ULTRAFAST SCIENCE

Ultrashort electron probe opportunities

Technology borrowed from electron accelerator and beam physics looks set to push the performance of ultrafast-electron-diffraction-based pump–probe studies of matter.

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A strong desire in ultrafast science is the development of instrumentation that makes it possible to monitor the spatiotemporal dynamics of excited atoms or molecules — yielding important fundamental insights into how different material systems behave. This is typically conducted using pump–probe experiments, where an atomic system is first 'pumped' into an excited state with an ultrashort laser pulse and then 'probed' a short time later with a beam of X-rays or electrons that read out the atomic dynamics as the system relaxes.

For such pump–probe experiments to be successful on an atomic scale it is important that the probe beam has a sufficiently short wavelength to spatially resolve atomic positions and also a sufficiently short duration to observe atomic motion on the sub-100 fs scale. It is also vital that the pump beam and probe are well synchronized with minimal temporal jitter. Now, writing in *Nature Photonics*, Kim and colleagues describe how this feat can be accomplished using high-energy, temporally compressed, ultrashort electron bunches as a probe beam[1].

Recently, the bright, ultrashort X-ray pulses generated by the latest X-ray free-electron laser (XFEL) facilities around the world have become a popular tool for pump–probe experiments. However, the use of a short-duration electron bunch to perform ultrafast electron diffraction (UED) potentially offers an alternative probe approach that is compact, cost effective and can provide complementary information[2,3].

Contrary to X-rays though, electrons are negatively charged particles and obtaining very short bunch durations is hindered by the repulsive space charge forces that cause the bunch to lengthen during propagation. Improving temporal resolution has thus been the main challenge at the core of recent ultrafast electron scattering instrumentation development. Progress in this area has been fuelled by the introduction of relativistic electron photoguns and other accelerator physics techniques such as radio-frequency (RF)-based velocity compression[4,5].

Using an electron beam with relativistic electron energy (having a speed close to the speed of light, *c*) for UED has unique advantages in terms of penetration depth (higher-energy electrons can probe thicker samples) and group velocity mismatch (probe electron beams and pump laser have similar speed).

Most importantly, the use of relativistic energies also suppresses space charge repulsion — the relativistic length contraction phenomenon in the rest frame of the electron beam means that the electron density and hence the repulsion forces are much smaller. Therefore, electrons with megaelectronvolt (MeV) energies can be packed into very short (<100 fs) bunches containing 1–10 million electrons. This opens the way to the study of irreversible ultrafast processes that cannot be investigated by other techniques. In addition, by further taking advantage of RF compression techniques ultrashort bunches of less than 10 fs duration can be obtained[6,7], enabling direct observation of the fastest atomic processes.

Unfortunately, fluctuations in the amplitude and phase of the RF fields used in the MeV acceleration and compression equipment severely impacts the time of arrival of these electron beams with respect to the pump laser. This has thus prevented the use of these beams in pump and probe studies and effectively limited the temporal resolution in MeV UED to above 100 fs.

Now, inspired by accelerator and beam physics, Kim and colleagues introduce a double-bend achromat to compress the electron beam output from a RF photogun, while at the same time reducing its jitter with respect to the pump laser. The double-bend achromat is a particular arrangement of dipole and quadrupole magnets, well known in the design of storage rings and synchrotron light sources, where the trajectory of the output beam does not depend on the beam energy (achromatic condition).

In addition, the dependence of the path length on the electron beam energy can be tuned in such a way to compensate the time-of-arrival dependence on the energy of typical drift regions. As shown in Fig. 1, space charge effects cause the beam exiting the gun to have higher-energy particles at the front and lower-energy particles at the back. After the double-bend achromat beamline this situation can be reversed, leading to a much shorter, compressed beam at the sample plane. In fact, the entire system can be arranged to meet a so-called isochronous condition for the transport from the photocathode in the gun to the sample. In this configuration, fluctuations in the high-power RF waves will not affect the arrival time jitter of the electron beam with respect to the pump laser.

Kim and co-workers apply this concept to a novel instrument that utilizes an RF photogun and a 90° achromatic bend and successfully compress a relativistic electron bunch to less than 20 fs in duration, while simultaneously reducing the timing jitter between the laser pump and the electron probe at the sample to below 30 fs.

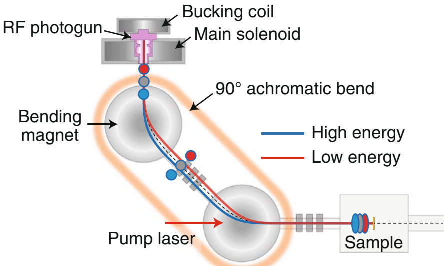

Fig. 1 | The principle of the double-bend compression and jitter suppression. The arc trajectory temporally compresses the electron bunch exiting the gun, while at the same time minimizing the time-of-arrival dependence as a function of electron energy. The red, grey and blue dots represent electrons with different energies — low, medium and high, respectively. The high-energy (blue) electrons are found at the head of the bunch at the gun exit but have a longer trajectory and thus arrive at the sample at the same time as the lower-energy (red) electrons, which are at the tail of the bunch but have a shorter trajectory. Figure adapted with permission from ref. [1], Springer Nature Ltd.

