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Laser action in very white paint

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The subtle interplay of light scattering, interference and amplification in disordered materials such as paint and powdered laser crystals has opened up a new field in optics

Laser action in very white paint

DIEDERIK WIERSMA AND AD LAGENDIJK

NOWADAYS lasers are well known to everyone. They are used in industry and in hospitals, in supermarket bar code scanners, compact disc players and laser light shows. The difference between light from a laser and that from a normal electric light bulb is well known to physicists. Laser light has a well defined colour and direction, whereas light bulbs emit radiation with a range of colours and directions.

Disordered systems are less well known, although they are common in our everyday lives – for example, clouds, fog, sugar, sand, human tissue and white paint. On a macroscopic level, the propagation of light through these materials is treacherously simple: incoming beams get attenuated and a diffuse “glow” is formed. On a microscopic level, however, this problem is extremely complicated. White paint, for instance, consists of a large collection of small particles which are distributed randomly. Light incident on white paint is randomly scattered thousands of times before it leaves the paint again, thereby performing a complicated random walk. Multiple scattering in all kinds of disordered systems is currently an active area in both fundamental and applied research (see box).

Two key processes in the interaction of light and matter are scattering and absorption. If an object strongly absorbs light at all wavelengths, it appears black. On the other

hand, if it just scatters light, it looks white. Scientists who study multiple light scattering are always fighting against absorption because it kills the higher-order scattering events – due to absorption, the light simply disappears after being scattered a few times. The opposite applies when one tries to optimize the operation of a laser: one

studies the absorption (and gain) and wants to minimize scattering because it leads to losses from the cavity. Recently, however, physicists have been able to make a connection between laser action and multiple scattering.

There are many types of laser: gas, glass, dye and semiconductor lasers to name a few. For a laser material to be capable of amplifying light, it must be excited: that is, some upper lasing state must have a larger population than the ground state. This can be achieved with a suitable “pump” such as a strong flash lamp or another laser. After excitation, incident light at the lasing wavelength can be amplified by stimulated emission.

What would happen if we took a beautiful laser crystal and ground it into a fine powder? If we could excite the powder, it would still amplify. But the small grains would also multiply scatter the light! What would the output of such a powder look like? Would laser action still be possible in such an amplifying and multiple scattering material?

1 A model of a random medium with gain. The green light is multiply scattered and excites the system. The red light is both multiply scattered and amplified. The glass spheres in the photograph are typically a few mm across.

Multiple light scattering

We all know that light travels in straight lines until it hits an obstacle and is either absorbed or scattered (in which case it changes direction). But what exactly happens when light is scattered? Scattering occurs because the refractive index of the obstacle is different from that of its surroundings. The type of scattering depends on this contrast in refractive index, the size of the obstacle and its shape. If the particle is small compared with the wavelength of the light (e.g. a sugar grain, paint particle or water droplet), the light is scattered in all directions. Larger objects reflect and refract light.

What happens when we shine light on a large collection of scattering particles, for instance white paint or a bowl of sugar? In this case, the light wave is scattered by many particles and performs a random walk from one grain to the next. This phenomenon, known as multiple light scattering, occurs in almost all the objects around us. The concept of multiple scattering is not restricted to light waves, however, and applies to all wave phenomena in nature. For instance, the diffusion of sound, heat and electrons are all based upon multiple scattering.

Experiments on multiple light scattering can be performed in various ways. For instance, one can

follow the diffusion of a short light pulse through a random material in a time-resolved transmission experiment. Another possibility is to study the angular dependence of light that has been scattered, or back scattered, from a disordered sample. Back-scattered light provides information on even the deepest part of the sample (see main text). An important parameter in all experiments is the mean free path, which is defined as the average distance a light wave travels between two scattering events.

Multiple light scattering is an everyday experience and is important in many industries. Most objects around us are visible because they scatter light, and knowledge of multiple scattering is relevant when optimizing the quality of coatings, such as various types of paint. Multiple light scattering can also hamper our attempts to see objects. Early applications of this work included research to improve imaging through turbid media such as fog and clouds.

