Characterizing Fano resonances during recollision

Graham G. Brown, Dong Hyuk Ko, Chunmei Zhang, \* and P. B. Corkum

Joint Attosecond Science Laboratory, Department of Physics,
University of Ottawa, Ottawa Canada K1N 6N5
National Research Council of Canada, 100 Sussex Drive, Ottawa, Canada K1A 0R6

(Dated: October 13, 2020)

When intense light irradiates a quantum system, an ionizing electron recollides with its parent ion within the same light cycle and, during that very brief (few femtosecond) encounter, its kinetic energy sweeps from low to high energy and back \[1\]. Therefore, recollision offers unprecedented time resolution and it is the foundation on which attosecond science is built. For simple systems, recolliding trajectories are shaped by the strong field acting together with the Coulomb potential and they can be readily calculated and measured \[2, 3\]. However, for more complex systems, multielectron effects are also important because they dynamically alter the recolliding wave packet trajectories \[4, 5\]. Here, we theoretically study Fano resonances \[6\], one of the most accessible multielectron effects, and we show how multielectron dynamics can be unambiguously isolated when we use in situ measurement \[7\]. The general class of in situ measurement can provide key information needed for time-dependent ab initio electronic structure theory and will allow us to measure the ultimate time response of matter \[8\].

Attosecond pulses generated through recollision are measured using two broad classes of methods: (i) ex situ \[9\] and (ii) in situ \[10\]. While we will focus on in situ techniques, we begin by briefly explaining ex situ techniques for comparison. Ex situ methods, such as photoelectron streaking, rely on the photoelectric effect to create an electron replica of the attosecond pulse by photoionizing a selected atom in the presence of an infrared pulse \[9, 11\]. For a well-understood photoemitter, attosecond streaking fully characterizes an attosecond XUV pulse. However, from a fundamental physics perspective, underlying any streaking measurement is information about both the generating and measuring atom, including their multielectron dynamics and frequency dependent time delays due to the static structure of the residual ion (the latter is encoded in the phase of the transition moment). These effects can be difficult to deconvolve. In contrast, in situ methods add a perturbing field to the recollision process \[7\] and observe the signal by photon emission. The relative phase between the driving and perturbing fields is used to characterize the electron continuum dynamics. In situ methods are insensitive to the recombination dipole matrix element and associated structural effects \[10\]. That is, they are only sensitive to the recollision electron trajectory dynamics.

The purpose of this letter is to confirm that in situ measurement provides a unique window into the effect of dynamic multielectron interaction during recollision. We do this by investigating how multielectron Fano resonances manifest themselves within the restricted domain of attosecond in situ measurement.

Fano resonances are a broad class of resonances that can occur during photoionization or photorecombination due to electron-electron interaction \[12–15\]. They can result from the interference between a continuum state and a bound doubly excited state that is embedded in the continuum \[16\]. These resonances represent the simplest dynamic electron-electron interaction reproducible by exact numerical methods. Within this context, there are two pathways to recombination, which are depicted in Fig. 1 and justified by the strong field model \[17\] and in the Supplementary Information. The first pathway, labeled as the direct recollisional pathway (3.1 in the figure) corresponds to the well-known three-step model \[1\].

\* chunmei.zhang@uottawa.ca

FIG. 1. An illustration of the recollision process in a one-dimensional helium atom. (1) The electron (blue) tunnels into the continuum from the ground state, denoted by the red arrow, leaving a hole (yellow) in the ion. (2) The electron is accelerated by the field and eventually driven back towards the parent ion. The electron can recombine with the parent ion in two different ways: (3.1) the electron can recombine with the hole which was generated when it left, denoted by the left purple arrow; or (3.2) the electron can exchange energy and momentum with the remaining electron and excite it, denoted by the green arrows, subsequently recombining (right purple arrow) through a doubly-excited state embedded in the continuum (shaded grey area). The interference of these two pathways gives rise to an asymmetric resonance profile.
FIG. 2. The normalized dipole acceleration spectral intensity around the Fano resonances in a simulated one-dimensional helium atom. The driving field has a wavelength of 800 nm, pulse duration of 2.5 fs and a peak intensity of $2 \times 10^{14}$ W cm$^{-2}$. At energies of 50.7 eV and 52.4 eV, two sharp Fano resonant profiles dominate the recollision spectrum and, at higher energies, similar resonances are observed with softer features.

