



# Quantum aspects of attoscience

Carla Figueira de Morisson Faria<sup>1,a</sup> and A. C. Brown<sup>2</sup>

<sup>1</sup> Department of Physics and Astronomy, University College London, Gower Street, London WC1E 6BT, UK

<sup>2</sup> Centre for Light Matter Interaction, School of Mathematics and Physics, Queen's University Belfast, 1 University Road, Belfast BT7 1NN, UK

Published online 7 October 2022

© The Author(s), under exclusive licence to EDP Sciences, SIF and Springer-Verlag GmbH Germany, part of Springer Nature 2022

## 1 Introduction

The online workshop ‘Quantum Battles in Attoscience 2020’ explored areas of tension related to matter in intense laser fields, and it was this unusual conference [1], which gave rise to this topical issue. Strong-field and attosecond science deal with systems under extreme conditions: external fields whose intensity rivals that of the binding forces involved—typically of the order of  $10^{13}$  W/cm<sup>2</sup> or higher—and some of the shortest time scales in nature—on the order of attoseconds ( $10^{-18}$  s). Because of this, standard theoretical approaches break down and novel methods and ways of thinking are required. This novelty, together with the extreme conditions, has triggered unprecedented progress, and the dream of steering electron motion in real time is becoming a reality.

However, this novelty has not come without controversy. Over the past three decades, our community has witnessed a great deal of debate over a wide range of topics and unfortunately, although vibrant, this debate has not always been constructive. On a number of issues (see e.g., [2–4]), the attoscience community has tended toward siloed factions holding opposing views. In organizing Quantum Battles in Attoscience we pictured these groups as the scientific equivalent of street gangs, and our proposed solution was to organize a martial-arts-style tournament, with a rigorous code of conduct and where respect would be maintained.

The Quantum Battles in Attoscience workshop took place on July 1–3 2020, and, although it was originally planned to happen at University College London was moved online due to the COVID-19 pandemic. It brought together leaders in the field, who gave invited talks, and highly promising early-to-mid-career researchers from groups holding opposing views, who co-organized and participated in three panel discussions, labeled the ‘quantum battles.’ In order to make the workshop accessible to all across the globe, we charged no fees and made pioneering use of online

resources such as social media and a dedicated YouTube channel to engage with the participants and general public.

The battles were a huge success, gathering over 300 registered participants from across the globe, and breaking several paradigms [1]. Quantum aspects of attoscience played a huge role in the debates, and the present topical issue records and extends this discussion. It highlights the three battles as perspective articles: ‘Tunneling,’ ‘Quantum Interference and Imaging,’ and ‘Analytical versus Numerical Methods,’ and brings other contributions from the attoscience and quantum technologies communities around those topics. We also had a few contributions from speakers in the ‘Atto Fridays’ seminar series, which started as a spin-off of the workshop in 2021 and meanwhile has established itself as one of the key seminar series in the field.

## 2 Three battles

A common thread in the three articles highlighted below is the discussion of different, sometimes conflicting viewpoints around a topic, aside from providing an idea of the overall landscape. They are also unusual in that they include the most important questions posed during the workshop, as well as results from opinion polls addressing the participants and the audience. All were written with great care by the battles’ participants and we hope that they will be a useful resource in the years to come.

*Tunneling* Apart from having puzzled scientists since the advent of quantum mechanics, tunneling plays a vital role in attosecond physics. For low-frequency, high-intensity fields, strong-field phenomena result from a multi-step process, in which the first step is often tunneling. For instance, high-order harmonics, the seed for many modern laser systems, may be generated by a process in which an electron tunnel ionizes, propagates in the continuum and is driven back to recollide and recombine with the ionic core. Upon recombination, the

<sup>a</sup> e-mail: [c.faria@ucl.ac.uk](mailto:c.faria@ucl.ac.uk) (corresponding author)



