

squares. The precursors would then seed the growth of a bulk SbTe crystalline phase. To make sure the bulk crystal could grow seamlessly from the precursors, the ABAB squares would need to closely match the geometry and spacing of SbTe's rock-salt lattice.

Guided by density functional theory calculations, the team whittled the three dozen contenders down to one, scandium. Molecular dynamics simulations confirmed that under typical set-operation conditions, ScTe squares survived 10 times as long as their GST counterparts and, as shown in figure 2, could effectively seed the growth of SbTe crystals. But the proof was in the experimental pudding: The researchers fashioned a prototype memory cell from their scandium-antimony-tellurium concoction, dubbed SST, and showed that they could perform set operations with the device in just 700 ps.

"I was surprised that such fast crystallization is possible," says SangBum Kim, a research scientist at IBM. The write-speed improvements are especially astonishing, he adds, because they come at no loss to data retention time. "Usually there's a trade-off."

Indeed, the Xi'an Jiaotong-SIMIT collaboration effectively lowered the energy barrier for the transition between logic

states, and intuitively, that should also raise the odds of unintended bit flips. But SST's estimated data-retention time is actually slightly better than that of GST.

Zhang chalks up the counterintuitive result to competing kinetic effects: Although the scandium-laced precursors lower the energy barrier for nucleation, they also increase viscosity, which stifles diffusion and thereby slows proliferation of the crystalline phase. At the elevated temperatures that prevail during a set operation, the energy-barrier effect wins out. But under the much cooler conditions at which bits are stored, the viscous effect wins.

Big step for big data?

Although the switching speeds of the SST-based device are suitable for a universal memory, the endurance currently is not. The scandium-doped cells become unreliable after about a million logic cycles, too few to stand up to the rigors of cache operations. Zhang and his colleagues think they can improve endurance with tweaks to the device setup and material composition—and there's no fundamental reason they can't. But they'll need to do so by orders of magnitude to make the technology a viable cache memory.

Even if phase-change memory never fully supplants SRAM, it is already durable, stable, and fast enough to contend with magnetoresistive random-access memory and resistive random-access memory in the competition to supplant DRAM as the workhorse memory of next-generation computers. Unlike DRAM, phase-change memory is nonvolatile: It retains information even without a power supply. That and its high storage density make it especially attractive for energy-intensive big-data applications such as machine learning and neuromorphic computing.

"If you use these nonvolatile memory devices with neuromorphic computing, you can save a lot of energy and power," says Kim. "And you could accelerate computations by orders of magnitude."

Ashley G. Smart

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Attosecond measurements reach electronvolt precision

With a train of ultrashort light pulses, researchers disentangle energetically similar photoionization channels—and solve a seven-year-old puzzle.

If you know where you are, you don't know where you're going, and if you know where you're going, you don't know where you are: The Heisenberg uncertainty principle fundamentally limits how precisely position and momentum can simultaneously be known. Another formulation of the principle, linking the uncertainties of energy and time, would seem to suggest that ultrafast measurements—such as the attosecond experiments that probe electronic motion in atoms, molecules, and solids—can't be both temporally and spectrally precise.

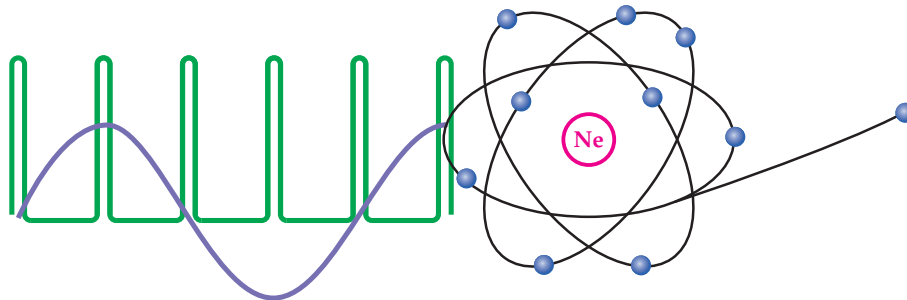


FIGURE 1. PHOTOIONIZATION OF NEON probed by a train of attosecond pulses (green) combined with an IR wave (purple). The ionization signal exhibits constructive or destructive interference, depending on the photoionization time delay and the timing offset of the two waves.

