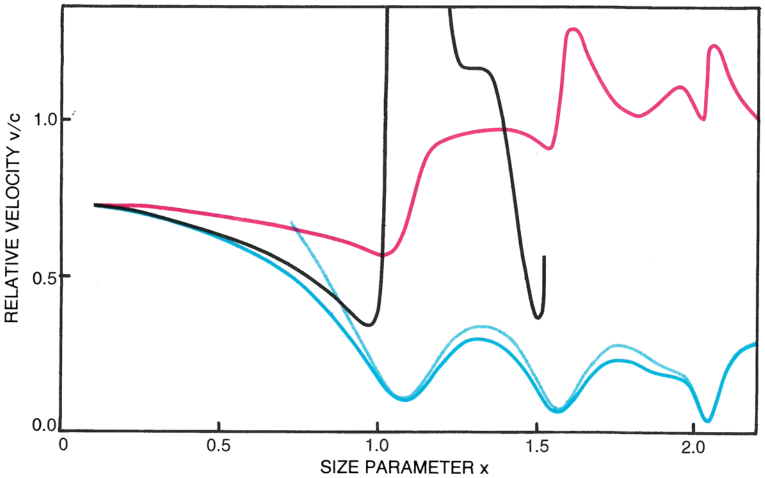
LIGHT TRAVELS MORE SLOWLY THROUGH STRONGLY SCATTERING MATERIALS

We expect light to travel through a medium at a rate given by the speed of light in a vacuum divided by the average index of refraction of the material. But a recent experiment performed in the Netherlands indicates that in some strongly scattering materials it may actually take light five to ten times longer to traverse a sample than one would expect from this simple formula for the phase velocity. It appears that the light resonates for a while with the dielectric microspheres of which the material is made so the light takes longer to travel through the sample. This result challenges some assumptions made in the analysis of data from light scattering experiments, especially searches for localized light.



Phase, group and transport velocities relative to the speed of light. These ratios were calculated for light passing through a strongly scattering medium made of dielectric spheres. The size parameter is a measure of the sphere diameter. When the spheres are the right size for the light to scatter resonantly within them, the phase velocity (red) and group velocity (black) grow larger than c. But the speed of energy transport (dark blue) remains well behaved. The second curve for transport velocity (light blue) is a more approximate calculation. (Adapted from ref. 2.)

Localization of light

Interest in light multiply scattered from random dielectrics has been piqued by the analogous phenomenon of electron scattering in disordered materials. Philip Anderson discovered in 1958 that in a sufficiently disordered material electrons can be localized: The electrons are scattered so strongly that they become confined to small regions and their transport is effectively halted. Once this behavior was recognized as essentially a wave phenomenon, researchers began to look for its occurrence in electromagnetic and even acoustic waves. (See the article by Sajeev John in PHYSICS TODAY, May, page 32.) In the case of photons, the role of the disordered material is played by a medium with a sharply and randomly varying dielectric constant, which can strongly scatter electromagnetic waves.

Localization is caused by the interference between waves scattered by the medium. As waves are scattered through the material they interfere with waves traveling other paths. When the average distance the photon travels between scattering events—the mean free path l—becomes comparable to a wavelength λ, the interference effects can affect the transport of light. Under the condition that $l \approx \lambda/2\pi$, known as the Ioffe–Regel condition, the extended normal modes can become localized. The electromagnetic energy is then concentrated in standing rather than traveling waves.

Although localization comes directly out of Maxwell's equations, it has only recently been appreciated. Previously, multiple scattering was either ignored or avoided because of the horrendous complications of keeping track of which waves interfere. Now, however, physicists are creating rather than shunning materials with random gradations in the index of refraction. Typically these are micron-sized spheres of one material embedded within another material. Because light localization is predicted to occur only within a certain range of frequencies, it is very difficult to achieve. To approach localization within real materials one must make the elastic scattering cross section large without correspondingly increasing the absorption. Experimenters must thus select just the right combination of composite materials, microstructures and packing density.

The hallmark of electron localization is the disappearance of the transport diffusion coefficient. Searches for photon localization have also focused on studies of the diffusion constant. According to the Boltzmann expression, the diffusion constant D equals $\frac{1}{3} v_t l$, where v_t is the transport velocity and l is the mean-free path. Traditionally, the transport velocity was expected to be closely approximated by the phase velocity c/n, where n is the average index of refraction. Thus a small value of D was thought to imply a small mean free path. Near the localization regime the diffusion constant gets renormalized by a factor that depends on the sample size, coherence length and absorption length.

The message of the Dutch experiment, however, is to inject some caution into how experimenters interpret the size of the diffusion constant: Because the transport velocity can be much less than the phase velocity, a small diffusion coefficient may imply a small l, a small v_t or both. The difference between the two velocities is expected to be especially great near resonant scattering—but that is just the strong scattering regime where many of the localization searches have concentrated.

