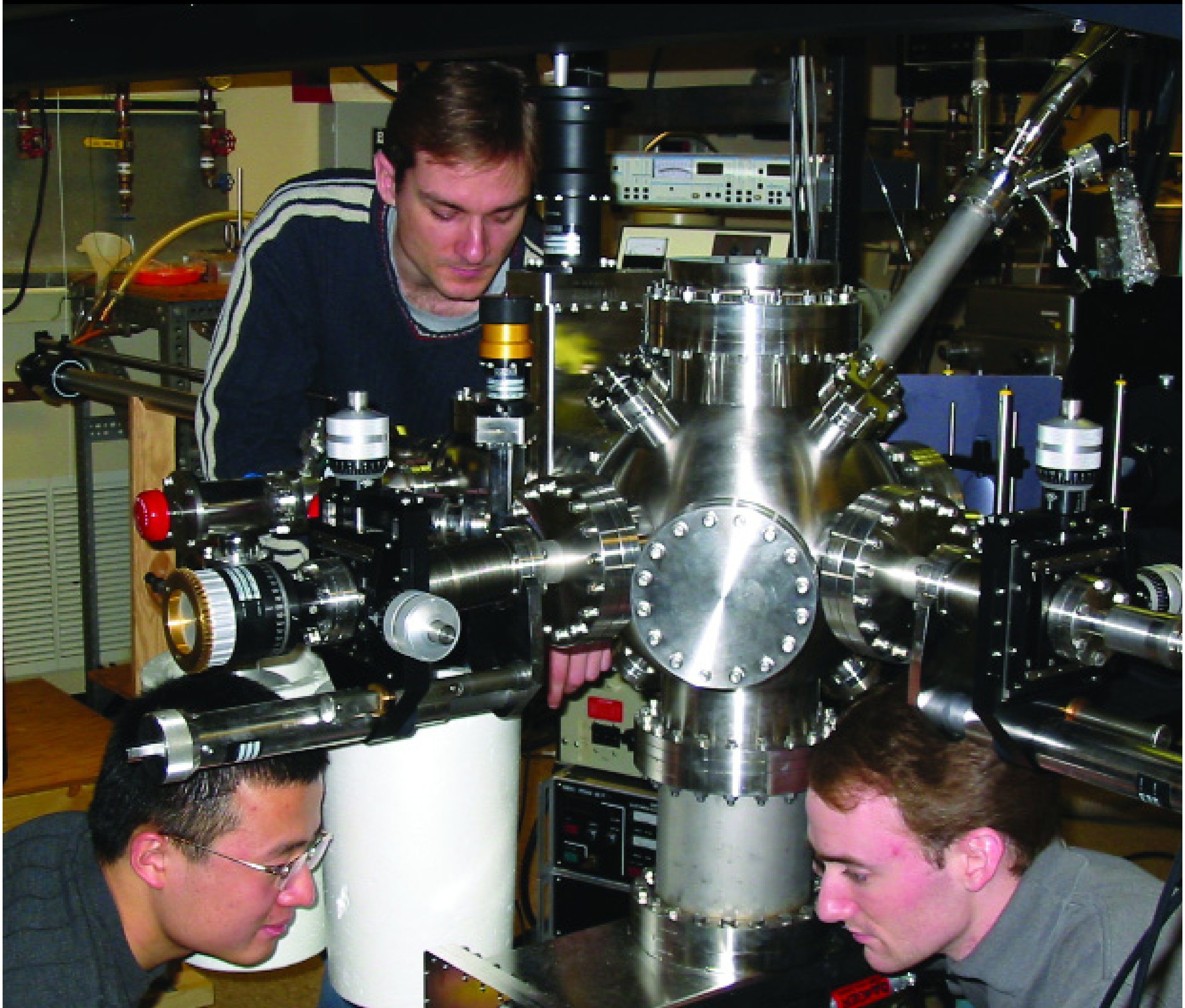

Bright Storage of Light

Stephen C. Rand

The author describes experiments related not to recent demonstrations of the slowing or stopping of propagating light, but to the direct generation of stationary light by electrical means and some of its implications for dynamic electromagnetic energy storage.

