Quantum bridges in phase space – Interference and Non-Classicality in Enhanced Ionisation

Heloise Chomet[1], Dhruva Sarkar[1], and Carla Figueira de Morisson Faria[1]

[1] *University College London*

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We perform a phase-space analysis of strong-field enhanced ionisation in molecules, with emphasis on quantum-interference effects. Using Wigner quasi-probability distributions and the quantum Liouville equation, we show that the primary cause of momentum gates is an interference-induced bridging mechanism that occurs if both wells in the molecule are populated. In the phase-space regions for which quantum bridges occur, the evolution of the Wigner function is essentially non-classical and non-adiabatic. Optimal conditions require minimising population trapping and using the bridging mechanism to feed into ionisation pathways along the field gradient.

Keywords: Wigner function, quantum optics, interference, non-classicality

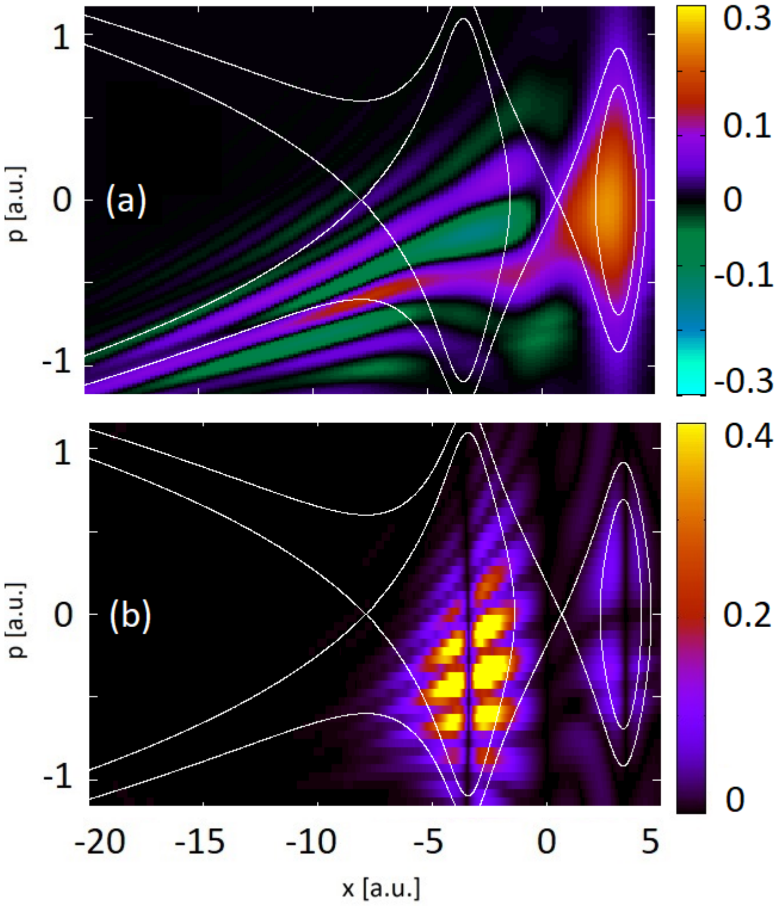

Figure.1: Comparison of (a) the Wigner quasi-probability distribution and (b) the quantum corrections Q(x, p, t), calculated for a model H_2^+ molecule of inter-nuclear separation R = 6.8 a.u. at time t = 24 a.u. in a static laser field of strength E = 0.0534 a.u. (intensity I = 1014 W cm^{-2}) using a Gaussian initial wave packet centred around the upfield potential well. The thin white lines in the figure give the equienergy curves (including the separatrices).

In this work, we perform a detailed analysis of strong-field enhanced ionisation using reduced-dimensionality models of diatomic molecules and phase-space methods.

Our studies show that enhanced ionisation stems from the interplay of at least two qualitatively different ionisation pathways, shown in Figure 1, with an optimal phase-space configuration chosen to minimise population trapping and maximise direct downfield population transfer.

One of these pathways follows the field gradient and leads to tails along separatrices that 'spill' into the continuum, while the other does not obey field gradients or classical barriers in phase space. The former pathway may be associated with quasi-static tunnelling mechanisms [1, 2] as well as the semiclassical limit of Wigner quasi-probability distributions [3], with oscillatory tails around separatrices and equienergy curves. The latter pathway has been first identified in [4, 5] for oscillating driving fields. It consists of a cyclic motion performed by the Wigner function in phase space and the emergence of momentum gates, along which there is a direct quasiprobability flow from one well to the other. Therein, momentum gates were explained as resulting from strongly coupled states and the non-adiabatic response to the time-dependent field gradients.

We find, however, that this pathway occurs also for static fields, and even in the absence of driving fields altogether. On top of that, near quantum bridges the Wigner quasiprobability distribution exhibits non-classical evolution, which we assess using the quantum Liouville equation:

$$\left(\frac{\partial}{\partial t}+\frac{p}{M}\frac{\partial}{\partial x}-\frac{dV_{\text{eff}}}{dx}\frac{\partial}{\partial p}\right)W(x,p,t)=Q(x,p,t)$$

By employing different types of initial bound states for the electronic wave packet, we show that the primary cause of the momentum gates in [4,5] is quantum interference. We also shed light on the behaviour observed for time dependent fields. The frequency of the quantum bridge being higher than that of the laser field, the quasiprobability distribution will sometimes counter-intuitively flow in the direction opposed to the electric-field gradient.

The fact that enhanced ionisation is an optimisation problem suggests that these ionisation mechanisms be controlled by appropriate coherent superpositions of states, targets and driving fields. This opens up a wide range of possibilities for studying quantum effects in enhanced ionisation.

References

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