But that's not quite true. The uncertainty principle does place a lower bound on the product of the time and energy uncertainties of an isolated electromagnetic pulse. An experiment's temporal precision, however, is not limited by the duration of the pulses used in it; rather,

it's often determined by the degree of control over the relative timing of two pulses. Extreme UV (XUV) pulses with durations on the order of 100 attoseconds can nevertheless make measurements with few-attosecond precision.

Spectral precision is another matter.

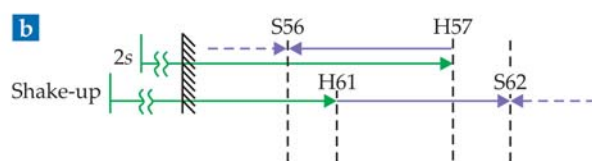
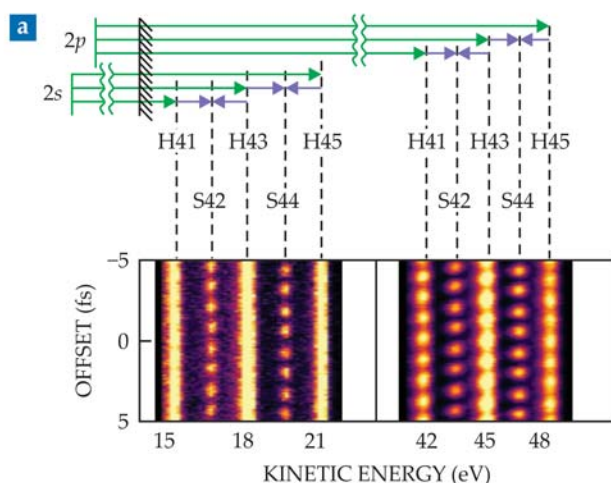


FIGURE 2. SEPARATING IONIZATION PROCESSES. (a) When neon is ionized by a combination of extreme UV (green) and IR (purple) photons, electrons from the 2s and 2p subshells are easily distinguished by their kinetic energy. The relative photoionization delay at each two-photon sideband energy (S42 and S44 shown here) can be inferred from the phase difference of the oscillating ionization signals. (b) Shake-up and 2s electrons overlap in energy. But their sideband signals (S56 and S62 here) can still be resolved. (Adapted from ref. 1.)

The spectral bandwidth of single attosecond XUV pulses is typically around 10–20 eV. Processes—such as atomic ionization channels—whose energies differ by less than that seem irretrievably blurred together.

Now Marcus Isinger at Lund University in Sweden, his thesis adviser Anne L’Huillier, and their colleagues have performed an attosecond experiment¹ that resolves two ionization processes in neon separated by just 7.4 eV. Instead of isolated attosecond pulses, they used a train of pulses (depicted in green in figure 1), similar to an optical frequency comb, created from a phase-locked superposition of high harmonics of an IR fundamental (purple). Each pulse in the train has the necessary subfemtosecond duration, and each tooth in the frequency comb has bandwidth less than 1 eV.

The technique, called RABBIT (for “reconstruction of attosecond beating by interference of two-photon transitions”), has been around since 2001.² But by using it to disentangle processes so close in energy, the Lund researchers have solved a puzzle that’s plagued the attosecond community for seven years.

