Diffusion Waves and their Uses

Diffusion waves lack wave fronts, can't be beamed, and don't travel very far, yet they form the basis of several new and revolutionary measurement technologies.

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The concept of waves is an integral part of our scientific culture and has nourished physicists, pure and applied alike, for centuries. Many important discoveries in physics, including quantum mechanics, have involved wave phenomena. The wave concept owes some of its scientific success to its mathematical tractability. Linear wave equations—the sort that describe the transmission of sound and radio waves through air—contain a nonzero second-order time derivative, which gives rise mathematically to the rich and familiar array of properties we associate with waves, such as wavefront propagation, reflection, and refraction.

There exists, however, a peculiar class of waves whose time derivative is only first order. Called diffusion waves, these wave-like disturbances involve the coherent, always driven, oscillations of diffusing energy or particles. They have complex wave vectors and do not exhibit square-law behavior.[1]

Diffusion waves have been known about since the days of Anders Jonas Ångström in the mid 19th century (see box on page 30). However, significant progress in their science and associated technologies has occurred only recently.[2] Now, their generation and detection in condensed and gaseous phases of matter form the basis of significant advances in the measurement and understanding of materials' optical, electronic, and thermal properties. Technologies based on diffusion waves have already improved biomedical diagnostics and the fabrication of optical and electronic devices. Diffusion waves have also spawned novel high-precision analytical techniques.

The mathematics of diffusion waves

Mathematically, diffusion waves arise when the classical diffusion equation is coupled to an oscillatory force function:

$$\nabla^2\Psi(\mathbf{r},t)-\frac{1}{D}\frac{\partial}{\partial t}\Psi(\mathbf{r},t)+F(\mathbf{r})\Psi(\mathbf{r},t)=q(\mathbf{r})e^{i\omega t},$$

where the driving force $q(\mathbf{r})e^{i\omega t}$ generates oscillatory solutions $\Phi(\mathbf{r},\omega)e^{i\omega t}$ for the field's wave function $\Psi(\mathbf{r},t)$. Here, D is a transport property of the medium, usually a diffusivity. $\Psi(\mathbf{r},t)$ may be an oscillating temperature (such as a thermal wave resulting from the optical heating of a medium), or an oscillating charge carrier density (such as a harmonically-photoexcited carrier plasma wave in a semiconductor), or a multiply-scattered photon fluence rate (such as a diffuse photon density wave in a turbid medium).

By applying a Fourier transform, we easily obtain a sort of pseudo-wave Helmholtz equation:

$$\nabla^2\Phi(\mathbf{r},\omega)-\kappa^2(\mathbf{r},\omega)\Phi(\mathbf{r},\omega)=Q(\mathbf{r},\omega),$$

where $\kappa(\mathbf{r},\omega)$ is the complex diffusion wavenumber.

The term $F(\mathbf{r})$, which is usually a constant, is the square of the characteristic decay length of the diffusion wave. In a photoexcited electronic medium, this length is the carrier diffusion length—that is, the distance a free carrier travels before it recombines with a carrier of the opposite sign (a hole with an electron) and disappears in the medium (ceases to contribute to electrical conduction). In a diffuse photon density medium (for example, milk with a strongly scattered laser beam propagating in it), $F(\mathbf{r})$ represents the distance a photon will travel under random motion until it is absorbed by the medium. For thermal waves, there is usually no delay in the energy conversion process, so $F(\mathbf{r}) = 0$.

Much of the physics of diffusion waves is embodied in the wavenumber. For thermal waves, $\kappa = (1 + i)/L(\omega)$, where the diffusion length $L(\omega)$ is given by $(2D_t/\omega)^{1/2}$ and D_t is the thermal diffusivity. For most other diffusion waves, the real and imaginary parts of the wavenumber are unequal—a fact that has important consequences in the spatial distribution of the wave field. If the real and imaginary components of the wave vector are equal, the waves appear at *all* frequencies. But if they are unequal, the imaginary part does not pick up enough strength to appear until relatively high frequencies are reached. As a result, at low frequencies there are no waves, and the wave field is equivalent to a DC signal whose magnitude oscillates in phase with the source everywhere in the medium. Once a critical frequency is reached—such as the inverse of the electronic recombination lifetime in a semiconductor—then the wave nature kicks in and a phase lag in the spatial coordinate appears.