– the electron tunnels into the continuum (1), is propagated by the strong driving field (2), and subsequently recombines into the hole from which it left (3.1). The second pathway, labeled as correlated recollision, involves multielectron interaction. Like in direct recollision, the electron tunnels into the continuum (1); is driven by the strong laser field (2); on its return, the recolliding electron interacts with a bound electron, transferring energy and momentum and thereby exciting the ion; and finally, the electron ion system recombines into the ground state through a doubly excited state (3.2). The interference of these two pathways gives rise to the well-known asymmetric Fano resonance profile, typically seen in photoionization spectra.

Since autoionization is not adequately described by the approximations of contemporary ab initio theory (e.g. Hartree-Fock, time-dependent density functional theory) [18, 19], we simulate a one-dimensional helium atom [20] in order to describe multielectron Fano resonances during recollision (see Supplementary Information for simulation details). The advantage of this model is that it includes quantum mechanically exact exchange and correlation effects, albeit at the expense of reduced dimensionality. The dimensionality has no effect on the general point we wish to make.

The calculated dipole acceleration intensity spectrum without the perturbing field is shown in Fig. 2 for energies in the range 50 - 60 eV, where the most prominent Fano resonances in the spectrum occur. Although we are observing photorecombination, the results agree with previously published XUV photoionization results [20] and they are very similar in form to the photoionization spectrum measured in helium [21]. The resonant structures are different for different photon energies. The lower energy structures are sharper while the higher energy ones are softened. The amplitude and phase around a Fano resonance are highly dependent upon the matrix elements relating the ground, doubly-excited, and continuum states involved in the resonance and can vary considerably, especially in the presence of another coherent channel [22].

The spectral phase of the spectrum shown in Fig. 2 is depicted in Fig. 3. The overall shape of the spectral phase is quadratic, as the spectrum is dominated by the direct-recollision channel due to the narrow linewidths of the Fano resonances. Near the resonances, however, the spectral phase deviates from the direct-recollision behaviour due to growing importance of the correlated channels. The variation of the spectral phase around the resonances at 50.7 and 52.4 eV appear more significant than the higher energy resonances and the total phase is well-described by a Fano resonance in the presence of a linearly chirped field (i.e. the direct-recollision channel). The variation of the resonance amplitude and phase profiles across the spectrum can be attributed to the variation of the recolliding electron wave packet phase. Within the context of photoionization, it has been shown that the shape of Fano resonances in XUV photoionization can be controlled by an infrared perturbing pulse [21]. That is, a perturbing field can modulate the phase of the doubly excited state. In the closely related recollision
FIG. 4. The simulated single-image in situ spectrogram for one-dimensional helium. The driving field has a wavelength of 800 nm, pulse duration of 2.5 fs and a peak intensity of $2 \times 10^{14}$ W cm$^{-2}$. The perturbing field has a wavelength of 800 nm, pulse duration of 8 fs, peak intensity of $2 \times 10^{10}$ W cm$^{-2}$, and with a relative angle of 30 mrad with respect to the driving field. The maximizing time delay for each spectral component is plotted in blue for each grating modulation. Around each Fano resonance, the variation of the maximizing time delay deviates from the expected linear behaviour of the direct-recollision channel. These variations indicate that the electron continuum dynamics are strongly affected by the multielectron interaction leading to the observed Fano resonances.

In conclusion, using the simplest multielectron model that can be solved exactly using numerical methods, we have shown how multielectron effects influence high-harmonic and attosecond pulse generation. We have also confirmed that in situ measurement isolates dynamics in correlated electronic systems. Thus, we have a new tool to study the collision-physics nature of recollision physics and an optical means of characterizing multielectron interaction occurring during recollision. This should allow collision-like measurements in transparent solids, where laser driven recollision also occurs and in situ measurements can be applied.