Photoionization delay

In a landmark 2010 experiment, researchers at the Max Planck Institute of Quantum Optics in Garching, Germany, showed that when an atom absorbs light and ejects an electron, the absorption and emission don’t happen at the same instant.³ Their technique, called attosecond streaking, combines an XUV pulse some 200 attoseconds in duration with a longer IR pulse. The XUV radiation ionizes the atoms, and the IR field, depend-

ing on its phase at the moment of ionization, either accelerates or decelerates the liberated electrons. A streaking spectrogram, which records the electron kinetic energy as a function of the timing offset between the XUV and IR pulses, displays clear oscillations whose phase depends on the delay between XUV absorption and electron emission.

Because there’s no way to tell where zero is on the offset axis, a single streaking spectrogram says nothing about the absolute photoionization delay. Simultaneously captured spectrograms of two photoionization processes, however, can reveal the difference between their time delays. The Garching researchers used an XUV energy centered at 106 eV, enough to pluck an electron from either the 2s or 2p subshell of the Ne atoms they studied. The binding energy of a 2s electron exceeds that of a 2p electron by 27 eV, so the spectrograms differ in energy by the same amount—comfortably in excess of the 14 eV bandwidth of the XUV pulse.

Comparing the two spectrograms showed that 2p ionization lags behind 2s ionization by 21 ± 5 attoseconds. That result had an important implication for the emerging field of attosecond science: The time at which a photoelectron is released could no longer be considered a reliable measure of when an attosecond pulse strikes its target.

But there was a problem. Of the 21 attosecond difference in delay times, theory could account for at most 9 attoseconds. Computational studies of Ne photoionization are unavoidably inexact: The time-dependent Schrödinger equation doesn’t admit exact solutions for atoms with more than 2 electrons, and

Ne has 10. But approximations are extremely sophisticated, and as theorists have refined their calculations over the years, their results have converged on a delay that’s much shorter than the Garching measurement. If the discrepancy stemmed from some undiscovered systematic error—theoretical or experimental—it could cast broad doubt on attosecond results.

Shake-up

One reason multiparticle quantum computation is so hard is because the particles are correlated: A 10-electron wavefunction, for example, can’t be decomposed into the products of 10 one-electron wavefunctions. In computational chemistry, it’s usually necessary to account for electron correlations to get results that are even qualitatively correct. And in atomic systems, correlations engender processes in which two or more electrons move simultaneously in response to a single stimulus. For example, in Ne, a single photon might remove an electron from a 2p orbital and promote a second 2p electron to the 3p subshell. That so-called shake-up process requires just 7.4 eV more energy than direct ionization from the 2s subshell. It would have gone unresolved in the Garching experiment.

“We thought about shake-up seven years ago,” says Vladislav Yakovlev, who coordinated the theoretical support of the experiment, “but we didn’t have means to properly simulate it in neon, and we didn’t believe it would be the major cause of the discrepancy.” If shake-up played an important role, the reasoning went, the measured delay would be sensitive to the XUV pulse

bandwidth. But there was no significant difference between delays measured with shorter, broadband pulses and those measured with longer, narrower-band ones.

By 2014 Johannes Feist of the Autonomous University of Madrid and collaborators had developed their calculations to the point that they could simulate shake-up's effect on the Ne streaking spectra.⁴ They found that shake-up was in fact prevalent enough to affect the measured delay. More recently, Garching's Martin Schultze and colleagues used attosecond streaking to study shake-up timing in helium. They found that direct ionization lagged shake-up by between 5 and 12 attoseconds, depending on the ionization energy.⁵ Shake-up in He is separated in energy from direct ionization by more than 40 eV, so it can be resolved in a streaking experiment. Measuring shake-up in Ne would require a different method.

Spectral resolution

The essence of the Lund RABBIT experiment is shown in figure 2. The attosecond pulse train is composed only of

odd-numbered XUV harmonics of the IR fundamental frequency—indicated by H41, H43, and H45 in figure 2a. Ionization at the energies of even-numbered multiples of the fundamental—such as sidebands S42 and S44—can be initiated in two ways: absorption of one XUV harmonic plus an IR photon or absorption of the next harmonic combined with IR emission.

