The Photorefractive Effect

A laser beam passing through a crystal can suddenly burst into a spray of light. This photorefractive effect may be the key to developing computers that exploit light instead of electricity

by David M. Pepper, Jack Feinberg and Nicolai V. Kukhtarev

When Arthur Ashkin and his colleagues at Bell Laboratories first noticed the photorefractive effect some 25 years ago, they considered the phenomenon a curiosity at best and a complete nuisance at worst. Today photorefractive materials are being shaped into components for a new generation of computers that exploit light instead of electricity.

Ashkin was experimenting with a crystal of lithium niobate (LiNbO₃) that he hoped would convert one color of intense laser light to another (a process technically called second-harmonic generation). As part of his tests, he directed a laser beam through the crystal. At first, the crystal performed quite admirably, allowing light to pass through undisturbed. But after a few minutes, the crystal began to distort the beam, scattering light around the laboratory. Somehow the laser light that bombarded the material had altered the optical properties of the crystal itself. This photorefractive effect would persist in the crystal for days. If the workers bathed the crystal in a uniform beam of light, however, the crystal would once again transmit an undistorted beam.

During the past 25 years investigators have discovered a wide variety of photorefractive materials, including insulators, semiconductors and organic compounds. Photorefractive materials, like film emulsions, change rapidly when exposed to bright light, respond slowly when subjected to dim light and capture sharp detail when struck by some intricate pattern of light. Unlike film, photorefractive materials are erasable: images can be stored or obliterated at whim or by design.

By virtue of their sensitivity, robustness, and unique optical properties, photorefractive materials have the potential to be fashioned into data-processing elements for optical computers. In theory, these devices would allow optical computers to process information at much faster rates than their electronic counterparts. Employing photorefractive materials, workers have already developed the optical analogue to the transistor: if two laser beams interact within a photorefractive material, one beam can control, switch or amplify the second beam. Photorefractive materials also lie at the heart of devices that trace the edges of images, that connect networks of lasers and that store three-dimensional images.

Because the optical properties of photorefractive materials can be modified by the very light that passes through them, they are categorized as nonlinear optical media. In linear optical media—such as lenses, prisms and polarizing filters—light beams merely pass through one another, without changing the properties of the material.

The photorefractive effect is closely related to another nonlinear phenomenon known as the photochromic effect. The light that strikes a photochromic material can change the amount of light that the medium absorbs. Photochromic materials, which are incorporated in certain brands of sunglasses, darken in bright sunlight and lighten in dark rooms.

In photorefractive materials, the light that bombards the material affects how fast light travels through it. More specifically, the photorefractive effect is a process in which light alters the refractive index of a material. (The refractive index is the ratio of the speed of light in a vacuum to that in the material.)

Most transparent materials will change their refractive index if bombarded by light of sufficient intensity. Light is a traveling electromagnetic wave whose electric field strength is proportional to the square root of the intensity of light. For instance, an optical beam whose intensity is 100 million watts per square centimeter is equivalent to an electric field strength of about 100,000 volts per centimeter. When such intense light is directed at a transparent material, it disrupts the positions of the atoms, changing the refractive index by a few parts in one million. As a result of the change, the material can act like a prism or a lens to deflect light.

The term “photorefractive” is usually reserved, however, for materials whose refractive index changes in response to light of low intensity. In photorefractive materials, light beams as weak as one thousandth of a watt per square centimeter can alter the arrangement of atoms in a crystal, changing the refractive index as much as a few parts in 10,000. And unlike most transparent materials, the change in photorefractive crystals is semipermanent: if an altered crystal is isolated from all sources of light, the change in the refractive index can last from milliseconds to years, depending on the material. In this manner, one can store information in the form of images in a crystal.

LASER BEAM striking a 5-mm crystal of barium titanate scatters into a fan of light owing to the photorefractive effect.

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How light alters the optical properties of a photorefractive crystal

1. Two laser beams interfere in a photorefractive crystal to form a pattern of bright and dark regions.

2. Mobile electrons migrate away from bright regions of the crystal.

3. Mobile electrons accumulate in the dark regions, leaving regions of positive charge.

4. An electric field forms between regions of positive and negative charge.

5. The electric field distorts the crystal lattice.

6. The distortion causes light to travel slower through some regions and faster through others. More specifically, the refractive index is altered periodically. The refractive-index grating is shifted one quarter of a period in space from the light pattern.