The bunch length and time jitter of the electron beam were measured using a cross-correlation technique based on a laser-generated terahertz wave impinging on a metallic slit located in the electron beam path[8]. When electrons arrive early (late) they get deflected by the terahertz field up (down) so that the time of arrival is directly encoded in the vertical beam position on a screen downstream[8]. An experiment studying the ultrafast structural change in a photoexcited bismuth film — a well-known ultrafast process that has a time constant of 150 fs — was used to validate the terahertz streaking measurements and to determine the instrument's temporal response (31 fs).

With multiple high-energy UED user facilities coming online[9,10], the technique described by Kim and colleagues sets a new milestone in the temporal resolution of MeV UED machines, which will now be able to probe hard phonons in condensed matter as well as the fastest molecular reactions in gas phase. Looking at the future, the authors claim that it might be possible to reach single-digit femtosecond temporal resolution and thus to detect electronic motion.

Many challenges remain for developing the next generation of instrumentation such as improving spatial resolution, signal-to-noise ratio and long-term stability, but we now have a new set of (very fast) eyes to peer into the depths of nature. ❐

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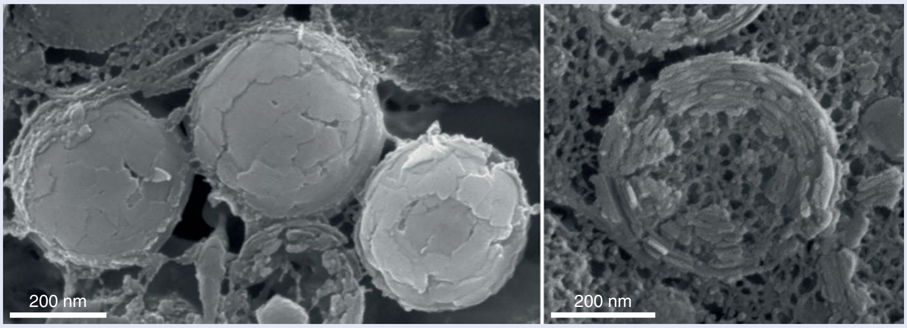
BIOPHOTONICS

Shrimp eye scattering

An ultrathin reflective layer in the eye of certain types of crustacean has been found to consist of an array of highly scattering nanometre-scale hollow spheres of an organic crystal. The findings (B. A. Palmer et al. *Nat. Nanotechnol.* **15**, 138–144; 2020) could provide inspiration for the design of new types of photonic crystal. Benjamin Palmer and co-workers from various institutions in Israel studied the compound eyes in the so-called whiteleg shrimp (*Litopenaeus vannamei*). They discovered that a reflective layer, the tapetum, at the back of the shrimp's eye, near the photon-absorbing retinal cells was found to consist of an arrangement of highly birefringent nanoscopic spheres of crystalline isoxanthopterin with an optimized lamellar structure (pictured) and exceptional scattering strength.



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Images obtained by transmission electron microscopy and cryoscanning electron microscopy showed that the isoxanthopterin particles were composed of many nanoscale platelets arranged in concentric lamellae around a hollow core. The platelets were irregular planar polygons with dimensions of approximately 50 × 50 × 10 nm and formed hollow spheres with an average diameter of 330 nm and a shell thickness of 70 nm.

Biogenic isoxanthopterin is a biaxial crystal, possessing three principal refractive indices along the *a*, *b* and *c* directions: $n_a = 1.40$, $n_b = 2.02$ and $n_c = 1.90$. The nanoparticles were composed of single-crystal isoxanthopterin plates, with the *a* axes of the individual platelets projecting radially from the surface of the sphere to form spherically symmetric birefringent particles. As a result, the isoxanthopterin particles exhibited as a uniaxial material with an in-plane ordinary refractive index (n_o) of 1.96 (the average of n_b and n_c) and an out-of-plane extraordinary refractive index (n_e) of 1.40.

The back-scattering efficiency of the isoxanthopterin particles was calculated using a modified Mie theory and it was found that the back-scattering efficiency of the birefringent nanoparticles was approximately two times higher than that of the effective isotropic material (with a refractive index equal to the average of n_a, n_b and n_c). Most interestingly, the maximum back-scattering efficiency with a minimum amount of material was obtained at the thickness of 70 nm, which was equivalent to the experimentally observed shell thickness. The design is thought to maximize the sensitivity and acuity of the shrimp's eyes.

"Our findings provide a rationalization for the optical functionality of the tapetum and offer inspiration for the development of previously unexplored photonic materials, made from spherically symmetric birefringent particles", Palmer said. ❐

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