



Figure 1 | Electron tunnelling and interference. The electrons in a solid form energy bands (here, 1–3) and can tunnel between these when a strong, terahertz-frequency field is applied. Band 1 can be reached directly from band 2 (solid arrow), but also through the path 2 to 3 to 1 (dotted arrows). The sign (+ or –) of the electrons’ wavefunctions (Φ ; solid and dotted blue lines) after each tunnelling event is determined by the polarity of the applied field (one cycle, shown in green). Hohenleutner *et al.*² observe that the electrons’ wavefunction does not change sign after each tunnelling event for positive field polarity (left), but does change sign for negative polarity (right). As a result, for positive polarity the wavefunctions interfere constructively (red line in band 1, left), whereas for negative polarity they interfere destructively (red line in band 1, right). Consequently, for positive polarities of the applied field only, the crystal emits a pulse of radiation (not shown) at frequencies corresponding to high harmonics of the applied field’s frequency.

and infrared light that are only several femtoseconds long. These pulses reveal the signature of the electronic states populated by the tunnelling process and allow accurate tracing of the dynamics of the crystal’s electrons. The authors record the high-harmonic radiation that is emitted from a gallium selenide crystal subjected to pulses centred at a frequency of about 30 THz. Their experiment measures the structure of the high-harmonic spectrum with femtosecond precision, providing greater insight into the electron dynamics than would be possible by measurements of the spectrum without ultrafast temporal resolution. Using sophisticated optical techniques, the authors are able to pinpoint the moment at which the high harmonics are generated during a terahertz-frequency pulse that is only a few cycles long.

These results would have been hard to interpret without the in-depth theoretical understanding and modelling that Hohenleutner and colleagues use to complement their experiment. The data can be explained by invoking inter-band tunnelling, but the authors show that more than two electron energy bands are involved — five are required. Moreover, quantum interference between the various tunnelling paths needs to be invoked for a proper explanation of the observations. In classic interference, waves combine constructively or destructively depending on whether they arrive at a particular spot in or out of phase, which hinges on the difference between the

distances the waves have covered. Analogously, in this experiment, different electron-excitation pathways give rise to quantum interference and couple the five energy bands together.

Using numerical modelling, the authors artificially ‘switched off’ the interference between the tunnelling pathways, demonstrating that the quantum interference is essential to fit the experimental data. The modelling also shows that the polarity of the driving electric field determines whether the interference is constructive or destructive (Fig. 1). It turns out that high-harmonic radiation is emitted only when the driving electric field is at its peak, and for one polarity of the field only. When this happens, the intensity of the emitted high-harmonic pulse is enhanced by a factor of 30, owing to constructive interference between the tunnelling pathways, compared with the case without interference.

High-harmonic emission in solids has been a vibrant area of research since it was shown that the excitation of crystals by a strong, long-wavelength electromagnetic field generates radiation at high-harmonic frequencies of the driving field’s fundamental frequency^{5,6}. The strong-field physics of individual atoms in the gas phase is well understood, but the different scaling of the maximum harmonic frequency with the strength of the incident field in solids had hinted at different underlying processes⁵. However, a recent comparative study⁷ found that some aspects of the gas-physics picture

apply to solids too, such as the importance of controlling the electrons’ pathways so as to generate high-harmonic radiation. In another study⁸, researchers applied optical pulses of sub-period duration to a silicon dioxide crystal and showed that the observed high-harmonic radiation reaches frequencies of 8,500 THz (corresponding to the extreme-ultraviolet domain), a record value for intra-band currents induced in a solid.

From insights into such observations, new ways may be devised to control the phase of the electron wavefunction in crystals, for instance through the instantaneous modification of the band structure by external electric fields. Related work with solids subjected to strong fields has also shown that the material may be reversibly changed from being a dielectric (insulator) to a semi-metal (conductor) in a femtosecond^{9–11}.

These studies, and Hohenleutner and colleagues’ work in particular, open the door to using electrons in solids as a quantum-physics playground. For example, fully understanding the radiation mechanism will allow us to infer the electron wavefunctions from the emitted high harmonics. This might enable the tomographic reconstruction of a crystal’s electronic band structure. Moreover, the ultra-short time-scales observed in this work could open up new ways of quantum information processing, in which information will be encoded in the electrons’ wavefunctions. Other applications can also be envisaged, including the generation of intense sources of coherent extreme-ultraviolet radiation. Because of the complex nature of any condensed material, further strong-field studies of crystals, disordered solids, and even liquids might lead to other surprise discoveries. ■

Peter Hommelhoff and Takuya Higuchi

are in the Department of Physics,
Friedrich–Alexander–Universität (FAU)
Erlangen–Nürnberg, Erlangen 91058, Germany.
e-mail: peter.hommelhoff@fau.de

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CORRECTION

The News & Views article ‘Astrochemistry: Fullerene solves an interstellar puzzle’ by Pascale Ehrenfreund and Bernard Foing (*Nature* **523**, 296–297; 2015) omitted the relevant reference citations and full credit information for Figure 1. This has now been corrected online.