How can a weak beam of light cause such a strong change in the refractive index of a crystal? In the late 1960s F. S. Chen of Bell Laboratories advanced the basic model of the photorefractive effect. Just as a single ant can move a large mound of sand one grain at a time, a weak beam of light can gradually build up a strong electric field by moving electric charges one by one. In photorefractive crystals, charges diffuse away from bright regions and pile up in dark regions. As more and more charges are displaced, the electric field inside the crystal increases, attaining a strength as high as 10,000 volts per centimeter. The electric field will distort the crystal lattice slightly (about .01 percent), thereby modifying the refractive index.

The source of these electric charges apparently lies in defects in the crystal lattice of the material. The defects can be mechanical flaws in the lattice structure (missing atoms at certain lattice sites), substitutional dopants (a foreign atom at some lattice site) or interstitial dopants (a foreign atom wedged between native atoms). Very small amounts of these defects, on the order of parts per million, can cause the photorefractive effect.

Each crystal defect can be the source of an extra charge, which can be either electrons (particles of negative charge) or holes (regions of positive charge), depending on the particular crystal. In the dark, these charges are trapped; in the presence of light, they are free to roam within the crystal until they eventually become caught again. If light illuminates charges in one region of the crystal, they will diffuse away from that region and accumulate in the dark, in the way cockroaches scurry underneath furniture to avoid light.

Each charge that moves inside the crystal leaves behind an immobile charge of the opposite sign. In the region between these positive and negative charges, the electric field is strongest, and the crystal lattice will distort the most. A beam of light that passes through this region of the crystal will experience a different refractive index from that of the unaffected regions.

The time it takes for light to rearrange charges in a crystal depends on the intensity of the light and also on how fast charges migrate in the crystal. Weak light takes longer than strong light to build up the same electric field. For low-intensity light (about .01 watt per square centimeter), it can take minutes for the changes to reach their equilibrium pattern. For high-intensity light (about a billion watts per square centimeter), the response time can be less than a nanosecond. A photorefractive crystal, like photographic film, requires a certain amount of light to complete its "exposure."

The change in refractive index is linearly proportional to the strength of the electric field if the crystal lattice lacks a certain property called an inversion symmetry. The electric field will remain in the crystal long after the light is removed, just as the mound of sand remains in its new location long after the ants have left.

One of the most useful consequences of the photorefractive effect is the exchange of energy between two laser beams, which is also known as two-beam coupling. If two laser beams of the same frequency intersect, they will interfere and produce a stationary pattern of bright and dark regions—or more specifically a pattern whose intensity varies sinusoidally with position in the crystal. If this sinusoidal pattern of light forms within a photorefractive crystal, electric charges will move to generate an electric field whose strength also varies sinusoidally. The resultant field will distort the
crystal lattice in a similar periodic manner, producing changes in the refractive index. Ultimately a “refractive-index grating” (also called a refractive-index volume hologram) will be formed within the crystal.

The electric field and the refractive-index grating will have the same periodicity as the light pattern, but they will be shifted by one quarter of a period in space from the incident light. This displacement—called a 90-degree phase shift—is the optimal configuration for the exchange of energy between the original two laser beams.

Once the refractive-index grating has been established in the crystal, some of the light of one beam will be deflected, or diffracted, by the grating in the direction of the other beam (and vice versa). Hence, the two deflected beams will interfere with the two original beams—constructively in one case and destructively in the other. In the case of constructive interference, the peaks of the light waves in one of the deflected beams combine with the peaks of one of the original beams, and the beams therefore reinforce each other. In the case of destructive interference, the peaks of the waves from the other deflected beam combine with the valleys from the other original beam, and the light waves diminish each other. The beam formed from constructive interference will emerge from the crystal stronger than when it entered, whereas the beam formed from destructive interference will emerge weaker. Hence, one of the beams will have gained energy from the other. Which beam gains and which beam loses is determined by the orientation of the crystal and whether the charge carriers are holes or electrons.

Photorefractive materials exhibit two-beam coupling because the optical pattern and the refractive-index grating are shifted in space. Two-beam coupling is not found in most nonlinear materials, however, because they respond “locally” to optical beams (for example, atomic orbitals are deformed by the intense electric fields of the laser beams). In most nonlinear materials, therefore, the optical pattern and grating precisely overlap. The light deflected by the grating interferes with each of the undeflected beams in exactly the same way. Thus, the two beams exchange an equal amount of energy, so neither grows in intensity.