As you might expect, diffusion waves are heavily damped—a feature they share with damped waves of the nondiffusive sort, such as electromagnetic radiation in dissipative (that is, conductive) media. For these waves, $\kappa = (1 + i)/\delta(\omega)$, where $\delta(\omega)$ is the skin or penetration depth. Diffusion waves also resemble waves attenuated

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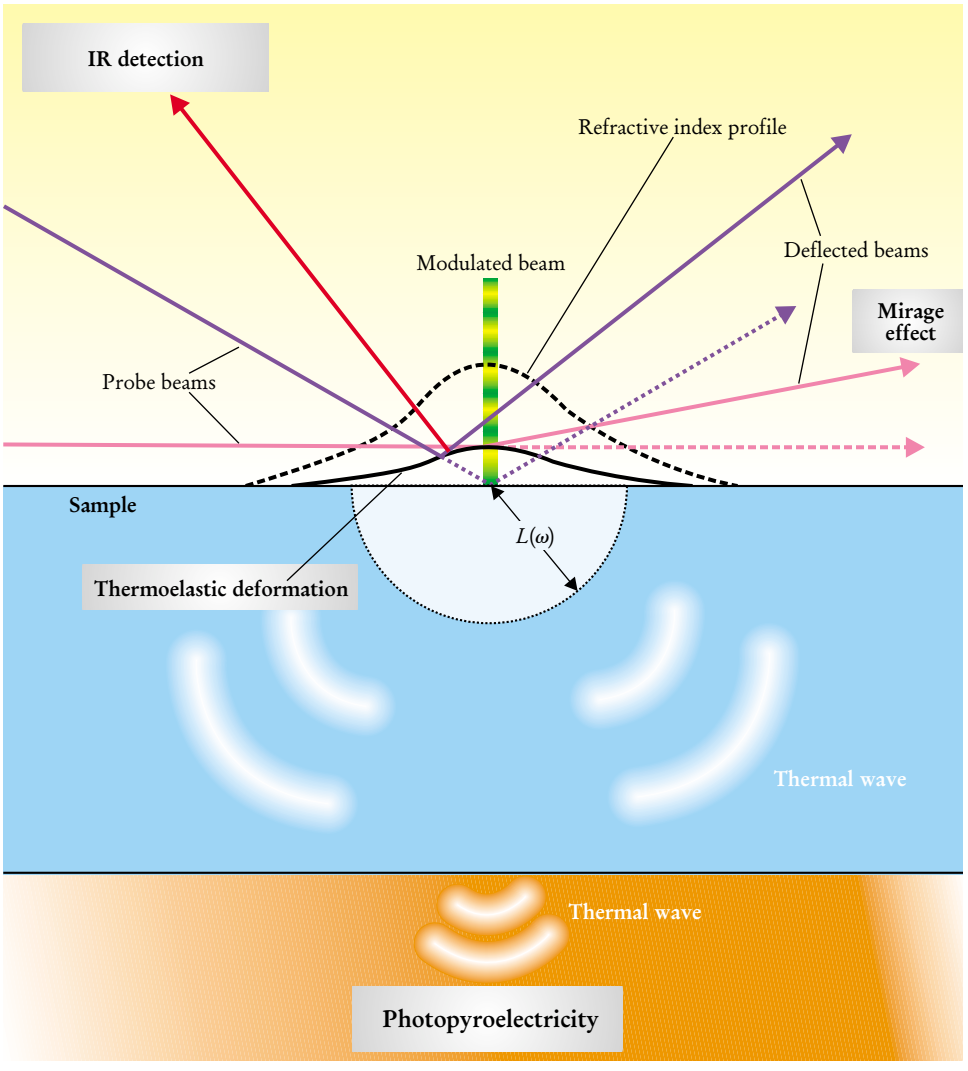
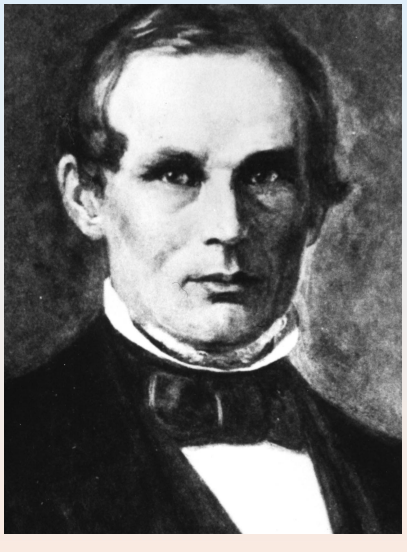


