

ULTRAFAST OPTICS

Electrons catch a terahertz wave

Far-infrared fields control ultrashort electron pulses

By Claus Ropers

Electron microscopy and diffraction are incredibly successful techniques for studying the structure of matter on the atomic scale. Making these techniques “ultrafast”—that is, using pulses of femtosecond duration—provides us with unprecedented microscopic vistas into how materials evolve on time scales typical of atomic motions (1, 2). One of the key challenges in this research area is the controlled generation of short flashes of electrons. On page 429 of this issue, Kealhofer *et al.* demonstrate a powerful means to shape electron beams in time through their interaction with terahertz electromagnetic fields (3).

Producing short bursts of electrons is far from easy, although the first step is straightforward: Illuminate a metal surface with a laser pulse, and electrons will be emitted into vacuum by the photoelectric effect. Unfortunately, these electrons will have different velocities, and although they may be ejected all at once, they will spread apart as they fly away from the surface (see the figure). Moreover, if many electrons per pulse are

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produced, they will repel each other by Coulomb forces. Both effects lead to a stretching of the pulse as it propagates. However, the time resolution of an ultrafast electron diffraction or microscopy experiment crucially depends on the electron pulse duration at the sample position, which should be as short as possible.

A number of strategies have been developed to alleviate this pulse spreading, in-

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volving the use of compact electron guns (4), nanoscopic electron-source designs (5–7), or active manipulation of the electron bunch (8, 9). A prominent example of this latter approach involves radio-frequency technology (8): An already stretched electron pulse is injected into a microwave cavity at exactly the right moment, such that the electrons at the front of the pulse

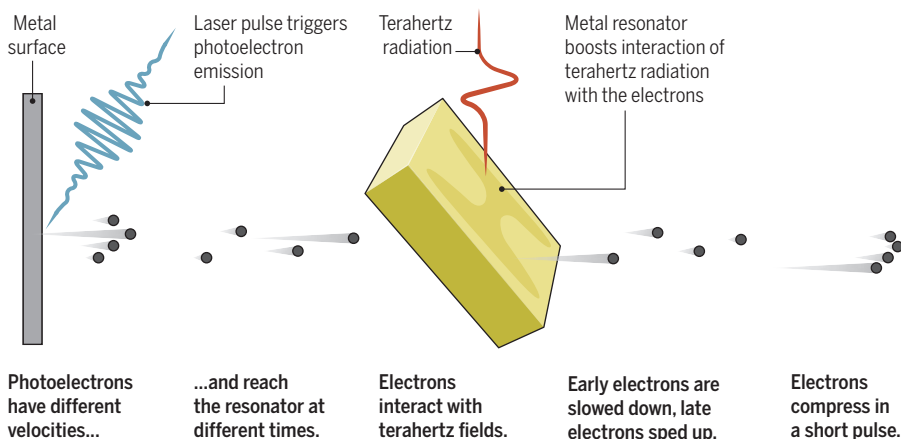
are slowed down a little by the microwave field, while the trailing ones are sped up by just the right amount. If the strength of the fields and the timing are perfect, one can make sure that the electrons will all meet up at a point further downstream in the beam; the pulse has successfully been compressed. A number of laboratories worldwide have mastered and continuously optimized this technique. It requires a synchronization of high-power microwave electronics with pulsed laser sources, but comes with the drawback of a small but unavoidable jitter in timing.

Kealhofer *et al.* take a different route that uses only the laser field itself, together with some nonlinear optics, to produce, compress, and measure the electron pulses. In this way, there are virtually no synchronization issues remaining. In particular, they use terahertz fields—far-infrared radiation at the intersection of optics and microwave electronics—to act on the electrons within bowtie-shaped microstructures enhancing the fields (see the figure). The compression that slows early electrons and accelerates late electrons is achieved by tilting one of these structures, thus allowing the terahertz fields to act in the forward direction.

The authors then characterize the electron pulses in what is essentially a miniaturized, extremely fast version of a cathode ray tube used in oscilloscopes or television tubes: The terahertz fields in a second microstructure deflect the electron beam on a screen, and the width of the resultant streak evidences the electron pulse duration.

Using their setup, Kealhofer *et al.* were able to measure electron pulse durations of 75 fs, a factor of greater than ten reduction from the value in the absence of the terahertz compression, and with negligible long-term jitter of only a few femtoseconds. With data of impressive quality, the study successfully joins various concepts involving intense terahertz radiation, such as near-field streaking (10), electron acceleration (11), and a proposed streak camera (12), to create a terahertz-driven electron-pulse beamline for time-resolved imaging and diffraction applications.