Although for many years scientists have been aware of the ease with which optical images can be stored in small volumes of transparent media, the idea of storing raw energy in optical form is quite new. Work in holography in the 1960s demonstrated that information carried by modulated electromagnetic waves could be "written" into gaseous, liquid or solid media and preserved for considerable periods of time. Very high quality archival storage of images was achieved, for example, by use of crossed beams from a laser source to "write" interference patterns (holograms) permanently into photographic emulsions. Complete three-dimensional (3D) images were retrieved by illuminating the storage medium with a beam diffracted from the interference pattern; this allowed information fixed in the emulsion at the time the hologram was created to be read out.

In the early days of holography, in a process akin to making a plaster cast of an original work of art, through photochemical reactions of silver atoms information transferred into the interference pattern was faithfully preserved as a grating structure. Reconstruction was accomplished by diffracting a probe beam from the encoded structure, much like reconstruction of a real physical object from its mold. In other light-responsive media, grating formation sometimes proceeds by very different mechanisms—such as light-induced changes of refractive index—and if the response of the medium is fast enough, or equivalently if the storage of frequency and wave-vector information has a broad enough bandwidth, even time-resolved aspects of the modulation envelope can be retrieved. In this way, entire pulse sequences have been restored after controllable delays in four-wave mixing experiments that make it appear the incident pulses themselves are slowed or stored inside the medium and then "released."

In experiments like these, it seems quite apparent that the output signals are merely delayed readouts of a diffractive structure created and preserved as a reaction product in the medium. The light wave itself is not actually slowed down, stopped or stored in the medium—only the information is stored, either as a stationary internal excitation of the medium that does not radiate (a non-dipolar polarization or, in current jargon, a "dark" polariton) or a slow-moving excitation that radiates very little (a "grey" polariton). Two pulsed fields may write a modulation pattern into a uniform medium for the lifetime of an electronic energy level or spin orientation (or for the lifetime of a chemical reaction product), but the original photons of the object wave cannot be "released" after an arbitrary delay. A delayed probe is needed to provide any output at all, and the output photons all come from the late-arriving probe wave. Since, as discussed below, however, it is known to be possible to slow down photons under coherent control, an obvious question that arises is whether light can genuinely be brought to a *complete standstill* by some means. Instead of just storing information, can we imagine ways to efficiently store significant amounts of energy in a dynamic "optical battery"? Also, can light incident on a medium or *created inside a medium* be captured in a borderline "radiative" state (as a "bright" polariton with a finite dipolar amplitude that cannot escape to the radiation zone), perhaps without using external fields at all?

Slow light

Recent dramatic advances have shown that it is indeed possible to slow light down to a small fraction of its speed in vacuum when external fields are applied in a special way to the medium through which it is traveling. The group velocity of light (i.e., $v_g = c/[n(\omega) + \omega\, dn/d\omega]$, where c is the velocity of light in vacuum and n is the refractive index) has been reduced to $c/10^7$ by tuning the incident light frequency to remarkably narrow electromagnetically induced transparency (EIT) features with exceptionally large positive $dn/d\omega$ values. Using the enormous dispersion in the vicinity of an EIT resonance, Lene Hau's group at Harvard, for example, slowed light to a mere 17 m/s. Other results achieved to date exploiting this technique in Bose condensates, solids and gases were reviewed recently.[1] When light is slowed by EIT to "bicycle speed," it is amusing to imagine that the sample could in fact (to paraphrase Ref.1) be mounted on a bicycle traveling in the opposite direction to make it completely stationary from the point of view of an observer standing nearby. Such an arrangement would clearly be awkward from the point of view of energy or information storage, requiring as it does an energy storage device balanced on a contraption of dubious mechanical integrity in precarious motion! A new approach—one that does not require a bicycle—is needed to realize dynamic electromagnetic storage that might not be limited by the lifetimes of internal excitations, provide storage without the application of external fields and preserve the original photons in the medium.