**Fig. 1** Conference picture with a small sample of the participants in the virtual workshop Quantum Battles in Atto-science 2020. It was impossible to bring together over 300 people from 34 countries at the same time. For details see [www.quantumbattles.com](http://www.quantumbattles.com)

kinetic energy acquired in the continuum is released as an XUV photon. The recollision process can also lead to high-order above threshold ionization (ATI) or laser-induced nonsequential double and multiple ionization (NSDI, NSMI), with the difference that the photoelectron rescatters elastically or inelastically off its parent ion, respectively. The tunneling debate has been most explicit in the context of attosecond angular streaking (attoclock) experiments which are often claimed to measure the so-called tunneling time. *The underlying question is whether tunneling takes time, and if so, how much?*

Cornelia Hofmann, Alexander Bray, Werner Koch, Hongcheng Ni and Nikolay Shvetsov-Shilovski address this question in [2], focusing on the physical observables and typical tunneling experiments, the nature of tunneling and the theoretical approaches to model tunneling (and possibly describe quantum tunneling times). Each of the topical introductions is followed by a debate, mirroring what occurred during the workshop. The question of observables is crucial due to several obstacles to timing the tunneling process, for instance the difficulty of defining the ‘beginning’ and ‘end’ of the tunneling process based on a spreading wavepacket. Several limitations to the attoclock experiment are also neatly outlined. The second topic discusses the nature of tunneling, and looks at the classical/quantum intersection, and if, in that context, one may define the ‘beginning’ or ‘end’ of tunneling. This is an important question, which many groups have tried to address from the classical perspective, or using trajectory-based methods. This brings the question of what quantities best characterize the onset of tunneling, the role and meaning of quantum-particle descriptions, and quantum-classical correspondence. Finally, they provide a summarizing account of the theoretical approaches employed to investigate tunneling, and list their key features, advantages and drawbacks. This includes quantum, semiclassical and hybrid methods. Although the main conclusion is that there is lack of

common ground in the understanding of tunneling and still much work to be done, the authors’ contribution has elucidated the key features of the debate and provided a foundation for this important, future work.

*Quantum Interference and Imaging* Another intriguing phenomenon since the early days of quantum mechanics is the quantum interference of matter waves. In attoscience, the external laser field is employed as a tool to create and control coherent electronic wave packets, which are then used to image targets in the sub-femtosecond and ångström regime. Imaging is made possible due to the electrons’ interaction with the parent ion, which happens as laser-induced rescattering and may take place along different quantum pathways. The review, written by Kasra Amini, Alexis Chacón, Benjamin Fetić and Matthias Kübel [3], focuses on photoelectron spectra and momentum distributions. Photoelectrons are excellent imaging tools due to their short de Broglie wavelengths and the associated high currents. However, throughout, a comparison is made with the other key imaging tool, namely high-order harmonic generation (HHG), and some interferometric schemes based on it. *Central questions are how to characterize a coherent electronic wave packet and use it to retrieve information about a target.*

Characterizing a wave packet requires retrieving the amplitudes and phase differences associated with its constituent partial waves. This invites the question of how to measure these quantities, which the authors address by pursuing unusual lines of inquiry. They start by asking why the measurements are performed in momentum space, and provide examples of experimental techniques to perform such measurements. Subsequently, the authors discuss different aspects of quantum interference, starting from the inter-cycle interference that leads to ATI rings, and going through interferometric schemes such as the Quantum Spectral Phase Interferometry for Direct Electron wave-packet Reconstruction (QSPIDER) and ultrafast photoelectron holography.

Furthermore, because rescattering itself modifies quantum interference, a whole section is dedicated to laser-induced electron diffraction (LIED), its variants and use for retrieving molecular structure. Extracting electron momentum distributions from *ab-initio* calculations is discussed, with emphasis on how to choose the correct scattering waves and boundary conditions. The last two sections deal with decoherence and whether it can potentially be avoided using quantum materials. Interestingly, HHG is coherent across the whole focus, while ATI is only coherent at the single-emitter level, i.e., one may determine which emitter has undergone ATI, but not HHG. Potentially, this may change for solids and lead to macroscopically coherent ATI. The article closes with a brief summary of solids in strong fields, and mentions several avenues along which imaging is expected to progress.

