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A method to resolve quantum interference between photoionization pathways with attosecond resolution

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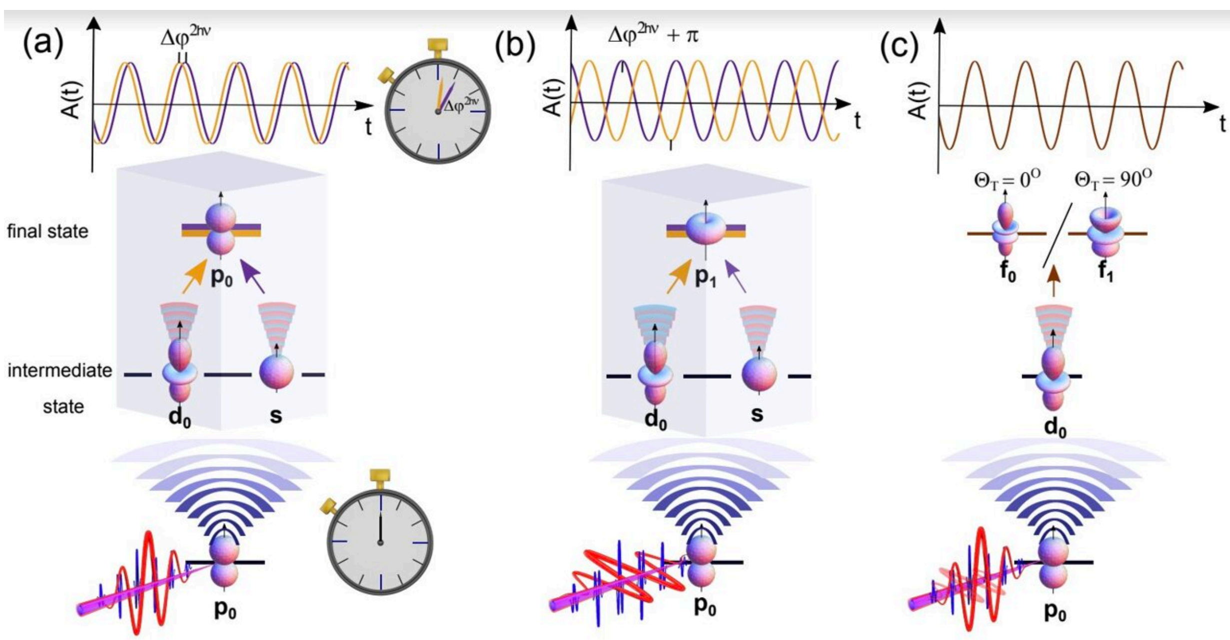

Figure outlining the mechanism in the context of the double-slit paradigm. Credit: Jiang et al, *Physical Review Letters* (2023). DOI: 10.1103/PhysRevLett.131.203201

The field of attosecond physics was established with the mission of exploring light–matter interactions at unprecedented time resolutions. Recent advancements in this field have allowed physicists to shed new light on the quantum dynamics of charge carriers in atoms and molecules

A technique that has proved particularly valuable for conducting research in this field is RABBITT (i.e., the Reconstruction of Attosecond Beating By Interference of Two-photon Transitions). This promising tool was initially used to characterize ultrashort laser pulses, as part of a research effort that won this year's Nobel Prize, yet it has since also been employed to measure other ultrafast physical phenomena.

Researchers at East China Normal University and Queen's University Belfast recently built on the RABBITT technique to distinctly measure individual contributions in photoionization. Their paper, published in *Physical Review Letters*, introduces a new highly promising method for conducting attosecond physics research.

"The RABBITT technique essentially provides an ultrafast stopwatch for electronic processes, so that we can measure (for instance) the time delay between the ionization of different electrons in an atom," Andrew C. Brown, co-author of the paper, told Phys.org.

"One of the difficulties with these experiments, though, is that when you have multiple, interfering processes, the picture becomes substantially more complex, and we can no longer make concrete claims about the timing of the various mechanisms. In essence, you have too many variables, and insufficiently many equations to solve for them.

"The real genius in Xiaochun and Jian's experiment was to provide more equations, or more accurately, more distinct measurements, which allowed us to unpick the different mechanisms."

In their experiments, Xiaochun Gong and Jian Wu, the authors who led the project, used two laser pulses, which is the standard practice when implementing the RABBITT technique. However, they changed the polarization (i.e., skew angle) of these pulses, to gain further control over the measurements they collected.

Initially, the researchers set out to resolve time delays in photoionization for different emission angles. In other words, they wished to determine if an electron behaves differently when it is emitted in different directions relative to the laser field. Once they started examining the data collected in their experiments, however, they realized that it painted a picture far more complex than what they had anticipated.

"Our current work is also a further step forward with respect to our previous atomic partial wave meter work," Gong said. "Our dream is to push the attosecond photoionization measurement into partial wave level, which is the original definition of the scattering phase shift."