FIGURE 1. PHOTOTHERMAL diffusion waves can be generated in several ways. When a modulated laser beam (shown in green) strikes a surface, it generates a thermal wave field, which, in turn, causes a refractive index gradient to appear. A probe laser beam traveling parallel to the surface (pink) will be deflected harmonically, a phenomenon known as the mirage effect or photothermal deflection spectroscopy (PDS). Another source of deflection is the thermoelastic deformation bump generated by intermittent laser heating and thermal expansion. A probe laser beam directed at the surface (purple) will be deflected by the bump. Blackbody radiation (red) may also be intercepted from the thermally oscillating surface, a technique called infrared photothermal radiometry (PTR). A thermal wave field is generated inside the medium with characteristic skin depth $L(\omega)$. This wave can be detected with a pyroelectric sensor made from a material such as polyvinylidene fluoride—a technique known as photopyroelectric spectroscopy (PPES).

Early Studies



Anders Jonas Ångström (shown here) was the first to publish, in 1861, an experimental and theoretical study of diffusion waves.[11] In this pioneering work, he calculated the thermal diffusivity of solids as measured by periodically heating a long bar and then detecting the alternating temperature field at a point in the bar some distance away from the heat source.

In 1880 Alexander Graham Bell observed that intermittently chopped sunlight incident on a strongly absorbing substance causes audible sound to emanate from the substance.[12] But he did not connect his discovery, dubbed the photoacoustic effect, to Ångström's mathematical treatment of the heat diffusion wave. Bell subsequently developed an instrument, which he named the photophone (a differential spectrometer of sorts), to investigate the audibility of various substances in the solar spectrum. He used a sewing machine to construct the driver of the light chopper. Much activity followed the early excitement and the effect was attributed to the generation of sound by the thermal bending of a solid.

It was not until almost a century later that the photoacoustic effect was put on the correct theoretical foundation by Allan Rosencwaig and Allen Gersho, who explained the audibility by means of an acoustic piston in the gas surrounding the optically absorbing solid.[13] The acoustic oscillation is generated by the expansion and contraction of a boundary layer created by thermal diffusion waves from inside the solid, following the absorption and optical-to-thermal (nonradiative) energy conversion of harmonically chopped radiation. The physical picture of the effect was completed by Allan McDonald and Grover Wetsel Jr, who showed that the piston-like motion of the gas boundary layer adjoining the sample is superposed on the mechanical vibration of the sample surface to give a composite piston displacement.[14]

by absorption, such as acoustic waves propagating in viscous media, whose complex wavevectors have unequal real and imaginary parts.

However, despite these similarities, diffusion waves differ in one very important respect from the more familiar sort of wave: Their transport, which takes the form of spatial diffusion gradients, obeys a linear law, rather than a square law. In general, if the energy density or particle concentration generated by a source oscillating at angular frequency ω in a medium of diffusivity D is designated $\rho(\mathbf{r},\omega)$, and if the field gradients are not too large (so that a linear approximation can be used), the resulting current density $\mathbf{J}(\mathbf{r}, \omega)$ is given by Fick's law of diffusion: $\mathbf{J}(\mathbf{r},\omega) = -D\nabla\rho(\mathbf{r},\omega)$.

The simple fact that the diffusion wave field propagates according to a linear law affects the waves' behavior at interfaces. When they encounter an interface, diffusion waves obey an accumulation–depletion law, rather than the reflection–refraction law of normal waves. Because detecting diffusion waves almost always involves the waves' crossing an interface of some sort, and because diffusion waves, being heavily damped, don't travel very far, their behavior at interfaces is of great practical importance.

Further exploration of the diffusion-wave equations yields the physical artifact of infinite speed of field propagation, though with vanishingly small amplitude at remote locations away from the source. This strange property, which correlates very well with experiments,[2] results in sudden perturbations over entire domains.

Even stranger properties emerge. Because propagation is instantaneous, the equations yield no traveling waves, no wavefronts, and no phase velocity. Rather, the entire domain "breathes" in phase with the oscillating source. In the world of diffusion waves, there are only spatially correlated phase lags controlled by the diffusion length. And in isotropic media, no field directionality exists; unlike ultrasonic or laser beams, diffusion-wave beams cannot be launched in a particular direction.