Those processes can interfere either constructively or destructively, depending on the timing offset of the XUV and IR waves. Adjusting the timing, therefore, creates oscillations in the sideband signals' intensity (not their kinetic energy, as in streaking), as evidenced by the trains of spots at the energies of sidebands S42 and S44 in the figure. The phase of the oscillation depends on the photoionization delay. At the S42 energy, then, the relative delay of 2s and 2p direct ionization corresponds to the vertical displacement between the spot trains at 16.5 eV and 43.5 eV. The delay, which amounts to tens of attoseconds, is too small to be perceptible by eye in the figure, but it can be extracted from a sinusoidal fitting of the data.

Figure 2b shows how shake-up enters the picture. The 7.4 eV difference between 2s ionization and shake-up amounts to a little more than five times the IR fundamental frequency. So the signal from 2s ionization at the energy of a sideband (such as S56) overlaps with the signal from shake-up initiated by a harmonic (such as H61), and vice versa. But the two processes' sideband oscillations—and thus their delays—are well enough separated that they can be measured individually. And importantly, the Lund researchers could confidently conclude that the sideband 2s ionization data in figure 2a are uncontaminated by shake-up.

The results are summarized in figure 3. To avoid overlap between the 2s and 2p ionization signals, the researchers used filters to single out sets of harmonics spanning less than 27 eV; two such sets are shown in yellow and red in the top panel. The bottom panel shows the photoionization delays, each measured relative to 2p direct ionization, at the side-

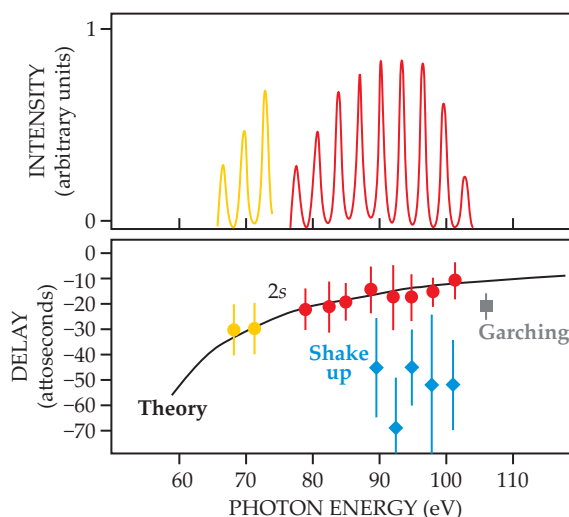


FIGURE 3. PHOTOIONIZATION TIME DELAYS, all shown relative to 2p direct ionization. With the photon spectra shown in the top panel, relative delays can be measured at the sidebands between each pair of neighboring harmonics. The negative delay values mean that 2p direct ionization lags both 2s ionization (yellow and red) and shake up (blue). The results of the Garching experiment³ and the latest theoretical calculations are shown for comparison. (Adapted from ref. 1.)

band energy between each pair of adjacent harmonics. Theoretical calculations and the Garching measurement are shown for comparison.

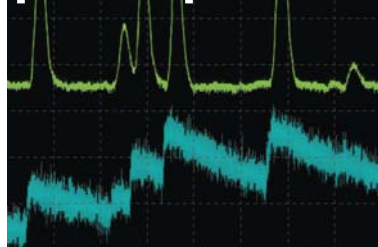
Although the Lund experiment doesn't quite reach the same photon energy the Garching researchers used, the results strongly implicate shake-up as the cause of the earlier discrepancy: The new 2s ionization measurements agree well with calculations, and the Garching measurement is in between the 2s and shake-up delays. With confidence thus restored to their methodology, attosecond researchers are ready to push their understanding to new frontiers. "Neon photoionization in this energy range seems quite understood now," says L'Huillier. "But there are many interesting questions in other energy regions and other systems. This is just the beginning."

Johanna Miller

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