So far, the scheme has been applied to pulses containing only a single electron on average, thus avoiding the Coulomb repulsion problem. Yet there is an ongoing debate as to the effectiveness of single-electron



Terahertz pulse compression. Electrons with a distribution of velocities are ejected from a metal surface by a laser pulse shown in blue. As the electron ensemble propagates, it spreads out in time and space. A properly timed terahertz pulse (red) incident on an apertured microstructure decelerates the early-arriving electrons while accelerating those arriving later. This process causes a compression of the electron pulse, which facilitates experiments with enhanced temporal resolution.

pulses in ultrafast imaging. Many processes are irreversible and must be probed with one massive electron burst, whereas others can be sampled in a stroboscopic fashion with many single- or few-electron pulses. It will be interesting to see whether and how effective terahertz compression techniques can also be applied to electron pulses containing large numbers of electrons. Most likely, adaptations would have to be made to account for the stronger initial pulse stretching arising from Coulomb forces. Lower-frequency fields, and perhaps cascaded stages with multiple interaction regions operating at different frequencies, may be a solution.

The rapid ongoing development in high-power laser sources is clearly widening the scope of possibilities, and Kealhofer *et al.* provide a very elegant solution to all-optical temporal beam shaping and characterization that could serve as the blueprint for future developments. More generally, the work is an excellent example of a trend toward optical manipulation of beams of electrons (9). In electron microscopy, lithographically produced phase plates are gaining interest as a means of preparing well-defined orbital angular momentum states and achieving miniaturized aberration correction. However, these devices are static and yield control only over the transverse properties of the beam, perpendicular to its propagation direction. Time-dependent fields, on the other hand, are required to influence the beam along its direction. In the future, quantum coherent optical interactions with free electron states (2, 13, 14) will enable tailored and configurable control over both the longitudinal and transverse variables of the quantum mechanical electron wave function, possibly resulting in electron microscopy and spectroscopy with attosecond precision. Beyond microscopy, the growing means to exert optical control over free electrons may also have an impact on other research fields, including benefits to the spatial and temporal beam properties of synchrotrons and free-electron laser sources. ■

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CLIMATE CHANGE

Mineral clues to past volcanism

A study of zircon minerals from around the world shows that volcanism is a key driver of long-term climate change

By Lee Kump

Over tens to hundreds of millions of years, Earth's climate has repeatedly swung from icehouse, with large ice sheets like today, to greenhouse, when even near-polar climates were temperate (1). The modern paradigm attributes these swings to a dynamic interplay of volcanism, which spews carbon dioxide (CO₂) into the atmosphere, and the chemical weathering of rocks on land, which removes CO₂ from the atmosphere (2). Documenting how these driving forces have varied through time has been a challenge. On page 444 of this issue, McKenzie *et al.* (3) argue that volcanic CO₂ emissions have been the main driver of climate change over the past several hundred million years.

The authors base their conclusion on an analysis of the age distribution of zircon (zirconium silicate) grains from sedimentary rocks around the world. Zircons crystallize from magmas, especially when oceanic plates are subducted beneath continental plates and form continental volcanic arcs, like those in the Andes and the Cascade volcanoes in North America. Because zircon grains are highly resistant to weathering, they are eroded as sand-sized grains, deposited in sedimentary basins, and incorporated into sedimentary rocks. They survive several rounds of subsequent erosion, deposition, burial, lithification, metamorphism, and uplift. As a result, zircons in a given sedimentary rock include some that may have just formed (often associated with volcanic activity) and others of various older ages.

Because zircons contain uranium, thorium, and lead impurities, their ages can be determined precisely through isotope analysis. A high proportion of young zircons in rocks of a given age suggests high volcanic activity at the time of sediment deposition. If, instead, old zircons predominate, volcanic activity is presumed to have been low when the sediment formed.

When McKenzie *et al.* compiled published analyses of ~120,000 zircons, they found an intriguing pattern: Greenhouse intervals, like the Cambrian (~500 million years ago) and Jurassic/Cretaceous (from 200 to 65

million years ago) have sedimentary rocks with a relatively high proportion of young zircons. In contrast, young zircons were rare in the Neoproterozoic (the "Snowball Earth" interval ~700 million years ago), the Carboniferous and early Permian (a time of extensive glaciation ~300 million years ago), and the Cenozoic (the past 65 million years, during which extensive ice sheets have spread across Antarctica, Greenland, and cyclically across North America and Northern Europe).

The swings from icehouse to greenhouse roughly follow the drift of the continents (1). The large continents of the icehouse Neoproterozoic rifted apart, were widely dispersed by the greenhouse Cambrian, and then gradually amalgamated into the supercontinent Pangea by the icehouse Carboniferous-Permian. They then rifted

"The correspondence... between zircon age distributions and long-term climate change is compelling evidence that volcanism has been an important climate driver for the past 700 million years."

apart and dispersed again during the greenhouse Jurassic-Cretaceous. McKenzie *et al.* argue that rifting and dispersal are driven by the creation of new ocean crust, which also demands that old ocean crust be destroyed in subduction zones. Subduction fuels continental arc volcanism, releasing CO₂ to the atmosphere and creating zircons that become part of the sedimentary record. Continental amalgamation leads to the slowing of subduction and a reduction in continental-arc volcanism and associated zircon formation.

The correspondence documented by McKenzie *et al.* between zircon age distributions and long-term climate change is compelling evidence that volcanism has been an important climate driver for the past 700 million years. Of course, a full consideration of the

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