Although the physics of diffusion waves differs fundamentally from that of conventional waves, the substantial deviations from the conventional reflection and refraction laws mostly occur at large angles away from normal incidence.[1,3] As a result, theoretical developments to date have been largely built on assumptions borrowed from conventional square-law behavior.[2,4] However, regardless of the often misleading conventional-wave language used to describe diffusion waves, these disturbances always take the physical form of coherent diffusive flow of energy and particles in condensed or gaseous media—like, for example, a periodically switched-on flashlamp that generates diffuse photon waves of scattered light in thick fog.

But perhaps the most intriguing aspect of diffusion waves—and the one that lies at the heart of their applications—is that they offer a relatively simple tool for creating spatial coherence out of random ensembles of diffusive energy or particles.

In the 1970s, after the advent of the laser, sensitive new diffusion-wave methodologies for investigating condensed and gaseous matter were introduced. Laser beams—thanks to their spectral selectivity, spatial coherence, resolution, and enormous range in fluence—proved ideal for putting diffusion waves to work.

Today's most important and popular diffusion-wave methodologies fall into three major classes: photothermal spectroscopies and microscopies (which include photoacoustic beam-deflection,[5] photopyroelectric spectroscopy,[6] and infrared radiometric techniques[7]; see figure 1); laser pump-probe methods (known as photomodulated thermoreflectance[8]); and diffuse photon density waves (see Arjun Yodh and Britton Chance's article "Spectroscopy and Imaging with Diffusing Light," PHYSICS TODAY, March 1995).

These techniques have been the cornerstones of diffusion-wave investigations throughout the last 25 years and are credited with significantly improving the dynamic range of optical, electronic, and thermal measurements of condensed and gaseous media compared with conventional methods. Below, is a *fin de siècle* selection of a few key diffusion-wave applications that best exemplify how their unique physics leads to significant new advances in materials and metrology.

Ultra-low absorption in photonic thin films

When light propagates across a refractive index gradient caused by a temperature gradient, it is deflected. Known as the mirage effect, this phenomenon forms the basis of photothermal deflection spectroscopy (PDS). In PDS, two laser beams are used: a probe that sends photons into the medium being studied and a modulated pump, usually placed at right angles to the probe, whose photons are absorbed by the medium (see figure 1). Inside the absorbing medium, the modulated pump creates a thermal diffusion wave field following the conversion of optical energy to thermal energy. The optically generated thermal oscillation penetrates the surrounding gaseous or fluid medium within a thermal diffusion length and causes a modulated change in its refractive index right above the optically pumped region. The amplitude and phase of the deflected probe beam carry information about the optical and thermophysical properties of the absorbing solid or liquid.

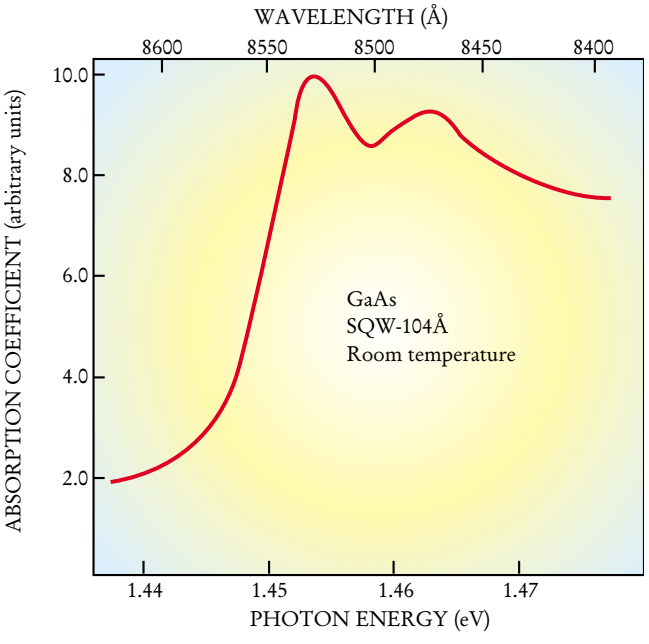


FIGURE 2. ABSORPTION SPECTRUM of a 10.4-nm-thick single quantum well of gallium arsenide at room temperature derived from photothermal deflection spectroscopy. The clear spectral resolution of the peaks attests to the high quality of the quantum well and its potential use in laser fabrication. (Adapted from ref. 9, Penna *et al.*)