We turn now to some recent developments in the field of light scattering that offer intriguing possibilities.

Recurrent scattering and stationary light in random media

As early as 1950, Lethokov[2] discussed a different idea for restricting the propagation of light and making use of it without either using any deliberately engineered structure or applying any external control beams. His concept was that if enough gain were made available, light could be scattered by a random medium to provide enough optical feedback—despite the losses from the random walk of the field distribution—that laser oscillation could be achieved. A laser of this type would operate by spatially diffusive, distributed feedback

Facing page: (*Left to right*) Graduate students Meng Cui, Shawn Redmond and Patrick Shea working on the ultrahigh vacuum chamber in the Nonlinear and Ultrafast Laser Spectroscopy Laboratory at the University of Michigan.

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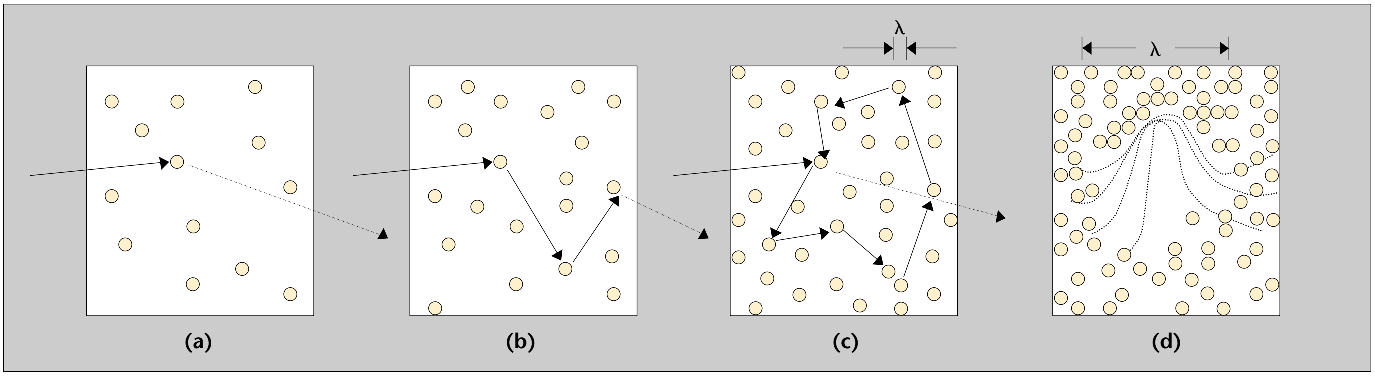

Figure 1. (a) Single, (b) multiple, (c) and recurrent scattering trajectories in a system of random scatterers and (d) a schematic drawing of the field distribution of stationary light.

rather than by reflection of freely propagating light from mirrors, and it would store light for brief periods of time. Figure 1 illustrates several important regimes of light propagation that affect the implementation and characteristics of lasers in random media with varying degrees of scattering, from those that scatter hardly at all to multiply scattering media that localize light.

Early experiments by Markushev,[3] Lawandy[4] and Noginov[5] showed that Letokhov's basic concept could be realized in practice and that recirculation of light over macroscopic trajectories within a strongly scattering medium could provide enough feedback to achieve laser threshold. Later, Cao[6] showed that when the effective size of the cavity was reduced by decreasing the scattering mean free path, a limited number of modes could be identified that mediated coherent laser action over fixed, closed trajectories. These fixed, closed trajectories are called recurrent scattering paths [see Fig. 1(c)]. The length of the propagation path through the medium is the key quantity that determines the frequency separation of allowed modes. In experiments on ZnO powders, Thareja[7] used this relationship to determine from the emission spectrum an effective cavity length that was only about ten optical wavelengths. With laser light propagating in such small, lossy loops, through these experiments a form of bright light storage had already been achieved. Yet this situation did not differ in any fundamental way from the simple transient storage of light between two mirrors—in a macroscopic, low-loss, reflective cavity—where the storage time is short and is known to be limited by finite reflectivity, absorption and diffraction around the mirrors.

