

The extremely cold ensemble of atoms has immediate benefits for EIT-induced optical effects. At these very low temperatures, the thermal motion of the atoms is essentially eliminated. "In all geometries, there is no Doppler shift," explains Hau, "and so we can use very low coupling powers." The steepness of the refraction dispersion, and the resulting effects such as reduced group velocity, actually increase with decreased coupling laser power, as long as the coupling laser is strong enough to overcome any Doppler broadening. The experimenters therefore saw their greatest effects with their smallest coupling laser power and their coldest temperature, 50 nK. Also to their advantage is the high number density of the atoms at these low temperatures, on the order of 10^{12} – 10^{14} atoms/cm³. The steepness of $n(\omega)$ is proportional to the density.

Hau and her coworkers saw significant effects above the BEC transition temperature, with group velocities of about 100 m/s for temperatures of a few μ K. As the temperature is lowered past T_c , though, there is a marked decrease in the group velocity due to the increase in atom density when the condensate is formed.

Some like it hot

The ultracold temperatures that Hau and her colleagues achieved with their sodium atoms are not necessary for achieving such dramatic optical effects, if one accepts restrictions on the experiment's geometry and carefully controls other experimental parameters. Through such means, both Scully and his coworkers and Budker's group have seen results like Hau's at room temperature and above.

At such warm temperatures, Doppler broadening of the atomic transitions can kill the effects. To overcome this hurdle and obtain the desired steep dispersion, the probe and coupling lasers must be collinear and have nearly the same frequency. Then, because the physics is mostly governed by the difference in the coupling and probe frequencies seen by the moving atoms, the response is essentially independent of the atoms' velocities. Thus most of the effects of Doppler broadening on the EIT resonance can be avoided.

Scully and his coworkers use a rubidium vapor whose density is comparable to Hau's, a few times 10^{12} atoms per cubic centimeter. They also carefully control the amount of neon buffer gas in their sample and the size of their laser beams to reduce the ground state decoherence rate. With this preparation, they have observed a propagation delay of 0.26 ms for an amplitude-

modulated probe beam passing through the 2.5 cm length of their sample, which corresponds to a group velocity of about 100 m/s.

Budker's group has achieved an even slower group velocity, 8 m/s, in a rubidium vapor at room temperature. In their experiments, the states |1) and |2) are degenerate but couple to different laser polarizations. A brief rotation of the linear polarization of the coupling laser has the effect of sending in a probe pulse of orthogonal polarization. The coupling and probe beams are therefore inherently collinear. A paraffin coating on the walls of the sample cell makes the ground state coherence very long-lived by suppressing relaxation during atom-wall collisions. Although the rubidium number density is low, Budker and colleagues have observed pulse delays of up to 13 milliseconds. They are restricted, however, to very long pulses, which limits the pulse delay to a small fraction of the pulse width. The nonlinear optical processes in the EIT medium, says Budker, are closely related to nonlinear magneto-optic (Faraday) rotation, the primary focus of investigation of his group.⁵

A unique nonlinear medium

The remarkable optical properties of these ensembles of atoms have additional appeal due to the nonlinear effects produced by the dispersion, recently discussed by Harris and Hau.⁶ "These are the largest nonlinearities ever produced," explains Harris. With them, it may be possible to perform nonlinear optics at the single-photon level. Compared to current nonlinear optics materials, says Scully, "the nonlinear couplings are millions of times larger." Because of its low efficiency, nonlinear optics at present requires very large laser powers. With such

greatly improved efficiencies, the energy density requirements may drop down to the order of nJ/cm². (For comparison, the energy density of current optical fibers is about 1 J/cm².) Using milliwatt diode lasers, Scully and his colleagues have already produced one example: A new field, coherently generated by wave mixing within their EIT medium, had an amplitude comparable to the transmitted probe beam.