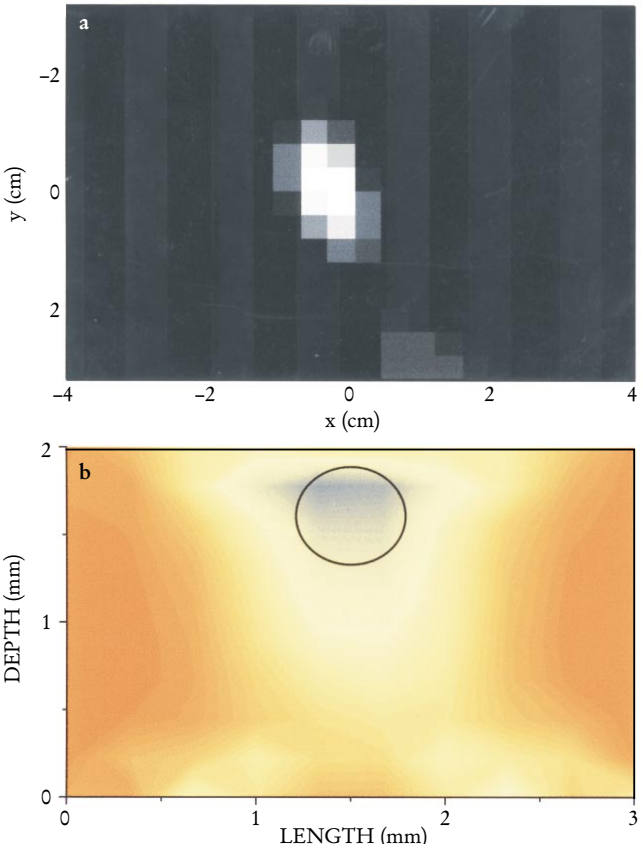


FIGURE 3. SLICE TOMOGRAPHIC IMAGES of diffusion-wave fields. (**a**) A diffuse photon density wave from a 1.2-cm diameter, highly scattering resin sphere situated 2.6-cm deep in a turbid intralipid solution. The image was obtained at modulation frequency of about 200 MHz (from ref. 3, Zhu *et al.*). (**b**) A thermal wave from a defect (hole) of 0.635-mm diameter, situated 1.5 mm deep in a mild steel obtained at 15 Hz. (From ref. 10, Nicolaides and Mandelis).

PDS is a popular and important technique because it can probe surfaces irrespective of the size of the sample. PDS is also free of the limitations of the photoacoustic cell enclosure. Moreover, samples can be studied in the open air, in gases, solids, or liquids, without contacting the sample and without microphonic noise limiting the signal-to-noise ratio.

Since the introduction of PDS in 1979 by Claude Boccara, Daniele Fournier, and their collaborators,[5] the technique has been widely adopted as one of the most sensitive solid-state spectroscopies for probing ultralow absorptances (absorptance is the product of a sample's absorption coefficient and its thickness). In particular, PDS is used to study electronic-defect-dominated spectral regions well below the optical gap of amorphous thin-film semiconductors. For such samples, the signal amplitude is proportional to the absorptance, so ultralow absorption spectra can be obtained directly from the amplitude of the thermal wave signal induced in the thin films upon nonradiative conversion.

PDS has also been successful in studying the excitonic structures of quantum wells in spectral ranges where optical spectroscopy has proven inadequate due to its high background level. Figure 2 shows the absorption coefficient spectrum[9] (proportional to the PDS thermal-wave signal amplitude) of a single-quantum-well gallium arsenide sample grown by molecular beam epitaxy. The well consists of a 10.4-nm layer of GaAs sandwiched between 40-period superlattices of GaAlAs/GaAs. The two peaks are related to the heavy-hole and light-hole excitons, resulting from the lifting of the degeneracy of the valence band in a two-dimensional system.

The ability of PDS to reconstruct density-of-states spectra from ultralow photothermal absorption spectra has led to the effective control of electronic defect populations and energetics. The fabrication of devices based on amorphous thin films, and very recently on porous silicon nanostructures, has been largely accomplished with the aid of precision measurements based on photothermal techniques (Ref. 2, volume IV).

Inverse problems

The heart of mathematical physics has always included inverse problems, a field in which one is called upon to reconstruct the cow from the hamburger meat, so to speak. When applied to square-law wave fields, inverse methods have created several important physical and medical breakthroughs in the 20th century, such as computer assisted tomography (CAT), ultrasonic imaging, and magnetic resonance imaging. What kind of inverse problems are diffusion waves capable of solving?