This work on strong localization made it plausible that novel laser sources less than a cubic wavelength in size might be possible in random scattering systems where the transport mean free path *l was comparable to or less than a wavelength.**

A very different possibility began to emerge with the experiments of Wiersma,[8] which showed that, under the right conditions, electromagnetic transport could be "halted" in random media by elastic multiple-scattering interactions alone. Transmission experiments revealed that the field amplitude decayed exponentially in space away from the origin. When closely packed particles were reduced to a wavelength in size, light propagation underwent a transition from diffusive to strongly localized transport. By monitoring the density of states available in the scattering process directly, Chabanov[9] confirmed and clarified the conditions necessary for Anderson localization of electromagnetic waves to take place. This work on strong localization made it plausible that novel laser sources less than a cubic wavelength in size might be possible in random scattering systems where the transport mean free path l^* was comparable to or less than a wavelength.

The path length l^* sets the distance scale for directional randomization of the optical wave vector. In systems with $l^* < \lambda$, it is apparent that the coherence length for light is less than a wavelength too, since scattering events that redirect a wave also alter its phase. Light encountering the severe scattering of this transport limit cannot propagate even a single spatial period without being dephased as well as directionally randomized. In its interaction with the medium, the field is repeatedly scattered over a distance much shorter than a wavelength, so that the resulting polarization is critically damped and cannot propagate. Goodman described this odd regime in his book, *Statistical Optics*, where he remarked that a perfectly incoherent wave has infinitesimally fine spatial structure and that "spatial structure finer than a wavelength corresponds to nonpropagating evanescent waves. Hence the perfectly incoherent surface does not radiate." In the $l^* < \lambda$ limit, in the presence of gain, even amplified spontaneous emission would be generated with a coherence length shorter than its own wavelength, while at the same time being confined "losslessly" to a volume on the order of a cubic wavelength. This is a remarkable situation indeed, suggesting that "laser" action may be possible in high-quality, subwavelength cavities enclosing a localizing random medium that cannot support constructive interference!

We should be cautious in assessing this conclusion, however, because cavities in which constructive interference cannot occur might be thought by some to be incapable of supporting "laser action" in any meaningful sense at all. After all, the average transport distance would then be too short to allow rectilinear propagation of light waves across the cavity and the cavity length would be too short to support mode selectivity or coherent feedback, properties that are often associated with lasers. However, many lasers produce output that is very nearly spatially incoherent, operate with distributed feedback mechanisms or emit broadband temporally incoherent light—yet still qualify as laser oscillators. This is because they manifest the only two essential properties that distinguish oscillators from amplifiers, namely stimulated emission and feedback that balances the "cavity" losses in the active region (regardless of whether the feedback is coherent or incoherent). Hence the occurrence of stimulated emission with a threshold and feedback that result in linear output meet the physical criteria for a laser oscillator as opposed to an amplifier. Moreover, in a system characterized by a transport length of $l^* < \lambda$, the combination of *incoherent* stimulated emission and *oscillator* behavior jointly imply confinement (and amplification) of the light in a subwavelength-sized volume. Clearly nanocavity confinement again qualifies as bright storage of light, although the character of the light itself is unusual in this case, to say the least. Under these circumstances the light field in the "cavity" does not propagate at all. It merely oscillates in time at the optical frequency in one spot [see Fig. 1(d)] and might best be described as stationary light. While such circumstances lead in theory to bright storage of this stationary light, we must turn to experiments for confirmation that such an intriguing possibility may be realizable in practice.