The researchers collected their measurements on helium, neon and argon samples. Examining helium is straightforward, as it only contains two electrons and there is really just one method to ionize it, while neon and argon are far more complex systems.

"More precisely, when you ionize helium, there's only one possible residual ion state," Brown said. "For neon and argon, however, things are significantly more complicated. For one thing, there are more electrons to worry about, and for another, there are multiple residual ion states, all of which contribute in some (previously) unknown way to the measured signal. The way we interpreted/explained this was to think of the classic 'Young's double slit' experiment, wherein light passes through two apertures before being 'measured' on a screen."

In a classical Young's double slit experiment, light passing through two apertures produces an interference pattern on a screen. This is because the waves passing through each aperture arrive at the same location through different routes, resulting in so-called "fringes" of constructive or destructive interference.

"The key for that experiment, and the reason it has formed such a compelling metaphor, especially for quantum theorists, is that you can't tell which slit the light passed through, as this cannot be measured," Brown said. "All you can measure is the interference, and the 'which-way information' is inaccessible."

In the experiments carried out by Brown, Gong and their collaborators the two apertures in classical Young's double slit experiments were two different residual ion states in Neon. In contrast, the interference pattern they measured was the photoelectron angular distribution produced by the two skewed laser pulses.

"By performing the measurement for two different skew angles, and then working out all the different routes the electrons could take to arrive at some final state, we could then solve the equations to give us both the amplitude and phase for each different pathway," Brown said. "In other words, we worked out which slit the electron passed through, and how. "

Most studies in experimental attosecond physics use lightweight theoretical calculations to explain their findings after the fact. However, this project required much more detailed simulations to account for the complex dynamics at play and, in essence, provide a prediction for the experiment to confirm.

"The method we used to reconstruct the different pathways in the experiment has a solid theoretical grounding, but the dynamics are so complex that it would be hard to make an airtight case that the numbers we extract from the experiment are reliable," Brown said. "We performed simulations with the R-matrix with Time-dependence (RMT) code, which can handle all of these dynamics from first principles, and from there we were able to extract the amplitudes and phases directly."

When they compared their experimental results with those from the simulation, they found that they were closely aligned. This suggests that their experiment truly did measure what they theoretically claimed it did.

"In summary, we try to use the laser field to attach one additional phase to the intermediate d-wave," Gong said. "We can identify the s-wave and d-wave, but we can disturb their phase property and observe their final interference property. For example, we can open the box to know the 'quantum cat' to be alive or not, but we can add some perturbation and check whether the box has any response or not, where the responses are a must from the reaction of the cat in it."

The researchers view their proposed experimental method as a "partial wave meter," or in other words a tool that can effectively measure individual contributions in photoionization. Notably, their proposed method is based on two distinct experimental techniques, namely changing the laser polarization and measuring the photoelectrons and ions coincidence, which were not previously used together.

"Our work combined these techniques in such a way as to make this new measurement possible," Brown said. "That's not to say that the measurements were straightforward by any means, but it would not be a surprise to see that same combination of techniques used to make more interesting measurements of ultrafast dynamics in the coming years. "

A further unique aspect of this recent study is the simulation used to validate the team's experimental results. For a long time, scientists have tried to interpret experimental data using theoretical models, yet Brown, Gong and their colleagues decided to use a simulation instead.

"The results that RMT provides are less intuitive because the model is far from simple, Brown explained. "However, by including a description of all of the interesting multielectron effects and doing that in a general way so that you aren't limited to specific atoms or specific laser parameters, we can actually start to lead experiments in this field in a way that just hasn't been possible for the thirty or so years of attoscience up to this point."

The recent work by this team of researchers offers new insight into the fundamental dynamics of photoionization. While Brown, Gong and their collaborators are primarily focusing on the physics of this phenomenon, in the future their efforts could help to identify new strategies to control electrons using light. This could inform the development of ultrafast electronic circuitry and photovoltaic technologies (solar panels), or could perhaps even help to design medical tools that prevent radiation damage to cells.

"We are working on building out a more comprehensive theory of higher-order processes in photoemission," Brown said. "In other words, we are trying to theoretically describe what happens when you absorb multiple (more than two) photons in these RABBITT-type experiments. Although we have this RMT code which can simulate the dynamics from first principles, if you want to interpret the findings you also need some relatively simple model to explain the different pathways."

While working on a theoretical model that can explain the data collected in their experiments, the researchers plan to continue conducting experiments and running simulations at increasingly higher intensity regimes. They hope that this will allow them to further examine transitions from few-photon to multiphoton systems and ultimately to strong-field physics.

"The development of strong field physics is away from the traditional scattering theory and a large gap exists between them," Gong added. "An intermediate bridge is necessary to be built to provide a soft understanding from one photon to multiphoton."

More information: Wenyu Jiang et al, Resolving Quantum Interference Black Box through Attosecond Photoionization Spectroscopy, *Physical Review Letters* (2023). DOI: 10.1103/PhysRevLett.131.203201