Because of their mathematical peculiarities, diffusion-wave fields belong squarely in the realm of the so-called ill-posed or ill-conditioned problems. For such problems, reconstructing the scatterers involves the discrete approximation of a Fredholm integral equation of the first kind, for which small perturbations in the data can lead to large-amplitude fluctuations and nonphysical artifacts in the reconstruction. The spatially damped nature of diffusion waves, however, presents two major advantages over conventional tomographic imaging techniques. Not only do diffusion waves provide high near-surface feature resolution, but they are also far less susceptible to ghosting—that is, the appearance of spurious signals from remote interfaces. For these reasons, physicists, mathematicians, biophysicists, and engineers turned their attention in the 1990s to tomographies that use diffuse photon density waves and thermal waves.[3,10]

A diffuse photon density wave is readily generated by sinusoidally modulating the amplitude of an optical point source (delivered through an optical fiber, for example) in a turbid medium, such as tissue or milk. Injected photons are elastically scattered following a random walk toward a detector (another optical fiber), and some are absorbed in midstream. The resulting oscillation limits the random walk and has been shown to be a spherical diffusion wave with a wavenumber $\kappa(\omega)$ given by $\kappa^2(\omega) = (v\alpha - i\omega)/D_\gamma$. Here, v is the speed of light in the medium, α is the absorption coefficient and D_γ is the diffusivity of light.

Physically, the collective motion of coherently driven and randomly scattered and absorbed photons creates a diffuse optical field with well-defined spatial phase lags with respect to the source phase and a characteristic diffusion length $\sqrt{2}/|\kappa(\omega)|$ of about 10 cm. Typical modulation frequencies are on the order of a few hundred MHz.

Phase sensitive demodulation of the received signal is possible with heterodyning instrumental schemes or, directly, with today's high-frequency lock-in amplifiers. The inequality of the wavenumber's real and imaginary parts and the very high speed of light require high-frequency operation to create a diffusion wave.

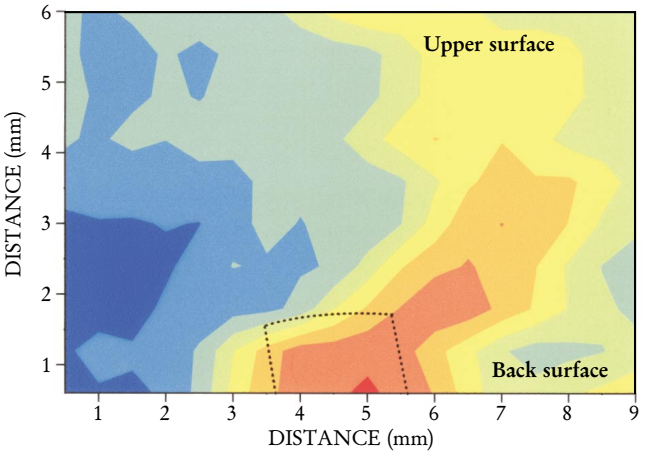


FIGURE 4. ELECTRONIC PLASMA-WAVE recombination lifetime image from a silicon wafer probed radiometrically from the upper (polished) surface, with a back-surface mechanical defect (delineated). The image was obtained at 10 kHz. (Adapted from M. E. Rodriguez, A. Mandelis, G. Pan, L. Nicolaides, J. A. Garcia and Y. Riopel, *Journal of the Electrochemical Society*, volume 147, page 687, 2000.)

Tomographic techniques based on diffusion waves have been developed to solve the inverse problem of obtaining the shape of an inhomogeneity in a turbid medium. Even though the algorithms used to analyze the data are not conventional CAT-scan algorithms, the instrument is operated in a CAT-scan mode. To stabilize the inversion for a given level of accuracy, elaborate computational signal conditioning procedures known as regularization techniques have been developed.[10] At present, objects of about 1 cm in diameter can be imaged (see figure 3a), but new methodologies are being developed to improve the spatial resolution for clinical applications.

Thermal-wave tomographies operate on similar principles, but with important differences. Because the two parts of their wavenumber are equal, thermally driven diffusion waves can be manifested at any frequency. Moreoever, their much smaller wavevector magnitude allows the identification of sub-surface features in the submillimeter range, such as the one shown in figure 3b.