Exciting luminescence from within

The electric fields associated with plasmon excitations in fractal *metal-dielectric* systems have been shown in recent years to manifest enormous enhancements in tiny, subwavelength regions.[10] Since the

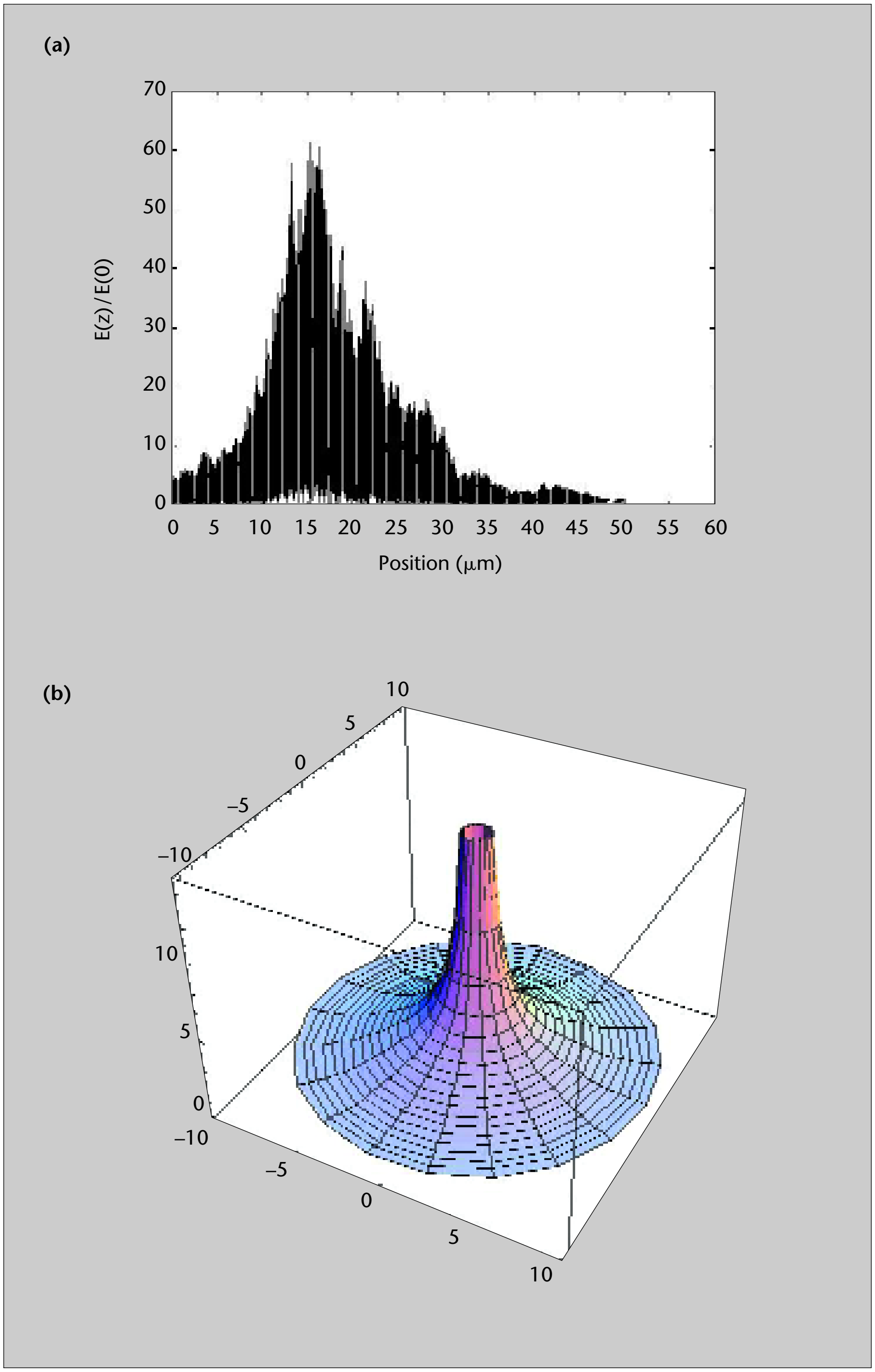

Figure 2. (a) Calculated electric-field enhancement relative to the input field E_0 inside a medium corresponding to a random configuration of dielectric particles arranged in 1D. The Gaussian nanoparticle (ε=3.06) size distribution has mean diameter ϕ=(80±30) nm and the Gaussian spacing distribution has a mean of (15±3) nm at λ=405 nm. Note that the widths of enhancement peaks and field fluctuation intervals are less than λ. (b) Distribution of the stationary light field amplitude calculated in 3D (and plotted versus x,y) around a single dipole emitter at the center of a subwavelength-sized density depression of particles in a dielectric powder. The cube sides are $2l = 0.4\lambda/\pi$, where l is the radius of the density fluctuation. In the model, the field amplitude outside the density fluctuation is less than one percent of its normal value.

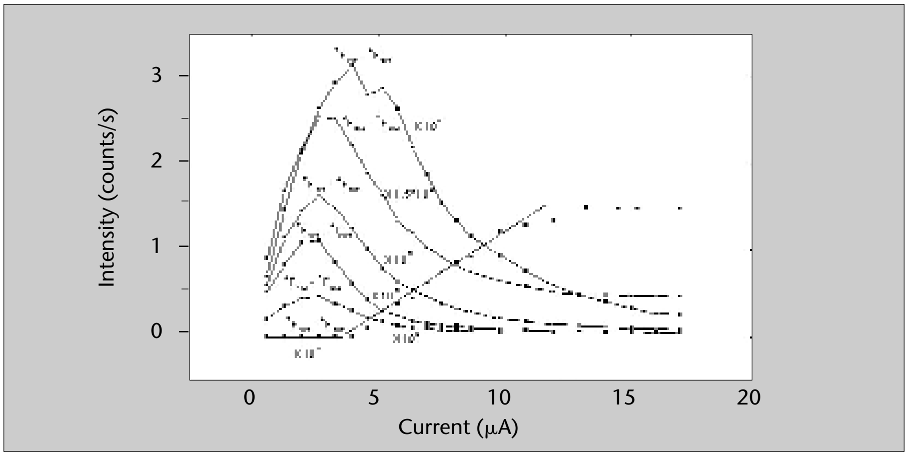

Figure 3. Luminescent emission curves from Nd^{3+}:Al_2O_3 nanopowder pumped by an electron beam in a ultrahigh vacuum chamber. The cw laser emission corresponds to the one curve that does not bend over as the pumping increases, at the onset of stimulated emission. This curve uniquely shows a large change in slope and linear output above a well-defined threshold current. Each curve is labeled with the corresponding transition assignment for the active Nd^{3+} ion.

nanoscale structure of such systems has no apparent order or obvious correlation with field-enhanced locations, one might naively hope that large optical field enhancements are possible in *purely dielectric* random media too. Remarkably, this turns out to be true, although for rather unexpected and unrelated reasons. An exact calculation for a one-dimensional medium of nanoparticles reveals that internal field amplitudes as large as 100 times the input field can arise, even when particle positions and sizes are determined by a Monte Carlo routine [Fig. 2(a)]. As shown in Fig. 2(b), subwavelength density fluctuations in random systems can achieve similar enhancement and localization of light in three-dimensions in theory—equalling the properties of carefully engineered, high-quality macroscopic cavities. The amplitude of the electric field from a point dipole situated at the origin in this model is reduced by a factor of 100 in all directions outside the near field or induction zone, compared to corresponding values for a homogeneous medium. If large theoretical field enhancements and high internal reflectivities can be realized experimentally, the way is clear to create stationary light, bright light storage and continuous-wave laser action in nanodielectrics, provided that a means can be found to excite luminescence from *within* such media.

