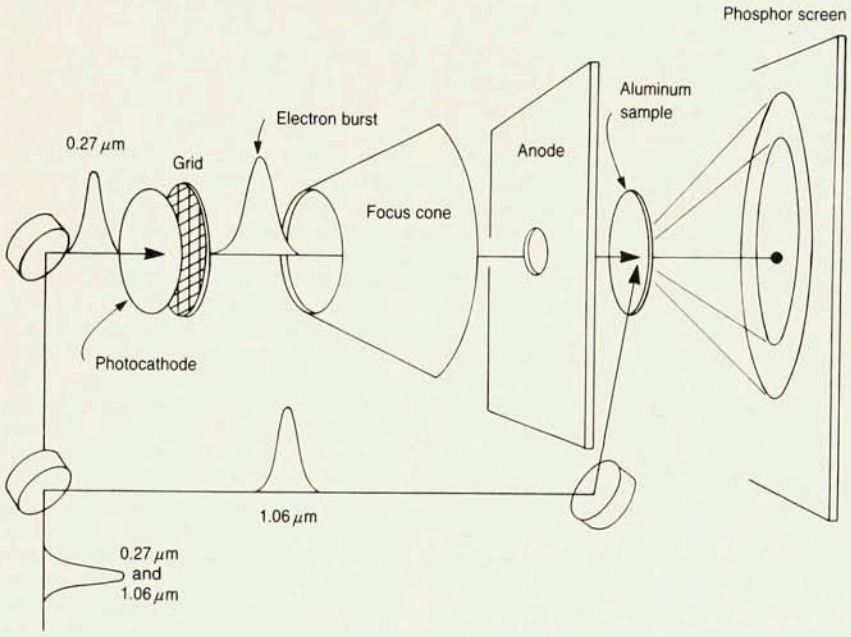
Picosecond time resolution for electron diffraction

Gerard Mourou and Steven Williamson at the University of Rochester have reported the demonstration of a new technique that makes possible the direct observation of laser-induced structural changes and phase transitions in a crystal lattice on a picosecond time scale. They have been able to observe electron diffraction patterns with electron bursts photoelectrically generated by 30-picosecond laser pulses. With this technique Mourou sees no impediment to the production of electron diffraction patterns time resolved to a few hundred femtoseconds.

What happens to a crystal lattice on a picosecond time scale when it is subjected to ultrashort laser pulses is a question of considerable theoretical and practical interest. Annealing of doped semiconductor materials with picosecond laser pulses can produce doping concentrations orders of magnitude above the limits of normal solubility. A controversy has arisen about the kinetics of the laser–lattice coupling on this time scale. Although the weight of the indirect experimental evidence has supported the widely held view that the laser-generated hot electron plasma comes quickly into thermal equilibrium with the lattice, melting it locally in less than a picosecond, this picture has its challengers.

One would like to be able to examine such ultrafast structural changes directly by diffraction analysis. But time-dependent kinetics has until now gone unmonitored on this time scale for lack of suitable picosecond x-ray or electron probes that could be synchronized with a laser pulse. X-ray diffraction analysis has been limited to time resolutions not much better than a nanosecond.

Mourou and Williamson hit upon their picosecond diffraction technique while studying picosecond optical transients (in laser-induced fluorescence, for example) with a streak camera. Traditional streak cameras determine the time dependence of very fast phenomena by recording their photographic images on a rapidly rotating drum. Such mechanical devices are however limited to temporal resolution of about ten nanoseconds. Modern streak cameras achieve sub-picosecond resolution by photoelectrically converting an optical image to an electron burst, which is then resolved in time by deflecting the electrons in a time-dependent electric field.



A modified streak camera was used by Mourou and Williamson to demonstrate the feasibility of picosecond electron diffraction. A 30-psec pulse of 1.06-micron laser light is quadrupled in frequency and directed to the camera's photocathode, where it generates a 100-psec electron burst, which is accelerated, focused and passed through 300 Å of aluminum sample. The resulting diffraction pattern appears on the phosphor screen. In experiments now under way, the residual 1.06-micron component is split off by a dichroic mirror and sent directly to the sample surface. This annealing pulse arrives at the sample before the 0.27-micron probing pulse, the variable time delay being determined by the difference in optical paths.

The photocathode of a good streak camera reproduces a spatial and temporal electron replica of the incident light pulse faithful to a fraction of a picosecond. It occurred to Mourou and Williamson that they "were wasting such beautiful electron replicas" by using them merely to record optical transients. They set out to see if one could observe diffraction patterns by passing laser-pulse generated picosecond electron bursts from a streak camera through thin samples of polycrystalline and single-crystal material.