In recent years the medical community has been interested in multiple scattering as part of the effort to develop non-invasive diagnostics to image objects within the human body. Laser-based optical techniques are promising in this respect. □

And if laser action was possible, how coherent would the output be?

Disorder with gain

In 1968 Vladilen Letokhov of the Russian Academy of Sciences in Troitzk calculated the optical properties of a random medium that both amplifies and scatters light. Amplifying media are thermodynamically unstable because there are more atoms or molecules in the excited state than in the ground state. In an amplifying medium the intensity of a light wave increases as the light travels through it (figure 2). However, such a state can only be maintained in a finite region of space. Normal materials that absorb light are thermodynamically stable because, over long distances, all the energy eventually gets absorbed.

In 1994 Raymond Chiao and Jack Boyce of the University of California at Berkeley predicted further fascinating "thermodynamic" and optical properties. For instance, the energy velocity can, in theory, be larger than the speed of light in vacuum without violating special relativity.

For a long time Letokhov's pioneering work was not followed up by experimentalists. In 1993, however, Arnold Migus and colleagues at the Laboratoire d'Optique Appliquée in Paris and the authors showed that a random amplifying medium can be made by grinding a laser crystal into a powder.

It sounds simple to make an amplifying random medium: grind it and excite it. But optical excitation is

difficult for a random material because most of the excitation light is simply scattered back into the laboratory and very little is absorbed by the laser material. This means that very high pulse energies, which lie close to the thermal threshold of the material, are needed. We found that powdered titanium-doped sapphire (a widely used laser crystal) can withstand these large intensities. This crystal was ground to small particles ($\sim 10 \mu\text{m}$) and excited with very strong light pulses from a frequency-doubled Nd:Yag laser. The amplification was measured with a low-intensity "probe" pulse immediately after the excitation pulse.

Another approach is to introduce scattering particles into a laser medium. Nabil Lawandy and co-workers at Brown University in Rhode Island, US, have suspended titanium dioxide particles in Rhodamine laser dye. (A laser dye is a solution of large organic molecules that can amplify light at visible wavelengths.) In their first experiments in 1994 the scattering mean free path was of the same order as the sample size, so multiple scattering effects were not observed. By increasing the particle concentration, however, they have recently been able to increase the amount of scattering by

2 A red light wave incident from the left is randomly scattered by small particles. The wave is also amplified at each scattering event when the particles (e.g. small grains of a laser crystal) are excited with intense green light.

about two orders of magnitude.

The ability to perform multiple scattering experiments with gain – either with powdered laser crystals or micro-particle suspensions in laser dye – has opened up a whole new field of research. All the multiple scattering phenomena previously studied with passive (absorbing) materials can now be explored in amplifying systems. Moreover, the effect of strong scattering on laser action can also be investigated.

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The abbreviations of categories are: ED Editorial; NE News; FC Focus; FO Forum; L Letters; PA Physics in Action; FE Feature article; PCN Physics Community News; NP New Products; CR Careers and Recruitment. Lateral thoughts, obituaries and books reviewed are listed separately, in order of appearance.

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Diffusion laser

To see how multiple scattering and laser action interact with each other, let us start by comparing the emission from a powdered laser crystal with the emission from a regular laser. Migus and co-workers found that the diffuse light emitted from a powdered laser crystal (after excitation) can have a well defined colour, similar to that from a conventional laser. They also observed a transient spiked output when the powder was excited. This is similar to the phenomenon of spiking that occurs when some regular lasers are switched on.

However, different physical processes are responsible for the well defined wavelengths found in conventional lasers and powdered laser crystals. In a conventional laser the output is determined by an optical cavity. This traps the light, forcing it to go through the laser material many times, which yields a large overall gain. To obtain laser action, the gain must be larger than the loss. If this condition is fulfilled, a "coherent mode" builds up inside the cavity. Part of this light is coupled out of the cavity (through a partially reflecting mirror) to form a unidirectional monochromatic beam.

For a powdered laser crystal this external cavity is of course absent. However, the output can be spectrally narrow due to a process called gain narrowing. This is a general phenomenon for light propagating in a gain medium: the gain coefficient is a function of wavelength with a maximum at a particular wavelength. After propagating through the medium, light at this wavelength is amplified much more than light at other wavelengths and therefore dominates the colour of the output. Sajeev John at the University of Toronto has performed calculations on the output spectrum of a disordered laser material that confirm the effect of gain narrowing.