**Analytical versus numerical** Due to the extreme conditions involved, attoscience is the ideal testing ground for novel theoretical methods. Over the years, a wide range of analytical, numerical and hybrid approaches have been developed. This brings a great deal of tension and misconceptions, along with warring schools of thought which favor either type of approach. In this article, Gregory Armstrong, Margarita Khokhlova, Marie Labeye, Andrew Maxwell, Emilio Pisanty and Marco Ruberti try to dispel this tension and find common ground, by critically analyzing the key methods employed in attoscience [4]. They examine their advantages and shortcomings, their role in scientific discovery and try to find common ground by challenging established beliefs in the attoscience community. They also present two case studies, namely NSDI and resonances in HHG, as examples against which the propositions are tested.

The article brings several innovative features. First, it is written as a Socratic dialogue between two fictional theorists, Analycia Formuloff and Numerio Codeman, who favor analytical and numerical approaches, respectively, interwoven with more conventional, explanatory sections. Second, it includes the results of opinion polls associated with the workshop panel discussion, questions from the audience, a comprehensive summary of the existing theoretical methods in attoscience, and a compass going from analytical to numerical and from *ab-initio* to approximate to argue that these distinctions are more blurred than one may think.

To start the debate, the authors strive to define a rigorous framework for attoscience and the sources of confusion. Considerable effort is invested identifying tensions and highlighting misconceptions as precisely as possible. For instance, in our community the terms '*ab-initio*' and 'numerical' are often employed interchangeably, but, strictly speaking, such words carry different meanings. This is particularly true when electron-electron correlation is involved. Further to that, a hierarchy of numerical methods based on computational effort is established, which includes the single-active electron approximation to Hartree–Fock and post-Hartree–Fock methods, time-dependent density functional theory and full configuration interaction. Approaching the problem from the other side, the

authors question what 'analytic' really means, and if the word even makes sense when special functions and integral representations are used. A comprehensive analytical toolset is listed, which goes from quantum mechanical to semi-classical to purely classical, and hybrid methods, which incorporate analytical and numerical elements, are discussed. Thereby, several issues are raised, such as quantitative versus qualitative insights, and why *ab-initio* does not necessarily mean exact. They also touch upon the issue of modularity and predictive power, range of applicability, model-building and how to successfully challenge and benchmark the models, either using experiments or numerical methods. There are also several discussions, focusing on questions such as: *Is approximation a strength or a weakness? Are both analytical and numerical methods required in scientific discovery? Is attoscience really after discoveries or is it not more about finding and solving interesting puzzles?* This is a must-read article for anyone wanting to understand the landscape of theoretical methods used in attoscience.

### 3 How quantum is atto?

Apart from the three highlighted articles, there are several contributions from leading scientists in the field. They touch upon quantum aspects of attoscience, and raise important questions. In [5] Reiss calls into question the tunneling model widespread in our community, and explains why this can only be used as an approximation for a limited range of parameters. This stance is more radical than that of the first battle, and the very title, 'no light at the end of the tunnel,' highlights that a transverse field, such as light, does not produce a quantum tunnel. In [6], Chomet and Faria provide an overview of phase-space methods in a wide range of strong-field phenomena. These are powerful tools widely used in myriad areas, such as quantum optics, quantum information, physical chemistry and semiclassical theory, but underused in attoscience. Apart from a comprehensive list of the phenomena in which they are employed, the review article provides selected examples drawn from the authors' own work, on strong-field ionization and HHG.

Decoherence and entanglement have attracted a great deal of attention in the attoscience community in the past few years. Examples are the entanglement between a photoelectron and the ion, of two photoelectrons, or different electronic degrees of freedom. In [7], Busto and co-authors study the entanglement of radial and angular degrees of freedom of a photoelectron in two-photon ionization of helium by using high-resolution interferometric techniques in the vicinity of a Fano resonance. The studies show different degrees of coherence for specific sidebands, which are associated with different degrees of entanglement.

Quantum interference in the context of tailored fields and selection rules are also present in our Topical Issue, both for high-order harmonic and photoelectron emis-

sion. In [8] Habibović and co-authors study HHG in heteronuclear diatomic molecules in the presence of orthogonally polarized two-color fields of commensurate frequencies. They study what types of minima are present in the spectrum depending on the field-frequency ratio, and control harmonic emission and ellipticity by altering the relative intensities and offset phase between the two driving waves. In Kang et al. [9], conservation laws are derived for the orbital angular momenta of electrons undergoing strong-field ionization in linear and circularly polarized fields. They depend on the initial quantum magnetic quantum number of the initial bound states and on the number of cycles in a pulse. These studies provide a basic framework for understanding electron orbital angular momenta in this context.

