. Jiang, X. Y. & Soukoulis, C. M. Time dependent theory for random lasers. *Phys. Rev. Lett.* **85**, 70–73 (2000).
25. Cao, H. *et al.* Spatial confinement of laser light in active random media. *Phys. Rev. Lett.* **84**, 5584–5587 (2000).
26. Vanneste, C. & Sebbah, P. Selective excitation of localized modes in active random media. *Phys. Rev. Lett.* **87**, 183903 (2001).
27. Pradhan, P. & Kumar, N. Localization of light in coherently amplifying random media. *Phys. Rev. B* **50**, 9644–9647 (1994).
28. Polson, R. C., Chipouline, A. & Vardeny, Z. V. Random lasing in pi-conjugated films and infiltrated opals. *Adv. Mater.* **13**, 760–764 (2001).
29. Mujumdar, S., Ricci, M., Torre, R. & Wiersma, D. S. Amplified extended modes in random lasers. *Phys. Rev. Lett.* **93**, 053903 (2004).

30. Strangi, G. *et al*. Random lasing and weak localization of light in dye-doped nematic liquid crystals. *Opt. Express* **14**, 7737–7744 (2006).
31. Apalkov, V. M., Raikh, M. E. & Shapiro, B. Random resonators and prelocalized modes in disordered dielectric films. *Phys. Rev. Lett.* **89**, 016802 (2002).
32. Chabanov, A. A. & Genack, A. Z. Statistics of dynamics of localized waves. *Phys. Rev. Lett.* **87**, 233903 (2001).
33. Chabanov, A. A., Zhang, Z. Q. & Genack, A. Z. Breakdown of diffusion in dynamics of extended waves in mesoscopic media. *Phys. Rev. Lett.* **90**, 203903 (2003).
34. Skipetrov, S. E. & van Tiggelen, B. A. Dynamics of weakly localized waves. *Phys. Rev. Lett.* **92**, 113901 (2004).
35. Patra, M. Decay rate distributions of disordered slabs and application to random lasers. *Phys. Rev. E* **67**, 016603 (2003).
36. van der Molen, K. L., Tjerkstra, R. W., Mosk, A. P. & Lagendijk, A. Spatial extent of random laser modes. *Phys. Rev. Lett.* **98**, 143901 (2007).
37. Vanneste, C., Sebbah, P. & Cao, H. Lasing with resonant feedback in weakly scattering random systems. *Phys. Rev. Lett.* **98**, 143902 (2007).
38. Angelani, L., Conti, C., Ruocco, G. & Zamponi, F. Glassy behavior of light. *Phys. Rev. Lett.* **96**, 065702 (2006).
39. Mujumdar, S., Tuerck, V., Torre, R. & Wiersma, D. S. Chaotic behavior of a random laser with static disorder. *Phys. Rev. A* **76**, 033807 (2007).
40. Cao, H., Jiang, X., Ling, Y., Xu, J. Y. & Soukoulis, C. M. Mode repulsion and mode coupling in random lasers. *Phys. Rev. B* **67**, 161101 (2003).
41. Anglos, D. *et al*. Random laser action in organic–inorganic nanocomposites. *J. Opt. Soc. Am. B* **21**, 208–213 (2004).
42. van der Molen, K., Mosk, A. P. & Lagendijk, A. Intrinsic intensity fluctuations in random lasers. *Phys. Rev. A* **74**, 053808 (2006).
43. Viswanathan, G. M. *et al*. Lévy flight search patterns of wandering albatrosses. *Nature* **381**, 413–415 (1996).
44. Sharma, D., Ramachandran, H. & Kumar, N. Lévy statistics of emission from a novel random amplifying medium: An optical realisation of the Arrhenius cascade. *Opt. Lett.* **31**, 1806–1808 (2006).
45. Lepri, S., Cavalieri, S., Oppo, G. L. & Wiersma, D. S. Statistical regimes of random laser fluctuations. *Phys. Rev. A* **75**, 063820 (2007).
46. van Soest, G. *Experiments on Random Lasers*. PhD thesis, Univ. Amsterdam (2001).
47. Schuurmans, F. J. P., Vanmaekelbergh, D., van de Lagemaat, J. & Lagendijk, A. Strongly photonic macroporous GaP networks. *Science* **284**, 141–143 (1999).
