Double ionization with intense laser pulses: Insights from chaos theory

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One of the most striking surprises of recent years in laser-matter interactions has come from multiple ionization by intense short laser pulses. Multiple ionization of atoms and molecules is usually treated as a rapid sequence of uncorrelated events. However, in the early 90's, experiments using intense laser pulses found *correlated* double ionization yields could exceed uncorrelated ones by several orders of magnitude. The precise mechanism that makes electron-electron correlation so effective follows the "three-step" scenario: An ionized electron, after picking up energy from the field, is hurled back at the ion core upon reversal of the field and dislodges the second electron.

Remarkably, it turns out that entirely classical interactions are adequate to generate the strong electron-electron correlation needed for correlated double ionization in this "deep quantum regime". This is in stark contrast with the traditional application of chaos theory to atoms in the semiclassical regime [1], where the electrons are almost classical. In this presentation, I will revisit the recollision mechanism using a nonlinear dynamics perspective. I will show that the three-step scenario has to be complemented by the dynamical picture of the inner electron [2, 3], which, far from acting as a spectator, plays an active role in the double ionization. Using this global picture of the dynamics, we are able to derive verifiable predictions on the characteristic features of the "nonsequential" process [2, 3, 4].

Analysis of the dynamics leads to reduced-dimensionality models for each electron between recollisions. A pivotal argument for this construction is the identification of an inner and an outer electron. This identification follows from a reduced number of hyperbolic periodic orbits which organize the motion [2, 3]: even though the dynamics is chaotic over the entire phase space, a typical trajectory follows one of these hyperbolic orbits (see Fig. 1) for a short time before jumping to another. For a two dimensional model enables one to identify a reduced number of candidate hyperbolic periodic orbits to organize the dynamics [5].

As an ultimate test for these reduced models, we built up a discrete time mapping [4], as a modified kicked rotator, which models the dynamics of the inner electron subjected to successive recollisions. This model captures the bare essentials of the nonsequential process and enables to reproduce both the nonsequential and sequential double ionization probability as a function of the intensity of the laser.

The traditional picture of the nonsequential double ionization is based on a little delay, if any, between the recollision and the emission of the second electron. However experimens have identified an alternative path for correlated double ionization: Recollision Excitation with Subsequent Ionization (or RESI). RESI arises from the inner electron being left in an excited state after the recollision and taking more time to ionize. We find that RESI events are a signature of chaos in nonsequential double ionization [6]. They result from the entanglement of stable and unstable manifolds of resonant hyperbolic periodic orbits. This entanglement generates a sticky region where the inner electron can be stuck for some time before ionizing. I will show how these structures naturally stand out and I will explain the mechanisms which regulate the RESI dynamics.

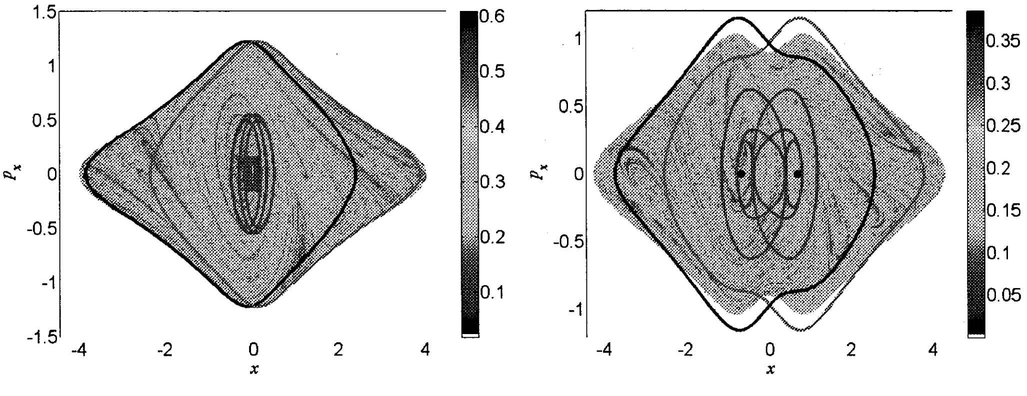

Figure 1: Finite time Lyapunov map for helium [2] (left panel) and the hydrogen molecule H_2 [3] (right panel). Initial positions are chosen on the surface (x, p_x), $y = 0$ and $p_y(\geq 0)$ adapted to be on the ground state energy surface. We also display projections in the plane (x, p_x) of four organizing periodic orbits for the model.

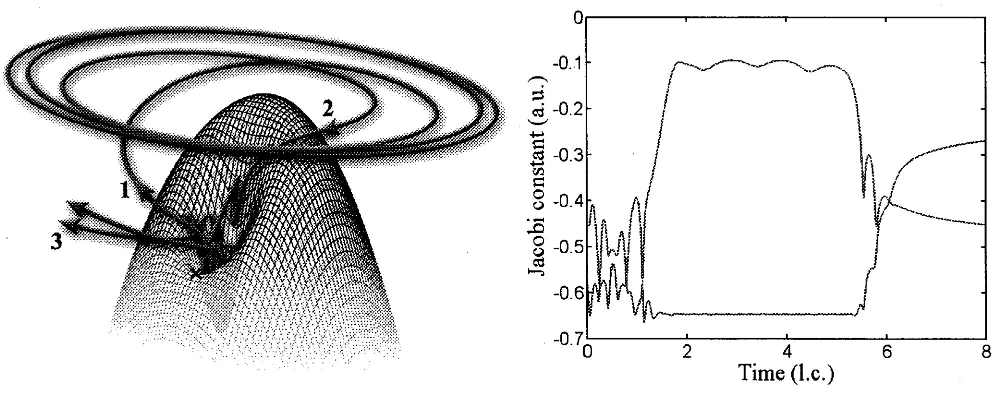

Figure 2: Typical nonsequential double ionization with circular polarization. Left panel: Dynamics of the two electrons in configuration versus Jacobi constant space. Right panel: Jacobi constant of each electron as functions of time. Both panels refer to the same trajectory.

The recollision scenario which works so well in linearly polarized (LP) fields is much more difficult to justify in fields in which the ionized electrons tend to spiral out from the core and to miss it like circularly (CP) or elliptically polarized fields [7]. In some experiments using CP fields, the double ionization yields follow the sequential mechanism, confirming current thinking, whereas in others these yields are clearly several orders of magnitude higher than expected, in apparent contradiction with it. The question we have recently resolved is [8]: Are recollisions possible in pure CP fields or does one have to rely on a small residual ellipticity? We explain these seemingly contradictory findings using a classical Hamiltonian model. In addition, we show that, contrary to common belief, recollision (see Fig. 2) can be the dominant mechanism leading to enhanced double ionization yields in CP fields.

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