However, light can also be "trapped" in a disordered system due to multiple scattering. A propagating light wave will make a long random walk before it leaves the medium, and will be amplified at every scattering event. This random walk can be orders of magnitude longer than the straight line along which waves would leave the

medium without scattering. Like in a regular laser, gain can become larger than loss this way and one can have "laser action with diffusive feedback" as first suggested by

Letokhov and recently taken up again by Lawandy's group. To compare gain with loss in this case, we have to look at the size of the system. The loss through the boundaries depends linearly on the area of the boundary surface of the sample, while the total gain depends linearly on the overall volume of the sample. As a result, gain becomes larger than loss above a critical volume. In this sense light scattering with gain is similar to neutron scattering in an atomic fission bomb: the bomb only explodes if the volume of the nuclear material is increased above the critical value. When an amplifying random medium is excited such that it becomes supercritical – that is when the gain becomes larger than the losses – the intensity will grow extremely quickly. This leads to an "explosion", a strong flash of light in all directions, during which most of the energy escapes and the system becomes subcritical. However, if the excitation mechanism is still present,

the system will slowly become supercritical again. This process can repeat itself many times, resulting in a spiked output (figure 3).

3 Calculated output intensity versus time for a powdered titanium-sapphire laser crystal. The pulses are strong flashes of light emitted when the powder becomes "supercritical". The excitation pulse is shown by the dashed line.

Interference effects

As mentioned before, a major problem for multiple scattering researchers is the presence of even the smallest amount of absorption. All conventional random media absorb to some extent. Even a seemingly very white piece of paper is slightly grey when looked at closely enough. The most interesting effects occur in the longest light paths, which have the most scattering events, and these are the most sensitive to absorption. The introduction of a small amount of gain can compensate for the natural absorption, thereby making the "perfect white" system.

Moreover, if more gain is introduced, the system can become even "whiter than white"!

About ten years ago, it was discovered that sizeable interference effects can survive random multiple scatter-

4 Interference between light waves that are back scattered from a disordered material. Every light path that exits in back scattering has a reciprocal counterpart (left), and each set of reciprocal paths gives rise to an interference pattern (right). The back-scattered intensity is the sum of all these patterns. The scattered intensity at exact back scattering ($\theta = 0$) is twice the intensity at large θ .

ing. The most important example of this is coherent back scattering or weak localization. This phenomenon has been studied now extensively by, for instance, Roger Maynard and Eric Akkermans of the CNRS in Grenoble, France, by Georg Maret at the Max Plank Institut für Festkörperforschung, Germany, by Akira Ishimaru's group at the University of Washington, USA and by our group in Amsterdam.

Coherent back scattering is an interference effect that manifests itself as a doubling of the scattered intensity in the exact back-scattering direction compared with other directions. This enhancement should occur for any scattering material, but is difficult to observe in daily life because it occurs in the direction towards the light source. To understand the origin of coherent back scattering, consider a light wave that propagates along a certain path from a light source, through a random medium and then back towards the source. Because of time-reversal symmetry, this path can be followed in two opposite directions. Waves that follow the path in opposite directions acquire exactly the same (random) phase. Therefore the two waves will interfere constructively in the direction back to the light source, despite the fact that they have been randomly and multiply scattered, maybe over a thousand times!

The net effect of this constructive interference is an enhancement of the scattered intensity in the form of a narrow cone around the exact back-scattering direction (figure 4). The width of this narrow cone is inversely proportional to the scattering mean free path and is therefore a measure of the amount of scattering inside the sample. The back-scattering cone contains information about parts deep in the sample, regions which cannot be accessed with normal optical techniques. These very long light paths determine the sharp top of the cone.

Furthermore, the increase of the back-scattered intensity means a reduction in the amount of transmitted light. If we make the mean free path shorter and shorter, for instance by increasing the density of scatterers, the transmission would be reduced due to interference. In theory it should be possible to decrease the mean free path by so much that all the light would be trapped inside the sample. In other words, interference phenomena would have brought about a halt to all transport within the sample, similar to the Anderson localization of electrons in a solid. A clear observation of Anderson localization of light has not yet been reported.