Using pulsed, external optical fields to excite highly scattering polymer films, Vardeny[11] found that it was possible to produce broadband amplified spontaneous emission that transformed into random lasing on a few narrow lines (modes) as pumping was increased. Ling[12] observed that the number of such modes increased dramatically as the transport mean free path approached the wavelength of light, at the boundary between the diffusive and "strong" scattering regimes. Consistent with these observations, but with an altogether different (internal) pumping scheme in media with transport mean free paths less than a wavelength,[13] it was recently demonstrated at the University of Michigan that continuous-wave (incoherent) laser action could be achieved by use of electron beam pumping in a rare-earth-doped oxide powder. In experiments on Nd^{3+}:Al_2O_3, Pr^{3+}:Al_2O_3 and Ce^{3+}:Al_2O_3, with particles only a fraction of a wavelength in size, stimulated emission was observed to follow a linear dependence on excitation rate above an abrupt threshold (see Fig. 3). Despite the short propagation and coherence distances measured in these powders by light scattering techniques ($l^* < \lambda/2$), cavitylike feedback evidently played a significant role in the emission process. Without feedback, the output intensity should grow exponentially—not linearly—with increasing pump rate, provided the gain coefficient is large. If the gain coefficient were exceedingly small, a process of pure amplification would not cause an appreciable change of slope at the inversion threshold, although the output would be linear. The data in Fig. 3, on the other hand, show an abrupt threshold with a very large change of slope and linear incoherent output, unlike either of the amplification limits described above. One is thus led to conclude that oscillation with feedback takes place inside subwavelength regions of the random medium.

A need for further studies

No evidence of frequency selectivity has been found in the spectra of continuous-wave random lasers, nor has any laser speckle been resolved. While this is unexpected for any kind of coherent emission, it is entirely consistent with incoherent laser action. Hence, characteristics of continuous-wave random lasers that at first appear puzzling can be reconciled with known laser physics if the transport distance that limits the cavity size is less than a wavelength, and if all modes of space participate in the process through scattering-induced mode mixing.

These developments raise a number of intriguing questions. First, what possibilities exist for fundamentally new, high-quality 3D nanoresonators in dense systems of subwavelength-sized particles? It seems almost inconceivable that any randomly assembled nanosystem could compete with deliberate Bragg structures or conventional mirrors to produce highly reflective cavity confinement. Yet this is what the experimental results and model calculation of Fig. 2(b) indicate. Can these properties be reproduced by numerical techniques in 3D? If light is generated in a nonpropagating or localized form in subwavelength-sized regions, how does it emerge from nanocavities?

Questions like these call for electron beam studies in which the accelerating voltage of the pump, and therefore the penetration depth of the electrons impinging on the sample surface, can be varied. Some experiments along these lines have already been performed at