They removed the deflection plates from a Photochron II streak camera and inserted between its photocathode and phosphor detecting screen an aluminum sample thin enough so that a 20-keV electron passing through the sample would typically experience only a single elastic collision. In the camera's normal streak mode, the deflection plates are subjected to a rapidly ramped voltage that provides the instrument's time resolution by imposing a time-dependent deflection on the electron trajectories. But in these picosecond diffraction studies the plates were superfluous; the streak camera was being used only to generate, focus and accelerate the electron burst, and to record its diffraction pattern after passage through the sample.

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The first Rochester experiments, whose purpose was to demonstrate the feasibility of the technique, used 30-picosecond pulses from a 1.06-micron mode-locked Nd:YAG laser. Because the work function of the streak camera's gold photocathode was about four times the energy of the 1.06-Å laser photons, it was necessary to quadruple the frequency of the laser light in two successive frequency doubling crystals before directing it onto the photocathode. The resultant photoelectron burst was then electrostatically focused and accelerated to 20 keV in the camera before passing through 300 Å of aluminum sample to a phosphor detecting screen.

Because the aluminum sample, like all ordinary metals, is polycrystalline, one expects the 100-micron-wide focused beam of 20-keV electrons (with a deBroglie wavelength of about 0.1 Å) to produce a diffraction pattern of concentric rings on the phosphor screen. Mourou and Williamson were able to observe four distinct diffraction rings, whose radii indicate a face-centered cubic crystal with a lattice constant of 4.03 Å $\pm$ 0.08 Å—in good agreement with the known crystal structure of aluminum.

By operating the camera in its streak mode, Mourou and Williamson measured the duration of the photoelectron bursts to be about 100 psec, and they determined that their onset was well synchronized with the initiating laser pulses. The fact that the electron burst is significantly longer than the laser pulse, Mourou told us, is a temporary problem resulting from the need to get an adequate diffraction ring signal from a single laser pulse. The number of photoelectrons required in these single-shot polycrystalline demonstrations produces a space-charge density that significantly broadens the duration of the electron burst; one is forcing the streak camera to operate beyond its regime of linearity. One wants, of course, to keep the electron bursts as short as the laser pulses that generate them—ultimately on the order of a hundred femtoseconds. To do this, Mourou explained, one must operate the camera with lower laser intensities and consequently lower space-charge concentrations. To get an adequate signal-to-noise ratio at low intensity one can run the laser at high repetition rates and use signal-averaging techniques. Mourou points out that both his streak camera and the 70-fsec colliding-pulse ring laser recently developed at Bell Labs are capable of 100-MHz repetition rates (PHYSICS TODAY, December, page 19).

For some applications, the repetition rate must be kept low enough to permit the sample to cool between pulses. But single-crystal samples will require far fewer photoelectrons than does polycrystalline aluminum; their diffraction patterns are concentrated in a few reciprocal-lattice spots instead of being spread out over rings. Furthermore it may be possible to reduce the required photoelectron flux substantially by increasing the gain of the image intensifier used to amplify the diffraction pattern.

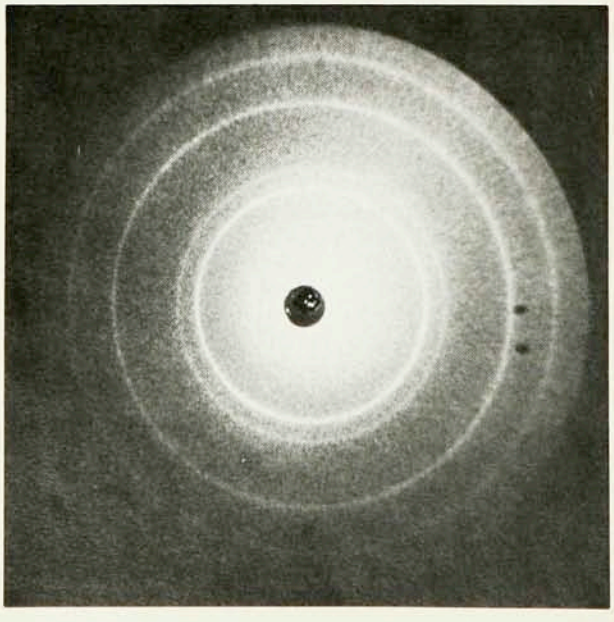
Observing phase transitions. The only result reported thus far[1] with the new Rochester device is the diffraction pattern of a cold, stable piece of aluminum. But the *raison d'être* of the scheme is to record the evolution of the ultrafast changes induced in crystal structures by picosecond laser pulses. The beginnings of this work are now under way in Mourou's laboratory. The same laser pulse that generates the probing electron burst is made to impinge directly on the sample surface.