To enhance the photorefractive effect, investigators have learned to control the flow of charge within a material. The two mechanisms that control the flow of charge in a crystal are diffusion and drift. They are analogous to the diffusion and drift of smoke from a burning ember. Left on its own, smoke will diffuse to regions of low smoke density. If a slight breeze is blowing, however, the smoke will drift in a particular direction. The particles in the smoke behave like mobile charges in a photorefractive material: the charges tend to move toward regions of lower charge density, and they drift in response to any electric field.

The simple diffusion of charges from bright regions to dark regions of a crystal does not produce the strongest electric field possible. In 1981 Jean-Pierre Huignard and Abdellatif Marakchi of Thomson-CSF Laboratories in Orsay, France, applied an electric field externally to a photorefractive crystal to build up a spatially varying field stronger than that produced by diffusion alone. The applied electric field, however, shifted the refractive-index grating away from the optimal quarter-cycle phase shift.

To prevent this nonoptimal spatial shifting of the grating, Sergei I. Stepashkov and Mikhail P. Petrov of the A. F. Ioffe Physico-Technical Institute of the Soviet Academy of Sciences in Leningrad developed a clever technique. When they applied an external electric field that rapidly alternated its direction, the charges would preferentially drift in one direction for the first half cycle of the applied field and in the opposite direction for the next half cycle. The process is similar to having two people alternately blow on a burning ember from opposite sides. The resulting smoke pattern is both intensified and spread out farther in space but has the same average location as if no net wind were present. In photorefractive crystals the process yields an internal electric field larger than that produced by diffusion alone, and the refractive-index grating has the same average quarter-cycle phase shift as if no drift field were present.

Workers have used this technique to enhance the efficiency of two-beam coupling as well as an effect called beam fanning (technically known as stimulated, forward photorefractive scattering). Discovered in the 1970s, beam fanning is perhaps one of the most intriguing nonlinear optical phenomena. It can be observed, for example, when a pencil-thin, weak beam from a helium-neon laser illuminates a crystal such as barium titanate (BaTiO3). Initially the beam passes through the crystal unaltered. After a second or so (the time depends on the intensity of the light), the beam begins to spread out in the crystal, curving to one side. In the process the curved beam divides into many rays that appear to spray out into a broad fan of light—hence the term “beam fanning.” Depending on the choice of photorefractive crystal, the emerging light in cross section can
PHASE-CONJUGATE MIRROR made from a photorefractive crystal allows communication through the atmosphere. The receiving station emits a beam of light. As the beam travels through the atmosphere, it spreads out and distorts. At the transmitting station a photorefractive crystal either reflects or transmits the light that hits it, depending on the voltage applied through the electrodes. Because the crystal acts as a phase-conjugate mirror, the reflected light is time-reversed. A message can be encoded by alternately reflecting and transmitting the light. As the time-reversed message beam interacts with the atmosphere, all distortions are removed from the beam, and the message can be decoded at the receiving station.

Because of this property, phase-conjugate mirrors have myriad applications in the fields of optical communications, high-power lasers and optical computing. As an example, they can be incorporated into a system to correct undesirable aberrations that laser beams sometimes acquire during propagation through distorting media or powerful laser amplifiers [see “Applications of Optical Phase Conjugation,” by David M. Pepper; SCIENTIFIC AMERICAN, January, 1986].

In 1977 Robert W. Hellwarth of the University of Southern California suggested a basic configuration for phase-conjugate mirrors, and two years later Sergei G. Odulov and one of us (Kukhtarev) and independently Huignard and his co-workers produced such a mirror that incorporated photorefractive materials. In 1982 one of us (Feinberg) serendipitously discovered a class of phase-conjugate mirrors that many investigators use today. Feinberg had focused three laser beams on a crystal of barium titanate. One beam contained the light waves whose time-reversed replica was sought; the two additional “pump” beams were needed to form the phase-conjugate mirror (or so Feinberg and his colleagues thought at the time). To check the experiment, Feinberg blocked the pump beams to ensure that the presumed time-reversed beam did not arise merely from a simple reflection from a crystal face. At first, the time-reversed beam obediently vanished. But after a short time, the time-reversed beam surprisingly reappeared. Feinberg had found a phase-conjugate mirror that required only a single beam. Feinberg’s elegant device is an example of a more general class of self-pumped, phase-conjugate mirror, which was pioneered by Jeffrey O. White, Mark Cronin-Golomb, Baruch Fischer and Amnon Yariv of Cal Tech.

Although phase-conjugate mirrors can be made from many classes of nonlinear optical materials, photorefractive elements have several distinct advantages: first, the mirrors require only one input beam—the very beam whose phase-conjugate replica is sought—thus forming the so-called self-pumped phase conjugator; and second, very low laser powers and intensities can initiate the process leading to the time-reversed beam.