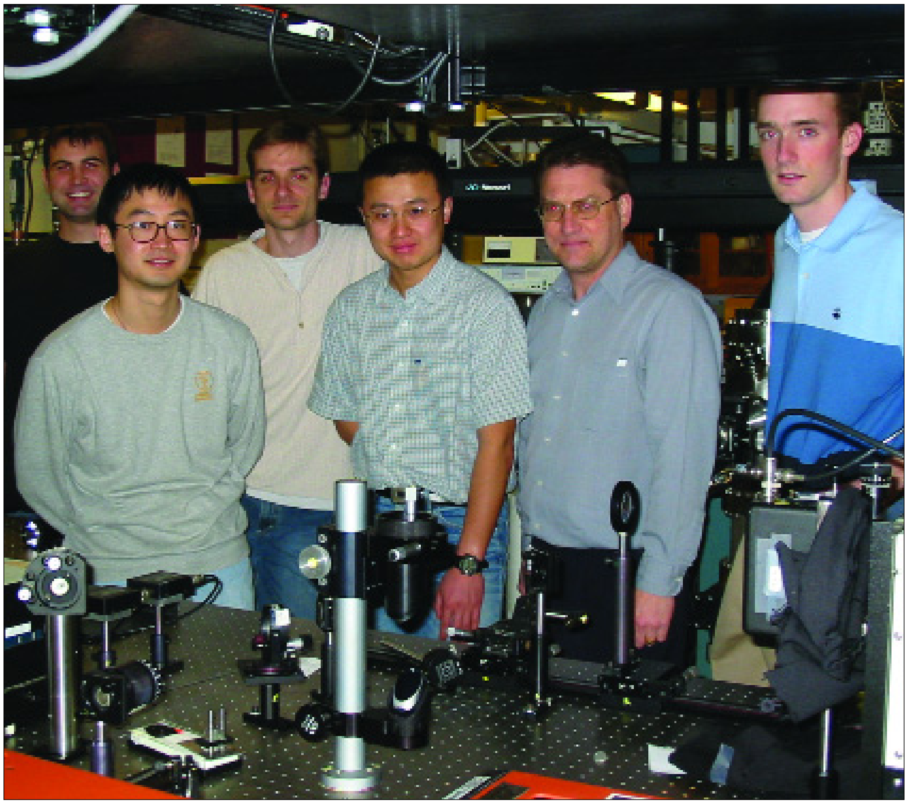

Figure 4. *(Left to right)* Graduate students Jeremy Potts, Bin Li, Shawn Redmond, Meng Cui, Professor Rand and Patrick Shea in the Nonlinear and Ultrafast Laser Spectroscopy Laboratory at the University of Michigan.

Michigan, and the slope efficiency of laser output is found to *improve* as light is generated at progressively greater depths from the surface. Hence it can be inferred that optical confinement is compromised near the powder surface by the proximity of free space, allowing the wings of localized light distributions to radiate. Light leaks out of near-surface "cavity" regions, where the vacuum interface slices through it, through what amounts to a voltage-controlled output coupler. In general, some light in the wings of the cavity distribution may also couple weakly to propagating modes outside the cavity region, where the particle distribution may be different. This is in stark contrast to information storage by four-wave mixing processes or dark storage in EIT experiments. Dark coherences—even those located only a few wavelengths from a vacuum interface— cannot radiate or couple in this way because there is no oscillating dipole in the medium.

Time limits of storage

If stimulated emission within subwavelength feedback cavities forces upon us the notion of generation of stationary light—light that is severely delayed in escaping from its own near-field region—it seems natural to ask how long light could be stored in this form in practice. Could it be stored longer than a beam confined between super mirrors? Slow-moving or stationary light cannot couple to transverse phonons in the customary way to form conventional polaritons, so one might guess that storage times could greatly exceed the excited-state dephasing times that typically limit dark polariton systems.

While this is at least partly true, it overlooks the fact that in the near-field region of the electromagnetic field, longitudinal components can couple to longitudinal phonons within the host particles, forming a bright polariton with a lifetime inversely proportional to the coupling energy. The coupling of the emitted field to the surrounding medium might be sufficient to prevent resonant reabsorption of the field by luminescent impurities. If not, the coupling between the bright polariton and local impurities would initiate a periodic exchange of energy between them, ultimately facilitating internal decay within the atoms. One or the other of these two quantum electrodynamic loss mechanisms is likely to limit the lifetime of these strange "optical batteries." Could the battery lifetime nevertheless be extended to seconds or hours by the use of nanocavity arrays? Is optical gain necessary or advantageous for efficient storage? We are only beginning to address many of these questions, so clear-cut answers will no doubt elude us for some time. What we can confidently predict is that the search for the road to bright storage of light will yield many exciting surprises in the future.

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