Characterizing behavior of individual electrons during chemical reactions

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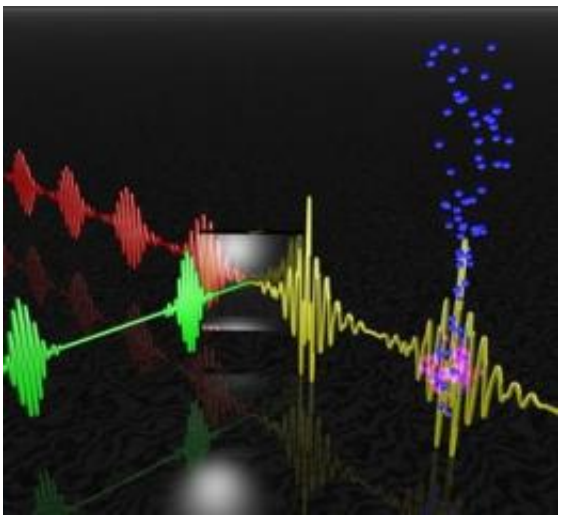

A schematic of a new design for a laser that emits ultrashort pulses of light. Light waves of different frequencies (red and green) are combined to form a new wave (yellow), which in turn passes through a gas (blue). The light excites the atoms of the gas, which release their excess energy as light of an even higher frequency. Image: Shu-Wei Huang

In a paper published in the latest issue of *Nature Photonics*, an international team of researchers takes an important step toward giving physicists the ability to effectively make movies of individual electrons. If the approach pans out, it would provide a way to gather data of unprecedented detail about how individual molecules interact during chemical reactions, with ramifications for not only the basic sciences but chemical engineering and pharmaceutical research as well.

The researchers, eight of whom are from MIT's Research Laboratory of Electronics (RLE), describe a technique that should be able to produce bursts of laser light that last only attoseconds, or billionths of a billionth of a second. The electron in a hydrogen atom takes about 151 attoseconds to orbit the nucleus, so catching it in the act during a chemical reaction would require attosecond pulses.

"If you can generate a pulse that has a shorter duration, then you can investigate dynamics that happen on that time scale," says Franz Kaertner, the adjunct professor in the Department of Electrical Engineering and Computer Science who led the work. "That connects back to this work from [MIT electrical engineering professor Harold 'Doc'] Edgerton, where he was able to make optical flash photography in the microsecond range and nanosecond range."

Attosecond pulses have been demonstrated in the lab before, but they didn't have the intensity required for so-called time-resolved spectroscopy, the technique typically used to measure electron dynamics. Not only should the new approach boost the pulses' intensity, but it should require a simpler setup, too, making it more practical.

Lockstep

The key to producing ultrashort bursts of light is to combine light waves of different frequencies. A wave can be envisioned as a regular, up-and-down squiggle, with the distance between the squiggle's crests indicating its frequency. When two waves intersect, they reinforce each other where their crests overlap, but the trough of one can cancel out the crest of another. The right combination of waves can thus produce a new wave with a radically different shape.

Other researchers have tried to produce short bursts of light by combining laser beams, but they've used a separate laser for each beam.

That makes it very difficult to synchronize the beams so that their troughs and crests coincide exactly where intended. The RLE researchers and their colleagues at the University of Sydney, Politecnico di Milano and Hamburg University instead pass a single laser beam through a crystal that splits it into beams of different frequencies. Because the beams are derived from a single source, they remain perfectly synchronized.

Short and shorter

Although this yields very short pulses of light, they're still not on the scale of attoseconds. So the next step in the process would be to send the pulses through a gas. When particles of laser light — photons — strike the atoms of the gas, they're absorbed, but usually, their energy is immediately re-emitted as new photons. Those photons, however, have frequencies that can be many times that of the original photons. And higher frequencies mean even shorter bursts of light.

The RLE researchers, however, have not yet performed this final step. Currently, they pass their laser beam through two amplifiers to increase its energy, but it needs more energy still to elicit enough higher-frequency photons from the gas. Adding another amplifier, the researchers say, should do the trick, but it does pose some engineering challenges.

Ian Walmsley, a physics professor at the University of Oxford and head of the university's Ultrafast Quantum Optics Group, says that the modularity of the researchers' design is one of its strengths. "The clever design that Franz and his collaborators have come up with is to use something where it's very straightforward to have all of these boxes work in a sort of plug-and-play fashion, a synchronous fashion," Walmsley says. "This new approach, I think, is a very important one because it allows, in principle, the generation of much higher energies in these very

short pulses."

Walmsley suspects, however, that adding the final step in the RLE researchers' process "won't be a trivial matter of turning up the pulse energy and getting shorter and shorter pulses." But, he adds, "there are some promising things that they can do with that, I think. It might be a bit of a challenge, but it could be done."

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