48. Garcia, P. D., Sapienza, R., Blanco, A. & López, C. Photonic glass: A novel random material for light. *Adv. Mater.* **19**, 2597–2602 (2007).
49. Noginov, M. A. *Solid-State Random Lasers* (Springer Series in Optical Sciences, Vol. 105, Springer, Berlin, 2005).
50. Wiersma, D. S., Bartolini, P., Lagendijk, A. & Righini, R. Localization of light in a disordered medium. *Nature* **390**, 671–673 (1997).
51. Scheffold, F., Lenke, R., Tweer, R. & Maret, G. Localization or classical diffusion of light? *Nature* **398**, 206–207 (1999).
52. Wiersma, D. S., Gomez-Rivas, J., Bartolini, P., Lagendijk, A. & Righini, R. Reply: Localization or classical diffusion of light? *Nature* **398**, 207 (1999).
53. Wiersma, D. S., van Albada, M. P., van Tiggelen, B. A. & Lagendijk, A. Experimental evidence for recurrent multiple scattering events of light in disordered media. *Phys. Rev. Lett.* **74**, 4193–4196 (1995).
54. Chabanov, A. A., Stoytchev, M. & Genack, A. Z. Statistical approach to photon localization. *Nature* **404**, 850–852 (2000).
55. Störzer, M., Gross, P., Aegerter, C. M. & Maret, G. Observation of the critical regime near Anderson localization of light. *Phys. Rev. Lett.* **96**, 063904 (2006).
56. Gottardo, S., Cavalieri, S., Yaroschuck, O. & Wiersma, D. S. Quasi 2D random laser action. *Phys. Rev. Lett.* **93**, 263901 (2004).
57. Laine, R. M., Rand, S. C., Hinklin, T. & Williams, G. R. Ultrafine powders and their use as lasing media. US Patent 6,656,588 (2003).
58. Dubois, J. & la Rochelle, S. Active cooperative tuned identification friend or foe (ACTIFF). US Patent 5,966,227 (1999).
59. Wiersma, D. S. & Cavalieri, S. A temperature-tunable random laser. *Nature* **414**, 708–709 (2001).
60. Milner, V. & Genack, A. Z. Photon localization laser: Low-threshold lasing in a random amplifying layered medium via wave localization. *Phys. Rev. Lett.* **94**, 073901 (2005).
61. Polson, R. C. & Varden, Z. V. Random lasing in human tissues. *Appl. Phys. Lett.* **85**, 1289–1291 (2004).
62. Choe, R. *et al*. Diffuse optical tomography of breast cancer during neoadjuvant chemotherapy: A case study with comparison to MRI. *Med. Phys.* **32**, 1128–1139 (2005).
63. Williams, G. R., Bayram, S. B., Rand, S. C., Hinklin, T. & Laine, R. M. Laser action in strongly scattering rare-earth-metal-doped dielectric nanophosphors. *Phys. Rev. A* **65**, 013807 (2001).
64. Mahler, L. & Tredicucci, A. Electrically pumped random laser in the Tera Hertz regime (2008, in the press).
65. Lang, R., Scully, M. O. & Lamb, W. E. Why is the laser line so narrow? A theory of single-quasimode laser operation. *Phys. Rev. A* **7**, 1788–1797 (1973).

Acknowledgements

I wish to thank the entire optics of complex systems group at LENS for discussions as well as S. Mujumdar, S. Lepri, S. Cavalieri, R. Righini, M. Colocci, T. Arecchi, F. Marin, A. Lagendijk, K. van der Molen, A. Mosk, D. Anglos, A. Genack, P. Sebbah, M. Noginov and H. Cao. In addition, I thank the authors of the various papers discussed in this review for providing high-quality reprints of their figures and F. Riboli and P. Barthelemy for the finite-difference time-domain calculations on the extended and localized field distributions. This work was partially financed by the European Network of Excellence Phoremost.

Author information

Reprints and permission information is available online at http://npg.nature.com/reprintsandpermissions.