After passing the laser pulse through the frequency doublers, one splits it with a dichroic mirror, directing the fourth harmonic to the photocathode and sending the remainder to the sample. By stepwise increase of the optical path of the probing component one can examine the diffraction pattern at different time delays after the "annealing" component strikes the surface. At present Mourou and Williamson are examining the diffraction pattern of aluminum in 100-picosecond steps after a 30-picosecond 1.06-Å laser pulse reaches the sample surface. For this purpose, several pieces of aluminum are arrayed on a carousel, a fresh piece being used for each laser pulse. They expect shortly to report the results of this work. The next step, Mourou told us, will be to reduce the time resolution to about one picosecond by using shorter laser pulses and lower photoelectron fluxes.

A very attractive feature of picosecond electron diffraction, Mourou stresses, is the fact that it can be done with a very small laser. Nanojoules of laser output should be adequate for seeing the diffraction pattern. "Unlike synchrotron radiation probes, which require a large accelerator, this is a table-top technique."

By observing the time evolution of the diffraction pattern in response to a picosecond laser pulse one can see directly when (or if) a phase transition takes place. In pulsed-laser annealing of materials and semiconductors, the photons interact at first only with the electrons of the materials, generating a hot plasma of conduction electrons (PHYSICS TODAY, June 1982, page 24). Because the electron plasma has very low specific heat, it rapidly attains a temperature much higher than that of the lattice. The time scale on which the plasma shares its thermal energy with the lattice has become an issue of debate between competing theoretical models. If this relaxation time is as short as the widely accepted "thermal melting" model suggests, annealing by picosecond laser pulses does indeed involve the melting (and subsequent recrystallization) of the material surface.

James Van Vechten and some of his colleagues at IBM have taken exception to this view. Their "plasma model" argues[2] that the electron–hole density generated by an annealing pulse is large enough to produce a Bose condensation of the plasma, inhibiting the electron–phonon coupling and thus delaying thermal equilibrium between the plasma and the lattice for much longer than a picosecond. The transitions that occur in picosecond laser



Diffraction pattern generated by 100-psec burst of 20-keV electrons passing through 300 Å of aluminum shows rings characteristic of a polycrystalline material. The radii of the four rings give a lattice constant of 4.03 Å, in good agreement with the known aluminum crystal structure.

annealing of silicon, Van Vechten suggests, involve a relatively cool quasi-liquid-crystal phase "distinctly different from molten silicon."

Experiments employing time-resolved reflectivity, surface photoelectric emission and nanoscond x-ray diffraction have all recently lent support to the thermal melting model, while a Raman-scattering experiment appears to support Van Vechten. But the new electron diffraction technique of Mourou and Williamson promises soon to provide our first direct look at lattice structures and their laser-induced changes on the crucial picosecond time scale. —BMS

References

1. G. Mourou, S. Williamson, Appl. Phys. Lett. **41**, 44 (1982).
2. J. Van Vechten, A. D. Compaan, Solid State Commun. **39**, 867 (1981).

Laser-driven electron–positron colliders

By 1987, two very large colliding-beam accelerators, LEP at CERN and SLC at SLAC, should be providing high-energy physicists with 100-GeV collisions of electrons and positrons. But what of the next generation? With the 27-km-circumference LEP storage ring costing about $400 million and consuming as much electrical power as a city of 150 000 souls, can one really afford to build significantly larger e^-e^+ accelerators at the turn of the next century?

Head-on linear colliders (of which SLC is a hybrid prototype), with no synchrotron radiation loss, would have substantially lower power bills. But a pair of conventional rf linacs firing 300-GeV pulses of electrons and positrons at each other would constitute a structure almost *forty* kilometers long.

What one wants for the next generation, therefore, is a technology that can provide a significantly higher accelerating gradient than the 15 or 20 MeV per meter to which rf linacs such as the SLAC 2-mile linac are limited. The extraordinarily intense electric fields that high-power lasers can generate offer a promising possibility. To investigate this possibility, a five-day Workshop on the Laser Acceleration of Particles was convened at Los Alamos last February. The Workshop looked in some detail at a half-dozen different laser accelerating schemes, some of which may eventually prove capable of accelerating electrons (and positrons) to hundreds of GeV with a gradient of several hundred MeV/meter.