Charge-plasma waves in semiconductors

When a semiconductor is optically excited with a harmonically modulated beam of monochromatic photons of energy greater than the solid's bandgap, several dynamic processes can occur that give rise to diffusion waves. In particular, the illumination creates energetic electron–hole pairs, which collide with each other and with lattice phonons until, on a picosecond time scale, thermal equilibrium is achieved. Called direct lattice heating, this phenomenon is a source of thermal waves in the semiconductor.

But another process occurs. Excess photogenerated charge carriers diffuse away from the source of the heating until, after an average lifetime τ, they recombine with carriers of the opposite sign or defects. The modulated densities of these randomly moving carriers constitute a charge plasma diffusion wave for which $F(\mathbf{r}) = -1/D_n\tau$ and $\kappa_n^2(\omega) = (1 + i\omega\tau)/D_n\tau$. Here, the subscript "n" is used for electrons, and the subscript "p" for holes. In a p-type medium, D_n is the minority (electron) diffusivity.

Because the wavenumber's real and imaginary parts are unequal, it is only when $\omega\tau \gtrsim 1$ (that is, when the imaginary part becomes as large as the real part) that this type of carrier oscillation behaves like a diffusion wave.

Several experimental techniques have been developed in recent years to detect carrier diffusion waves. Among them, photomodulated thermoreflectance has offered valuable physical insights into the interaction of radiation with the excited electronic states in semiconductors.[8] In this technique, a harmonically modulated pump beam generates thermal and carrier waves that, in turn, modulate the solid's optical reflectance. A second, CW probe laser measures the reflectance.

Plasma waves are increasingly used to investigate defects in electronic solids that are destined for high-density micro- and nano-devices. Under the general term infrared photothermal radiometry, various techniques have been developed that capture the oscillating blackbody radiation emitted by optically excited semiconductors.[7] This oscillation, which is superposed on the blackbody emission from direct lattice heating (also in the infrared), is the plasma wave created optically. When carriers recombine they emit an infrared photon. In effect, each recombining carrier acts as a discrete blackbody radiator. Recombination lifetimes in silicon are a few microseconds or longer, so the plasma diffusion wave appears when the incident photons are modulated at frequencies of 1 kHz and higher.

The amplitude and phase of the collected blackbody radiation flux measures not only the recombining free carrier density, but also the dynamics of the incoherent recombination process that creates the plasma diffusion wave. The superposed thermal wave from direct lattice heating is easily separated from the plasma wave because heat diffuses more slowly than the carriers.

When silicon crystals of normal quality are examined with this technique, the radiometric signals above 200–500 Hz are completely dominated by the de-exciting electronic blackbody radiators. The frequency dependence of the plasma wave involves the recombination lifetime, τ, in the bulk of the semiconductor and the electronic diffusivity, D_n (or D_p). It also involves the rate of recombination at the surfaces (or surface recombination velocity), which depends on the surface-state density and is an indicator of the preparation, cleaning, and processing of the sample. At low frequencies, the amplitude of the plasma wave is directly proportional to the photoinjected carrier lifetime (Mandelis, in ref. 7). As a result, an image of the laser-beam-scanned radiometric amplitude is, in effect, an image of the recombination lifetime (subject to a multiplicative calibration constant).

Because carriers diffuse rapidly to the back surface of the sample, depletion conditions there can affect the entire spatial density gradient. Taking advantage of this property, a novel technique has emerged that uses plasma diffusion waves to image the minority recombination lifetime, thereby probing conditions in the bulk of the sample (see figure 4). This nascent technology, which is unusually sensitive to electronic defects across the thickness of Si crystals and industrial wafers, has revealed that electronic defects in a substrate exert their influence on the local quality of micro- and nano-electronic devices over material regions much broader than anyone thought possible.

Looking ahead

The applications described here demonstrate what can be accomplished with diffusion wave physics. In the 21st century, we are bound to see many of these applications

transferred to industry, where they will lead to advances in materials, photonic devices, fundamental materials science, biomedical physics, and manufacturing yield. And thanks to their power to exert spatial control over dynamic diffusion processes, diffusion-wave technologies will have an important role to play in difficult cases where today's other leading diagnostic techniques, such as optics, x rays, and ultrasonics, cannot be used. As their spatial resolution improves, diffusion wave tomographies and inverse methodologies will be used in such applications as cancer diagnostics, dental imaging of caries, and the nondestructive evaluation of critical and valuable manufacturing components. For diffusion-wave researchers and users alike, the coming years will be very exciting!

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