The steepness of the dispersion opens the door to new ways of controlling and monitoring optical properties. For instance, Hau and her coworkers demonstrated in their sample a giant Kerr nonlinearity, in which the phase of one laser beam is controlled by the amplitude of another. This ability could form the basis for an all-optical switch. The steepness may also lead to magnetometers of greatly improved sensitivity.

In addition to being slowed upon entering an EIT medium to a fraction of the speed of light in vacuum, light pulses are compressed spatially by the same fraction. Thus the length of the 2.5 μ s pulses of Hau and company was reduced to as short as 40 μ m in their sample. Hau notes that such pulses may find application in the probing and manipulation of Bose-Einstein condensates.

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Progress Made in Near-Field Imaging with Light from a Sharp Tip

Researchers are striving to image objects optically at smaller and smaller scales. For example, they'd like to determine the chemical identity of individual molecules and examine the optical properties of semiconductor nanostructures. The required resolution is finer than that allowed by diffraction, which sets a lower limit of half the wavelength of light (hundreds of nanometers for visible light). But much higher resolution can be achieved by viewing objects at distances closer than the wavelength of light. Such near-field imaging has been hotly pursued since the early 1990s, when researchers demonstrated its promise for

Recent experiments, on realistic samples, take us further toward the goal of studying details smaller than 10 nm in objects that either emit light or absorb it.

imaging single molecules (see *PHYSICS TODAY*, May 1994, page 17 and November 1997, page 67). As Dieter Pohl (University of Basel) puts it, "Imagine the potential of microscopy that combines the resolving power of an electron microscope with the enormous spectral resolving power of light."

The standard way of accomplishing near-field microscopy is currently to

shine light onto a sample through a metal-coated optical fiber, tapered to a small opening. If the fiber end approaches the surface at distances less than a wavelength of light, the object is illuminated only by near-field radiation and the spatial resolution is not subject to the diffraction limit. A detector collects the reflected or reemitted light as the fiber scans the surface. This technique of near-field scanning optical microscopy (NSOM) yields pictures of the sample whose resolution is roughly the width of the fiber—on the order of tens of nanometers for images in the visible.

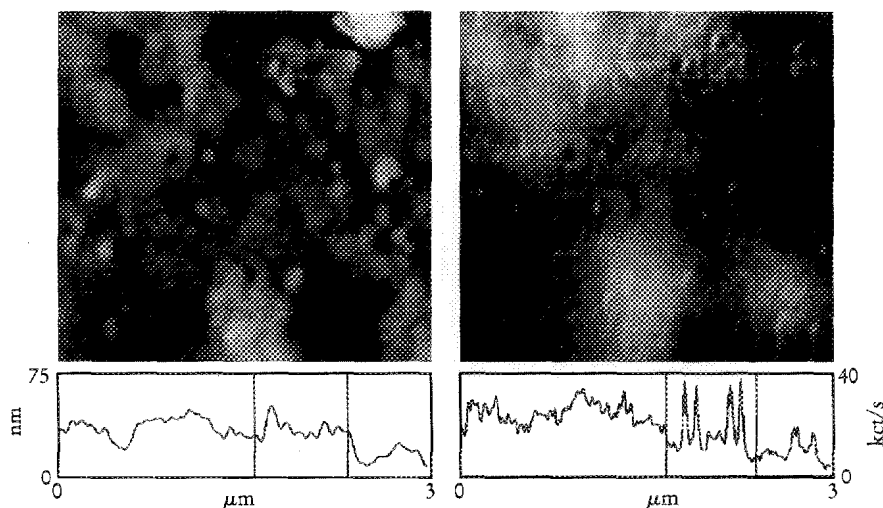
It's also possible to do near-field imaging without an aperture such as a tapered fiber. The basic idea of the apertureless approach is to shine radiation on the metal or semiconducting tip of a scanning probe microscope, from the side or, perhaps, from below. The radiation is enhanced at the very tip of the probe, much as the electric field is intensified at the point of a lightning rod. In this apertureless method, the sample is illuminated by the light from the irradiated scanning metal tip. Apertureless NSOM can be combined with scanning probe microscopy.