For some visionaries, such an order-of-magnitude increase over presently available accelerating gradients is still too modest. At a conference on future accelerator technologies held at Oxford in October, a number of the ideas developed during and following the Los Alamos workshop were communicated to the European high-energy-physics community. In his introductory remarks, Abdus Salam (Imperial College, London, and International Centre for Theoretical Physics, Trieste) challenged the participants to come up with a concept that could lead to a practical 100-TeV (10^{14} eV) accelerator early in the next century. While Andrew Sessler (Lawrence Berkeley Lab), chairman of the Los Alamos workshop organizing committee, thinks it more prudent to concentrate first on laser techniques that would offer a *single* order-of-magnitude advance over presently available accelerating gradients, Robert Palmer (Brookhaven) is urging the development of other laser accelerating schemes that he believes might indeed yield practical 10- or 100-TeV machines in twenty or thirty years.

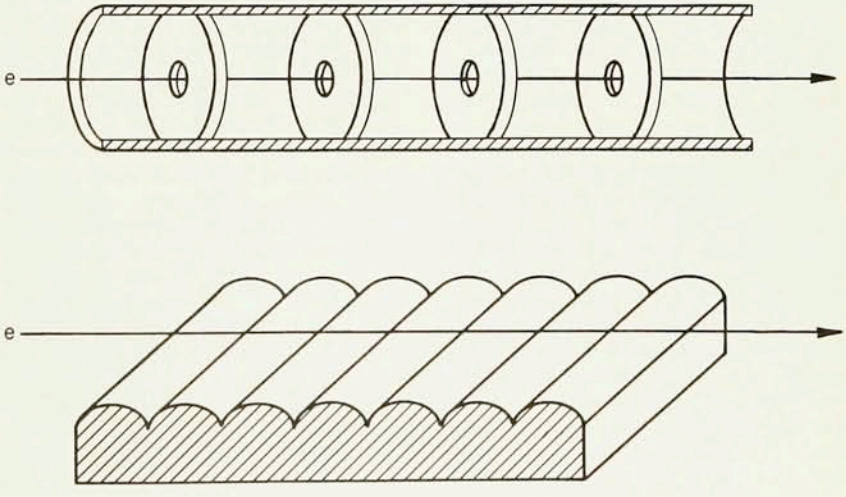
Although the Los Alamos workshop dealt primarily with the acceleration of electrons and positrons, these laser accelerating ideas are presumably also suitable for protons and ions. Because such heavier particles present much less of a synchrotron-radiation problem, we will have a 1000×1000-GeV $\bar{p}p$ synchrotron collider with a diameter of only 2 km—the Tevatron at Fermilab—by mid-decade. Frank Cole (Fermilab) suggests that a 20×20-TeV $\bar{p}p$ collider is a reasonable prospect for the first generation of laser accelerators.

Laser accelerating mechanisms. Because the electric field of electromagnetic radiation is always transverse to its direction of propagation, one cannot accelerate a charged particle simply by shining intense laser light at it. One needs a trick to couple the radiation to the particle by producing an accelerating field component parallel to the particle's motion. Such tricks fall into three broad categories:

▶ Media accelerators, in which the radiation couples to the particle by way of a medium through which the radiation propagates. Inverse Čerenkov acceleration and plasma beat-wave acceleration are two examples of this idea—exploiting, respectively, the refractive index of the medium and the electrostatic fields generated by density waves in a plasma.

▶ Near-field accelerators, in which the laser beam propagates within a wavelength of a periodic conducting structure. This is also the basic idea of the conventional rf linac. Substituting 1-cm radiation from a free-electron laser for the usual radio-frequency drive, one hopes to reduce linac dimensions by an order of magnitude. A far more radical miniaturization might be achieved by employing 10-micron, CO_2-laser drive—in which case one would use a 10-micron grating in place of the usual linac structure.

▶ Far-field accelerators would couple the particle beam to the laser radiation by perturbing the particles' trajectories to include an oscillating transverse component in phase with the radiation. In an inverse free-electron-laser accelerator this would be done by passing the particle through a magnetic wiggler of the kind used in free-electron lasers (PHYSICS TODAY, July 1979, page 19). Another such scheme, the



In all near-field accelerators, a periodic conducting structure converts incident electromagnetic radiation to an accelerating mode, with phase velocity matched to the beam. The accelerating field occurs only within a wavelength of the conducting surface. The conventional rf electron linac (top) uses periodic irises to slow the phase velocity. Sessler proposes scaling such structures down by using 1-cm radiation from a free-electron laser in place of the rf drive. Palmer proposes a more radical miniaturization, using a 10-micron laser drive, effectively reducing the linac structure to a 10-micron diffraction grating (bottom).