Why does nature love the phase-conjugate beam? A partial answer to this question posed by Hellwarth can be advanced at least in the case of barium titanate, as postulated by Kenneth Mac Donald, then at the University of Southern California, and one of us (Feinberg). After a short time, beam fanning causes...
es the incident laser light to be swept preferentially to one side of the crystal. If the incident beam and crystal are positioned so that the fanned beam is swept into a far corner of the crystal, the fanned beam undergoes two internal reflections, essentially folding back onto itself. This reflected beam fans again back along the direction of the incident beam. Out of all the scattered beams inside the crystal, the time-reversed beam—by virtue of its backward trajectory—gains more energy than the other scattered beams. This beam-reversal process can be very efficient: 60 percent of the power in the incident beam can emerge as the phase-conjugate beam.

One of us (Pepper) has added an additional twist to this novel phase-conjugate mirror. Pepper attached electrodes to a photorefractive crystal to apply a time-varying electric field across the crystal. When a laser beam strikes this crystal, it not only is time-reversed but also can be pulsed in time like a shuttered mirror. In this manner, pulsed information can be relayed from the conjugate mirror back to the laser source; the time-reversed nature of the beam guarantees that the two communication points remain locked onto each other. This scheme can be used to establish a communications channel between two satellites or to relay information from a remote sensor placed at one end of an optical fiber link (see illustration on page 70).

Another application that takes advantage of the energy-exchange mechanism is a device called a novelty filter, which highlights whatever is changing in a highly complex scene. Such devices can pick out moving airplanes against a background of buildings, a sportswoman diving into a still pond or bacteria swimming against a background of motionless algae.

One type of novelty filter demonstrated by Cronin-Golomb (now at Tufts University) involves two beams that illuminate a photorefractive crystal. An image is encoded spatially onto the first laser beam. The crystal is oriented so that this image-bearing beam transfers most of its energy to the second beam. After the image-bearing beam passes through the crystal, it becomes almost completely dark. Whenever something in the image-bearing beam changes, however, the energy-exchange mechanism is disturbed momentarily, and the part of the image beam that has changed will pass through the crystal. This part of the beam can then be viewed on a video monitor. Once the motion has ceased, the image-bearing beam will become dark once again after passing through the crystal.

Photorefractive crystals can also be employed in other applications to enhance the edges of an image. An image is encoded onto an "object" beam, which is directed into a photorefractive crystal along with a "reference" beam. The two beams interfere inside the crystal and produce a hologram of the original image. The image can be recovered by a third "readout" beam, which is aimed in a direction opposite to the reference beam. If the object beam is relatively weak, then the reconstructed image will be a faithful replica of the original picture. If the object beam is more intense than the other two beams, however, then the edges of the reconstructed image will be enhanced.

The intensity of the object beam varies locally in the crystal because the beam contains an image. Hence, its intensity will match that of the reference beam at every edge in the image, because an edge contains a full range of intensities, from dark to bright. Wherever the intensities of the two beams match, the optical interference pattern at that particular region in the crystal will have the largest modulation from bright to dark to bright and so on. The strong interference pattern will then generate a strong refractive-index grating. When the readout beam interacts with the strong grating, it will be most efficiently deflected, or diffracted, at that location, and so the reconstructed image will emerge with all of its edges highlighted.

Because photorefractive crystals can act as both energy couplers and phase-conjugate mirrors, they are particularly useful for reconfigurable optical interconnects and frequency-locking of lasers. Photorefractive crystals can relay information from one optical element (say, an optical fiber) to another (a data-processing element), free of complicated optical elements or electronic interconnects. This optical relaying scheme can also force two (or more) separate lasers to "lock" onto each other so that the two lasers oscillate at precisely the same optical wavelength; in this way, the two separate lasers essentially behave as one larger laser.

How do two or more mutually incoherent beams of light become connected in a photorefractive crystal? When the different beams illuminate a photorefractive crystal, each will produce its own armada of scattered beams and random refractive-index gratings (or holograms) within the crystal. If one hologram from one beam exactly matches one of the holograms from the other beam, then hologram will grow faster in strength than the others. The result is that the two arms will be connected to each other. This device is called a mutually pumped, or doubly pumped, phase-conjugate mirror.