We have shown that in situ measurement methods, by solely measure the continuum dynamics during electron recollision, provide an unambiguous means of studying these processes without obfuscation due to structural and recombination matrix element effects. The method we propose here is generally applicable to any process in which the momentum dynamics of the recolliding electron are altered with respect to the case of direct recollision, including autoionizing and collective resonances. This makes it a natural means to probe the ultimate time response of electronic matter.
Finally, due to the structural insensitivity of in situ measurement, the difference between in-situ and ex situ measurement on the same system will allow us to determine the amplitude and phase of transition moments over the full spectral region in which high-harmonic radiation can be generated.

The authors thank Drs. Michael Spanner and David Villeneuve for many fruitful discussions. Their demonstration that in-situ measurement methods are insensitive to photoionization time delay caused by static structure had a major impact on our thoughts. We also acknowledge important financial support from the US AFOSR (Grant # FA9550-16-1-0109); the Canadian Canada Research Chairs program; the Natural Sciences and Engineering Research Council of Canada; the Canadian Foundation for Innovation; and the Ontario Research Fund.

[1] P. B. Corkum, Plasma perspective on strong field multiphoton ionization, Phys. Rev. Lett. 71, 1994 (1993)
[2] M. Lewenstein, P. Balcou, M. Y. Ivanov, A. L'Huillier, and P. B. Corkum, Theory of high-harmonic generation by low-frequency laser fields, Phys. Rev. A 49, 2117 (1994)
[3] L. Torlina and O. Smirnova, Coulomb time delays in high harmonic generation, New Journal of Physics 19, 023012 (2017)
[4] A. D. Shiner, B. E. Schmidt, C. Trallero-Herrero, H. J. Wörner, S. Patchkovskii, P. B. Corkum, J.-C. Kieffer, F. Légaré, and D. M. Villeneuve, Probing collective multi-electron dynamics in xenon with high-harmonic spectroscopy, Nature Physics 7, 464 (2011)
[5] P. Pablé and R. Santra, Strong-field many-body physics and the giant enhancement in the high-harmonic spectrum of xenon, Phys. Rev. Lett. 111, 233905 (2013)
[6] M. F. Limonov, M. Y. Rybin, A. N. Podshaly, and Y. S. Kivshar, Fano resonances in photonics, Nature Photonics 11, 543 (2017)
[7] N. Dudovich, O. Smirnova, J. Levesque, Y. Mairesse, M. Y. Ivanov, D. M. Villeneuve, and P. B. Corkum, Measuring and controlling the birth of attosecond xuv pulses, Nature Physics 2, 781 (2006)
[8] J. Breidbach and L. S. Cederbaum, Universal attosecond response to the removal of an electron, Phys. Rev. Lett. 94, 033901 (2005)
[9] P. M. Paul, E. S. Toma, P. Breger, G. Mullot, F. Augé, P. Balcou, H. G. Muller, and P. Agostini, Observation of a train of attosecond pulses from high harmonic generation, Science 292, 1689 (2001)
[10] M. Spanner, J. B. Bertrand, and D. M. Villeneuve, In situ attosecond pulse characterization techniques to measure the electromagnetic phase, Phys. Rev. A 94, 023825 (2016)
[11] Y. Mairesse, A. de Bohan, L. J. Frasinski, H. Merdji, L. C. Dimu, P. Monchicourt, P. Breger, M. Kovačev, R. Taieb, B. Carré, H. G. Muller, and P. Agostini, and P. Salières, Attosecond synchronization of high-harmonic soft x-rays, Science 302, 1540 (2003)
[12] M. Wickenhauser, J. Burgdörfer, F. Krausz, and M. Drescher, Time resolved fano resonances, Phys. Rev. Lett. 94, 023002 (2005)