Research groups are taking the apertureless technique in a number of different directions. Two recent experiments—one¹ combining apertureless NSOM with two-photon fluorescence to study photosynthetic membranes and the other² measuring the mid-infrared absorption of polymers—have attracted particular note because they come closer to actual realization of high optical resolution on samples of interest.

Comparative advantages

In principle, the resolution achievable with aperture-based NSOM is limited only by the diameter of the fiber one can make. In practice, however, its potential is restricted by the exponentially diminishing amount of light that can get through a very narrow aperture. The resolution is also limited by the distance that light from the fiber penetrates into the fiber's cladding, which is typically a few tens of nanometers.

These barriers to higher resolution led experimenters to try the apertureless approach, both with particles³ and with sharp tips. But the work was hampered by the large background: One wants to image only that point on the sample immediately below the probe tip, where the light intensity is greatly amplified (by factors of perhaps hundreds or thousands), but the light shining on the probe tip inevitably illuminates a larger region of the sample. Although light scattered into the detector will come predominantly from the spot directly below the tip, some



TWO-PHOTON FLUORESCENT IMAGE (right) and topography (left) of photosynthetic light-harvesting complexes in a cellular membrane, both taken with the same atomic force microscope. The fluorescence was stimulated by light reflected off the probe's tip. The cross sections at the bottom show the surface height in nanometers (nm) and intensity in kilocounts per second (kct/s) measured along the horizontal dotted line. In the box in the upper right, four peaks—thought to be proteins—are seen only in the fluorescence image. (Adapted from ref. 1.)

light is also scattered from other regions as well. In 1994, Kumar Wickramasinghe and his colleagues at the IBM Research Center in Yorktown Heights, New York, achieved 1 nm optical resolution by implementing one idea for circumventing the background.⁴ They oscillated the tip up and down, perpendicular to the sample surface. Light reflected from a semiconductor tip was modulated at the same frequency and thereby distinguished from the background light scattered from the (unmodulated) incident beam. Independently, groups from Osaka University⁵ and from the Ecole Supérieure de Physique et Chimie in Paris⁶ used similar techniques to image surfaces, although their resolutions were not as good.

In addition to scattering studies, experimenters are turning to such incoherent processes as fluorescence or Raman scattering. In these processes, the background scattering is reduced because the signal detected has a different wavelength than the incident light. Nevertheless, if the area illuminated by the incident radiation is 100 times greater than the spot of interest, then the field under the tip has to be at least 100 times more intense for the signal to exceed the background. That's why experimenters are interested in nonlinear approaches, in which the probability of exciting a molecule no longer depends linearly on the intensity of the radiation, but has a quadratic or higher dependence.^{7,8}

Two-photon fluorescence

One such nonlinear process is fluorescence excited by two photons; the mole-

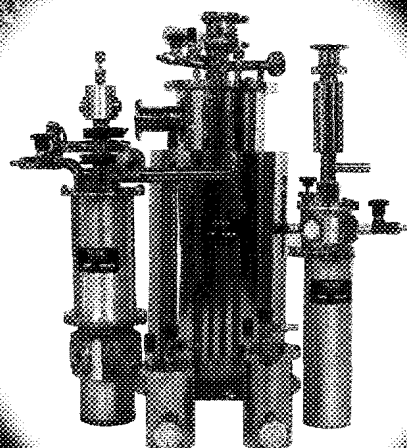
cule must absorb both photons, each roughly half the frequency of the absorption line, before it will fluoresce. The probability of excitation is proportional to the square of the intensity of the radiation bathing the sample. Erik Sánchez, Lukas Novotny, and X. Sunney Xie, working at the Pacific Northwest National Laboratory, combined such two-photon fluorescence with apertureless NSOM to image photosynthetic proteins with a resolution of 20 nm.¹ That's a slight improvement over fiber-based NSOM, but researchers hope to do even better. (Sánchez and Xie, who were also affiliated with Portland State University, are now at Harvard University; Novotny is at the University of Rochester.)