The device—first demonstrated in 1987 by Fischer, now at the Technion-Israel Institute of Technology in Haifa, and his co-workers, by Robert W. Eason and A.M.C. Smout of the University of Essex in Great Britain and subsequently by Mark D. Ewbank of the Rockwell International Science Center in Thousand Oaks, Calif.—can connect any two beams originating from any direction. If the two beams contain images, then each image will be converted into the other as the beams traverse the crystal (see illustration on page 74). If one beam is pulsed rapidly, the temporal information is relayed back toward the other beam.

If two beams come from different lasers that oscillate at slightly different frequencies, then the beam from one laser will be diverted into the approximate conjugate, or time-reversed, di-

PARAMECRIIUM is hidden among algae and debris (left), but when the scene is viewed through a novelty filter, only the swimming microorganism appears (right). Novelty filters can highlight any object that moves against a stationary background. This filter was designed by R. M. Pierce, R. Cudney, G. D. Bacher and J. Feinberg.
reduction of the beam from the other laser, and vice versa. If two such lasers are optically connected by a crystal, they can, under the proper conditions, be made to oscillate coherently, thereby locking the frequency of the two lasers. With this scheme, workers hope that thousands of semiconductor lasers can be efficiently combined into a high-power source of coherent light.

To construct better optical components and devices, investigators are searching among the many different kinds of photorefractive materials to find the most efficient and reliable crystals. Photorefractive materials vary greatly in their optical, electrical and structural properties—for example, the insulator barium titanate, the semiconductor gallium arsenide and the organic compound 2-cyclooctylamino-5-nitropyridine doped with 7,7,8,8-tetracyanoquinodimethane.

These seemingly different materials nonetheless exhibit similar photorefractive effects. They all have a crystal lattice that is relatively easy to distort, and they all contain defects, which act as a source of charge carriers and charge traps. Under the influence of an electric field, however, charges move about 10,000 times faster in gallium arsenide than they do in barium titanate. For the same incident light intensity, therefore, the photorefractive effect evolves much more rapidly in gallium arsenide than it does in barium titanate. The same incident light intensity, therefore, the photorefractive effect evolves much more rapidly in gallium arsenide than it does in barium titanate. The optical properties of the crystals in the different materials vary greatly as well: gallium arsenide and other semiconductors are typically photosensitive in the near infrared part of the optical spectrum, whereas most insulators and organic compounds are sensitive in the visible spectrum.

A flurry of experiments currently in progress aim to identify and possibly control the defects responsible for the photorefractive effect in various crystals. As an example, the source of the photorefractive effect in barium titanate is an open question: many researchers attribute the effect to the presence of various ionization states of transition metal impurities such as iron, cobalt and manganese; oxygen vacancies in the lattice may also contribute to the charge carriers. In gallium arsenide, on the other hand, the photorefractive effect is attributed to an intrinsic lattice defect thought to be formed by a combination of an arsenic atom replacing a gallium atom in the lattice and an additional arsenic atom placed in the same lattice cell, forming the so-called EL2 center.

Regardless of the character of the crystalline defects, the properties of most photorefractive crystals can be altered by doping them with impurities or by drawing atoms out of the lattice. For example, one of us (Feinberg) and Stephen Ducharme of U.S.C. found that when a crystal of barium titanate is heated in an oxygen-free environment to remove some oxygen from the crystal lattice, its photorefractive properties are altered markedly because the dominant charge carriers are changed from holes to electrons.

As workers increase the concentration of the defects in a photorefractive material, they have found that the number of available charges increases, thereby enhancing the strength of the internal electric field and the refractive-index gratings. On the other hand, defects scatter and absorb light from the incident beams. Defect concentrations of from one to 100 parts per million appear to be optimal in terms of providing a reasonable number of charges without appreciably attenuating the input light.

Perhaps the best photorefractive materials will be those painstakingly fashioned out of semiconductor materials. Stephen E. Ralph, David D. Nolte and Alastair M. Glass of AT&T Bell Laboratories have recently shown that layers of gallium aluminum arsenide alloys only a few atoms thick can be assembled into structures—called superlattices and quantum wells—that exhibit a measurable photorefractive effect. Such materials are currently being studied for their novel electrical properties and high speed of response. These crystals may also lead to a new class of integrated processors based on optics and electronics.

What makes photorefractive materials so promising for optical computing is their high sensitivity to light, coupled with their ability to connect light beams from different lasers and to change one information pattern into another. The future of photorefractive materials will depend on whether their optical properties can be as meticulously tailored as semiconductors are today. Ideally, photorefractive devices would be integrated with semiconductor lasers and detectors to form a single, compact device capable of processing millions of bits of data simultaneously in each microsecond, yielding a total data-processing rate of trillions of bits per second.

Further Reading