One of the past experiments whose interpretation may be affected by the new results is a 1989 study by Michael Drake (Exxon Research and Engineering) and Azriel Genack (Queens College of the City University of New York) of the transmission of light through a collection of small spheres of titanium oxide in air.[1] They reported a value for the diffusion coefficient that was half the size expected, and they inferred a correspondingly small value for the mean free path.

Trapped by resonant scattering

The Dutch transport velocity experiment was done by Meint P. van Albada of the University of Amsterdam, Bart A. van Tiggelen and Adriaan Tip of the FOM Institute for Atomic and Molecular Physics, Amsterdam, and Ad Lagendijk, who has a joint appointment at both institutions.[2] Lagendijk discussed the results at the March APS meeting in Cincinnati. His group studied the passage of 633-nm light through samples of what was essentially white paint—particles of titanium oxide, whose index of refraction is 2.7, surrounded by air. The size distribution of the particles was centered at 220 nm, and the particles filled 36% of the volume.

The experimenters made two types of measurements. In one type, the researchers measured both the backscattered and transmitted light under static conditions. Because these steady-state measurements should not depend at all on the transport speed, they were used in two ways to infer the mean free path.

In the other type of measurements, the FOM–Amsterdam group varied the frequency of the laser light and measured the changes in the speckle intensity of the transmitted and reflected light. The speckles are dark and light areas resulting from the patterns of constructive and destructive interference. The intensity–intensity correlations as a function of frequency gave a measure of the time τ between scattering events. Using the time determined from these dynamic measurements and the mean free path from the static measurements, Lagendijk and his colleagues found that the value of the transport velocity, defined as $v_t = l/\tau$, was $(5 \pm 1)\times 10^7$ m/sec. The phase velocity for this material would be about 2.5×10^8 m/sec, a factor of 5 faster.

This experiment was conducted quite near resonance, with the wavelength about equal to the diameter of the titanium oxide spheres. Thus Lagendijk suggests a picture in which the light, being just about the right length to create a standing wave within the spheres, bounces around inside them for a bit before resuming the journey through the sample. Lagendijk's picture also resonates with some other studies involving scattering in spheres.

The FOM–Amsterdam group derived a theoretical expression for the transport velocity and found a value of 4.2×10^7 m/sec. As a result of their experiment, the group recommends that the predictions of theories in which diffusion constants are calculated from the phase velocity should be reconsidered. In addition, they assert, one should be very careful in inferring a mean free path from a dynamic measurement.

Many observers find the result of this experiment to be at once quite striking and quite reasonable. One classic optics text notes that both the group velocity and the phase velocity become unphysical near a resonance but provides no details about how to treat this case. Lagendijk points out, however, that a 1960 text by Léon Brillouin (*Wave Propagation and Group Velocity*, Academic Press) derives an expression for an energy velocity. Brillouin was concerned with the actual speed of transport of light through media that have some internal resonances (such as two-level atoms or classical oscillators). In that case, as in the recent experiment with standing waves inside spherical scatterers, the energy is not available for transport part of the time. Lagendijk considers his group's treatment to be a generalization of Brillouin's.

The FOM–Amsterdam group has used their microscopic theory to calculate the phase, group and transport velocities. The ratios of these velocities to the speed of light are plotted in the figure on page 17 as a function of a size parameter x. Both the phase and group velocities are larger than the speed of light for some values of x and hence become unphysical, but the transport velocity remains well behaved.

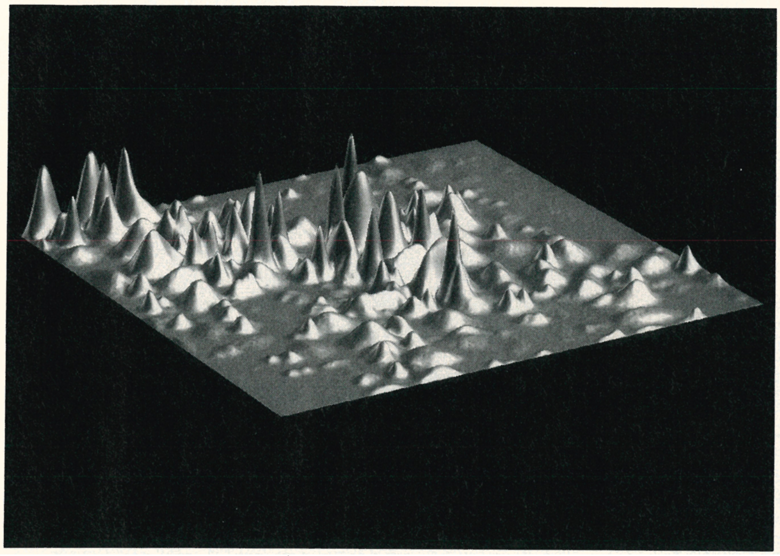
The scattering behavior is reminiscent of the phenomenon of self-induced transparency, observed by Erwin Hahn (University of California at Berkeley) and Samuel McCall (now at Bell Labs) in 1967. In both cases, the light is held up in some way as it traverses the material, but the self-induced transparency involves a nonlinear effect.