Xie and his colleagues demonstrated the two-photon fluorescence with two types of molecules. The figure above shows the image of photosynthetic light-harvesting complexes embedded within a cellular membrane; the group is interested in the spatial arrangement of those complexes within the membrane. The figure shows the topography determined by a metal-tipped atomic force microscope (left) and the fluorescent image made by illuminating the tip on the same instrument (right). The cross sections measured along the dotted lines, displayed at the bottom, indicate that the fluorescent image contains details not seen in the topography. The experimenters highlighted four peaks (see boxes near the upper right) which they believe correspond to protein molecules.

Fluorescent techniques suffer from a problem known as fluorescence quenching, in which the metal tip ab-

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sorbs the fluorescent radiation, especially when the tip is brought very close to the surface. Fortunately for Xie and his colleagues, in both the molecular complexes they studied, this problem is circumvented because the fluorescence emission occurs far from the metal tip. Two-photon fluorescence is also subject to photobleaching.

Infrared absorption

Not all molecules fluoresce, but all of them absorb in the infrared. IR absorption is thus a standard technique for identifying molecules. Far-field IR imaging, however, bumps up against the diffraction limit of several micrometers, and traditional NSOM hits a snag because it's hard to couple long-wavelength light into a fiber that is tapered to nanometer scales. Researchers have thus turned to apertureless techniques and obtained resolutions in the IR below 1/100th of a wavelength in scattering studies.⁹

Recently, Bernhard Knoll and Fritz Keilmann of the Max Planck Institute for Biochemistry in Martinsried, Germany, determined the vibrational absorption image of a sample with 0.1 μm resolution, 100 times smaller than the 10 μm incident radiation.² They examined the absorption as a function of wavelength to show that the response was consistent with the expected vibrational absorption resonances.

For their sample, Knoll and Keilmann embedded one polymer in the substrate of another. They found that the embedded polymer (known as PS) appears darker than the substrate when viewed at an incident wavelength of 9.66 μm , and lighter when seen at 10.17 μm . That agrees with expectations that PS is more (less) absorptive than the substrate at the shorter (longer) wavelength. Knoll and Keilmann believe that their measurements indicate that the polymer absorption is being enhanced by a dipolar coupling via the concentrated field below the tip: The absorption they observe is greater than calculated, based on the distance that the field from the tip penetrates into the sample. The topology they record is independent of the incident wavelength, and it manifests some features not seen in the absorption image.

Other approaches

Other researchers are exploiting in different ways the basic idea of field enhancement near a highly curved surface. One example is its use in second-harmonic generation, a nonlinear process involving the excitation of a highly dipolar molecule with a polarized field. Extending their own earlier work, a group of experimenters at the Hebrew University of Jerusalem recently re-

ported using second harmonic generation to study cell membranes.¹⁰ Rather than scanning the surface with a probe, they attached a tiny gold sphere to an antibody and let the antibody direct the sphere to a particular group of membrane proteins. The illuminated sphere stimulated a response from dipolar molecules in the cell membrane. Only asymmetrically distributed molecules contribute to the signal.

In yet another approach, the tip of a scanning probe microscope is not illuminated directly at all. Rather, the tip is placed in the evanescent field that is created just outside a glass surface within which light is being totally internally reflected. As the probe is lowered toward the surface, the evanescent field is concentrated at the tip, enhancing its interaction with the sample. Using the approach, Hendrik Hamann, Alan Gallagher, and David Nesbitt (the University of Colorado, JILA, and the National Institute of Standards and Technology, all in Boulder), have claimed high sensitivity and spatial resolution for resonant scattering images of gold nanospheres.¹¹ The same method could be used for single-photon molecular fluorescence studies as well.

The hope of many is that apertureless near-field imaging can be used for the chemical identification of individual molecules by Raman scattering; evidence suggests that such scattering may be greatly enhanced near or on the surface of a sharp tip.

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