What would happen to interference effects, such as

coherent back scattering, if we introduced so much gain into the medium that the gain became larger than the loss? Would the back-scattering cone survive in such a whiter-than-white system?

Last year we found that it is indeed possible to observe

5 Coherent back scattering measured from a powdered titanium-sapphire crystal for different levels of excitation or gain. The central peak becomes more narrow as the gain is increased due to the increased importance of longer light paths. The solid lines were calculated using diffusion theory.

coherent back scattering from an amplifying random medium. Moreover, we observed that long, deeply penetrating light paths become much more important with the introduction of gain: the top of the cone becomes narrower, while the wings of the cone stay behind (figure 5). This interpretation is consistent with theoretical work by A Yu Zyuzin at the University of Cincinnati, the late Sechao Feng of the University of California in Los Angeles, Zhao-Qing Zhang of the University of Science and Technology in Hong Kong, and by the authors (solid lines in figure 5).

Another, more mundane, interference effect

can occur in random systems in which the scattering particles do not move (fixed disorder). When one shines coherent light on a system with fixed disorder, the scattered light has a "grainy" intensity pattern with very bright and dark spots. At the bright spots many scattered waves interfere constructively while at the dark spots they cancel out. This effect is called "speckle" and can be observed with bulk systems (e.g. white paint) and surfaces (e.g. a dirty laser mirror). Indeed, the study of speckle from surfaces is of great interest for applied optics because the quality of many optical devices is limited by speckle. Speckle is not observed with an incoherent light source, or in scattering from systems with dynamic disorder (e.g. liquid suspensions), because the spatial intensity fluctuations are averaged out due to the movement of the scattering particles.

Last year we observed speckle patterns from a random amplifying system (powdered titanium-sapphire). This is a complicated experiment because all the scattering particles must remain fixed in their original position if we are to observe speckle. However, the large intensities required to excite the powder also heat up the system and disturb the positions of the particles. Theoretically the effect of gain on laser speckle was dealt with by Yu Zyuzin in 1995 and by Carlo Beenakker, Jeroen Paaschens and Piet Brouwer of the University of Leiden

6 An example of how random light paths could form closed loops. The light incident on the sample is not shown.

in the Netherlands.

The Leiden group considered an amplifying random medium that was embedded in a waveguide, such as an optical fibre. A waveguide restricts the amount of "modes" or speckle spots that are present. For instance, in a single-mode

fibre only one mode can propagate. The Leiden group found that due to interference, the scattered intensity from a random laser material in a fibre does not grow to infinity, even if the system is larger than its critical size. This is different from the powdered crystal systems discussed above. However, the scattered intensity increases as the fibre diameter increases because more modes can propagate. This work has important implications in telecommunication over optical fibres where absorption is a major problem.

Coherent random laser

We have discussed amplifying random media from two points of view: a diffusion laser in which interference plays no role; and systems in which randomly scattered light experiences both gain and interference. It seems natural to ask if it is possible to combine the two: can we build a coherent random laser in which the feedback is caused by interference in multiple scattering? This would require a medium in which extremely strong scattering is combined with gain.

We have found evidence that light paths can form closed loops in very strongly scattering passive media (figure 6). These loops could provide a coherent feedback mechanism. Extremely strong scattering in passive media should also lead to Anderson localization of light. Calculations by Prabhakar Pradhan and N Kumar at the Indian Institute of Science in Bangalore and by Zhao-Qing Zhang in Hong Kong suggest that the introduction of gain in a strongly scattering passive medium can help to realize the Anderson localization transition.

It is fascinating to speculate about what type of laser

such a fully coherent, but random, feedback mechanism would generate. The amount of scattering required would be very large, however, and it could be difficult to introduce sufficient gain. Nevertheless, a strongly scattering amplifying medium would be sure to illuminate even more connections between lasers and multiple scattering. Fascinating new avenues for fundamental research and important new applications would be guaranteed.

Further reading

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