Refinements

Most observers believe that the FOM–Amsterdam group has uncovered a very real effect. However, the exact determination of such parameters as mean free path and scattering times involves some very subtle considerations. Lagendijk and some colleagues issued a caution[3] several years ago about inferring the transport mean free path from static transmission measurements: The light may suffer some internal reflection at the boundary, causing the conditions at the boundary to differ from those in the interior, where one wants to know l. Based on some systematic studies done with his colleagues at Queens College,[4] Genack has found that this effect might cause the inferred value of the mean free path to differ from its true value in some systems, but he admits that the effect may be small in the recent FOM–Amsterdam experiment. Lagendijk contends that internal reflectivity is not an important factor in his group's experiment.

Paul Fleury (AT&T Bell Labs) has some reservations about deriving the mean time between scattering events from dynamic measurements because the quantitative relation between the speckle pattern and τ is a very sensitive function of such things as absorption, boundary conditions and launch conditions.[5] Genack, however, defends the dynamic measurements, citing experiments in which he has compared measurements in both the time and frequency domains and found good agreement.[6]

The theoretical treatment of the subject by the FOM–Amsterdam group has raised some questions as well, because at first glance it appears to violate a conservation law that is known to hold for electrons. This conservation law, the Ward identity, translates for electrons into a requirement that the square of the wavefunction remain constant. By analogy, theorists analyzing the scattering of light have required that the square of the amplitude of the classical wave be



Microwave electric energy density in a two-dimensional system enables one to visualize a localized state. Microwaves entering from the left traverse a 36×27 array of dielectric cylinders, half of which have been removed at random. The peaks near the center are part of one localized mode, unrelated to the neighboring mode seen at the upper left. The experiment was performed by a group from the University of California, at San Diego, and from AT&T Bell Labs. They showed a similar figure at the March APS meeting.

conserved. Past theories have considered only the electric field amplitude. However, Lagendijk and his colleagues point out that this approach is only approximate: The conserved quantity is the energy density, which involves both the electric and magnetic fields.

Ping Sheng of Exxon Research and Engineering was excited about the Dutch result because he and his colleagues have seen what they feel is an additional example of resonant behavior in experiments on acoustical transmission through dense colloidal systems.[7] In particular they have observed an unexpected low-frequency mode of propagation in addition to the expected, higher-frequency mode. The velocity of this new mode, like the transport velocity in the recent optical experiment, is very much lower than normal. The researchers believe that it results because tails of resonant waves in adjoining scatterers couple together. While the FOM–Amsterdam group finds that light is bouncing around inside spheres, which interact *incoherently*, the Exxon team finds a *coherent* coupling between the scatterers.

The search for localization

While the interpretation of the 1989 experiment by Drake and Genack is affected by the recent Dutch result, Genack and his Queens College colleague, Narciso Garcia, have recently done an experiment to which the new caution does not apply.[8] They have studied the scattering of microwaves in random mixtures of aluminum and Teflon spheres. (See the May cover of PHYSICS TODAY.) The radiation does not penetrate the aluminum spheres, so the phase and transport velocities are expected to be equal.

The absorption present in this system prevents the experimenters from seeing the diffusion coefficient go to zero. Thus Genack and Garcia do not claim to have reached localization in the sense of a vanishing diffusion constant. However, they do report several types of behavior that are predicted for localized systems. For one, the transmission falls off as $1/L^2$, where L is the thickness of the sample. Such behavior is predicted by the scaling theory of localization to occur at the threshold of localization. The experimenters saw this scaling behavior only when the spheres occupied a certain fraction (about 30%) of the volume.

Genack feels that their experiment satisfies the Ioffe–Regel criterion for localization as well as a criterion formulated by David Thouless (University of Washington), which requires essentially that the width of energy levels within the scattering medium be small compared with the spacing between levels. However, because the absorption in the microwave system complicates the data analysis, observers would still like to see additional confirmation of localization.

A collaboration between the University of California, at San Diego, and AT&T Bell Labs has produced some intriguing maps of the electromagnetic power distributions in a two-dimensional system, in which localization is far easier to achieve. The team consists of Sheldon Schultz, Rachia Dalichaouch, John Armstrong and David Smith of UCSD and McCall and Philip Platzman at Bell Labs. In their setup, microwaves scatter from a square grid of dielectric cylinders, with the electric field vector of the microwaves parallel to the z axis of the cylinders. At the APS March meeting McCall discussed their observation of a photonic bandgap with this arrangement. (Eli Yablonovich and his colleagues at Bellcore have solved the more difficult problem of fabricating a material with a complete microwave bandgap in *three* dimensions, as pictured in John's article in PHYSICS TODAY.)

Recently the San Diego–Bell experimenters made the regular array irregular by removing half of the 36×27 array of dielectric cylinders in a random pattern. They probed the resulting electromagnetic energy distribution at frequencies where they anticipated that the system was strongly localized. The distribution clearly shows the electric energy density of the localized mode. (See the figure on this page.) There may be little surprise in finding a localized state in two dimensions, but there is great satisfaction in visualizing it.

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