[13] Z. X. Zhao and C. D. Lin, Theory of laser-assisted autoionization by attosecond light pulses, Phys. Rev. A 71, 060702 (2005)
[14] S. Gilbertson, M. Chini, X. Feng, S. Khan, Y. Wu, and Z. Chang, Monitoring and controlling the electron dynamics in helium with isolated attosecond pulses, Phys. Rev. Lett. 105, 263003 (2010)
[15] H. Geiseler, H. Rottke, N. Zhavoronkov, and W. Sandner, Real-time observation of interference between atomic one-electron and two-electron excitations, Phys. Rev. Lett. 108, 123601 (2012)
[16] U. Fano, Effects of configuration interaction on intensities and phase shifts, Phys. Rev. 124, 1866 (1961)
[17] S. Patchkovskii, O. Smirnova, and M. Spanner, Attosecond control of electron correlations in one-photon ionization and recombination, Journal of Physics B: Atomic, Molecular and Optical Physics 45, 131002 (2012)
[18] M. S. Pindzola, D. C. Griffin, and C. Böttcher, Validity of time-dependent harree-fock theory for the multiphoton ionization of atoms, Phys. Rev. Lett. 66, 2305 (1991)
[19] V. Kapoor, Autoionization in time-dependent density-functional theory, Phys. Rev. A 93, 063408 (2016)
[20] J. Zhao and M. Lein, Probing fano resonances with ultrashort pulses, New Journal of Physics 14, 065003 (2012)
[21] C. Ott, A. Kaldun, P. Raith, K. Meyer, M. Laux, J. Evers, C. H. Keitel, C. H. Greene, and T. Pfeifer, Lorentz meets fano in spectral line shapes: A universal phase and its laser control, Science 340, 716 (2013)
[22] M. Kotur, D. Guénot, A. Jiménez-Galán, D. Kroon, E. W. Larsen, M. Louisy, S. Bengtsson, M. Miranda, J. Mauritsson, C. L. Arnold, S. E. Canton, M. Gisselbrecht, T. Carette, J. M. Dahlström, E. Lindroth, A. Maquet, L. Argenti, F. Martín, and A. L’Huillier, Spectral phase measurement of a fano resonance using tunable attosecond pulses, Nature Communications 7, 10566 (2016)
[23] D. H. Ko, G. G. Brown, C. Zhang, and P. B. Corkum, Near-field imaging of dipole emission modulated by an optical grating (2020), arXiv:1907.01219 [physics.atom-ph]
[24] S. B. Schoun, R. Chirla, J. Wheeler, C. Roedig, P. Agostini, L. F. DiMauro, K. J. Schafer, and M. B. Gaarde, Attosecond pulse shaping around a cooper minimum, Phys. Rev. Lett. 112, 153001 (2014)
[25] G. Orenstein, O. Pedatzur, A. J. Uzan, B. D. Bruner, Y. Mairesse, and N. Dudovich, Isolating strong-field dynamics in molecular systems, Phys. Rev. A 95, 051401 (2017)
[26] G. Vampa, T. J. Hammond, N. Thiré, B. E. Schmidt, F. Légaré, C. R. McDonald, T. Brabec, and P. B. Corkum, Linking high harmonics from gases and solids, Nature 522, 462 (2015)
[27] P.-C. Li, X.-X. Zhou, G.-L. Wang, and Z.-X. Zhao, Isolated sub-30-as pulse generation of an he + ion by an intense few-cycle chirped laser and its high-order harmonic
pulses, Phys. Rev. A 80, 053825 (2009).

[28] P. Bader, S. Blanes, and F. Casas, Solving the schrödinger eigenvalue problem by the imaginary time propagation technique using splitting methods with complex coefficients, The Journal of Chemical Physics 139, 124117 (2013) https://doi.org/10.1063/1.4821126.

[29] M. Feit, J. Fleck, and A. Steiger, Solution of the schrödinger equation by a spectral method, Journal of Computational Physics 47, 412 (1982).

[30] Y. Yu and B. D. Esry, An optimized absorbing potential for ultrafast, strong-field problems, Journal of Physics B: Atomic, Molecular and Optical Physics 51, 095601 (2018).

[31] K. Burnett, V. C. Reed, J. Cooper, and P. L. Knight, Calculation of the background emitted during high-harmonic generation, Phys. Rev. A 45, 3347 (1992).