Ultrafast processes in topological systems have triggered huge interest, not only due to their extremely rich dynamics, but also due to their potential use in quantum technologies. See, for instance, the discussion about macroscopic coherence in ATI brought up in this editorial and in the perspective article [3]. This issue also features articles on HHG in topological nanoribbons [10], and strong-field ionization [11] in nanoparticles. In [10], Jürß and Bauer investigate the influence of edge-states in topological nanoribbons on HHG. They show that the crossings and avoided crossings that occur in the energy of these states strongly affect the HHG properties. In [11], Rosenberger and co-authors focus on the near-field imaging of nanoparticles. By studying the time-of-flight spectra of ions emitted by silica particles, they assess the applicability of reaction nanoscopy to a wide range of field frequencies and polarizations, and aim to provide a benchmark for current theoretical models.

The main question unifying the Quantum Battles workshop, the Atto Fridays series and this topical issue is an open one: *How quantum is atto?* Classical methods are hugely popular in strong-field and attosecond science and the intuitive insights they provide into complex phenomena are invaluable. Nonetheless, the phenomena themselves seem to be inherently quantum and effects such as quantum interference, tunneling, and entanglement play a huge role. It is also possible that, due to the extremely short timescales, usual decoherence effects may not have time to develop. This implies that bringing attoscience and quantum technologies together may open completely new avenues in research. As the above articles demonstrate, considerable progress has been made, but we expect much more in the years to come, accompanied, of course, by a healthy dose of controversy. The fight continues.

**Data Availability Statement** This manuscript has no associated data or the data will not be deposited. [Authors' comment: The present article is an editorial, and so no data was generated in the process of writing it.]

## References

1. C.F. de Morisson Faria, A. Brown, Courting controversy online. *Phys. World* **33**(10), 19 (2020)
2. C. Hofmann, A. Bray, W. Koch, H. Ni, N.I. Shvetsov-Shilovski, Quantum battles in attoscience: tunneling. *Eur. Phys. J. D* **75**, 208 (2021)
3. K. Amini, A. Chacón, S. Eckart, B. Fetić, M. Kübel, Quantum interference and imaging using intense laser fields. *Eur. Phys. J. D* **75**, 275 (2021)
4. G.S.J. Armstrong, M.A. Khokhlova, M. Labeye et al., Dialogue on analytical and ab initio methods in attoscience. *Eur. Phys. J. D* **75**, 209 (2021)
5. H.R. Reiss, No light at the end of the tunnel. *Eur. Phys. J. D* **75**, 158 (2021)
6. H. Chomet, C.F. de Morisson Faria, Attoscience in phase space. *Eur. Phys. J. D* **75**, 201 (2021)
7. D. Busto, H. Laurell, D. Finkelstein-Shapiro et al., Probing electronic decoherence with high-resolution attosecond photoelectron interferometry. *Eur. Phys. J. D* **76**, 112 (2022)
8. D. Habibović, W. Becker, D.B. Milošević, High-order harmonic generation by aligned heteronuclear diatomic molecules in an orthogonally polarized two-color laser field. *Eur. Phys. J. D* **75**, 122 (2021)
9. Y. Kang, E. Pisanty, M. Ciappina, M. Lewenstein, C.F. de Morisson Faria, A.S. Maxwell, Conservation laws for electron vortices in strong-field ionisation. *Eur. Phys. J. D* **75**, 199 (2021)
10. C. Jürß, D. Bauer, Edge-state influence on high-order harmonic generation in topological nanoribbons. *Eur. Phys. J. D* **75**, 190 (2021)
11. P. Rosenberger, R. Dagar, W. Zhang, A. Sousa-Castillo, M. Neuhaus, E. Cortes, S.A. Maier, C. Costa-Vera, M.F. Kling, B. Bergues, Imaging elliptically polarized infrared near-fields on nanoparticles by strong-field dissociation of functional surface groups. *Eur. Phys. J. D* **76**, 109 (2022)