Decay rates of planar helium

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Abstract. – We compare the autoionization rates of characteristic configurations of the doubly excited helium atom, in one, two and three dimensions. We find that the decay rates of the two dimensional restriction are in quantitative agreement with those of the real physical system, whilst the one dimensional model can underestimate the actual decay rates by orders of magnitude.

The quantum mechanical treatment of the helium atom goes back to the early days of quantum mechanics: Einstein was the first [1] to realize that the then available quantization schemes which had been applied successfully in the analysis of atomic spectra of one electron atoms would be inoperative for this microscopic realization of the gravitational three body problem. As first noticed by Poincaré, the classical dynamics of the latter is nonintegrable, and this remains true when gravitational forces are substituted by attractive and repulsive Coulomb forces, such as to define the three body Coulomb problem. Indeed, the electron-electron interaction term in the Hamiltonian of the unperturbed helium atom — which otherwise is just the sum of two hydrogen Hamiltonians with amended nuclear charge — makes the doubly excited states to decay via autoionization [2] and renders the two-electron dynamics in general irregular or chaotic, with only rather small domains of the classical phase space occupied by regular, i.e., integrable, motion. On the quantum level, the loss of integrability is tantamount to the (at least partial) destruction of good quantum numbers, and leads to an abundance of intriguing and surprising effects, such as Ericson fluctuations in the photo-cross-section at high excitation energies [3], with nevertheless very stable configurations such as the highly asymmetric frozen planet configuration of the doubly excited atom [4,5]. Hence, even without any external perturbation, doubly excited states of helium represent one of the most challenging — and experimentally accessible [6] — test cases for the theory of quantum chaos [7], which deals with low dimensional, complex though completely deterministic (in the sense of the absence of any random forcing) quantum dynamics.
In the presence of an external perturbation such as an electromagnetic field [8,9], however, the complexity of the electronic dynamics and of the underlying spectrum increases dramatically, due to the perturbation-induced coupling of different total angular momentum states. This rendered an exact quantum treatment of strongly perturbed doubly excited states of helium so far prohibitive, and accurate quantum treatments [10–13] are only available for excitations from the atomic ground state or from weakly excited states by optical fields, together with some one or two dimensional, strongly simplified models of the helium atom [14,15], which possibly allow for some qualitative insight.

It is the purpose of the present Letter to indicate an alternative pathway towards the realistic modeling of doubly excited states of helium, through the comparison of the autoionization rates of characteristic two-electron configurations of the unperturbed atom with zero total angular momentum, \( \bar{L} = 0 \), in a one, two, and three dimensional configuration space (with no further restrictions). We shall see that the dependence of the autoionization rates on the dimension of the accessible configuration space sensitively reflects the dominant decay process of the three dimensional compound, and that the planar helium atom apparently accounts for all relevant decay channels, as it exhibits decay rates quantitatively comparable to those of the real, three dimensional atom. Since, on the other hand, the Hilbert space of the two dimensional model has significantly smaller dimension than that of the real atom, and a fortiori so in the presence of an external perturbation, the planar helium atom seems to offer a sensible compromise between the mere complexity of the full problem, and the quest for a theoretical treatment with the potential of predictive power.

Let us start with the helium Hamiltonian in the center of mass frame, in atomic units, assuming an infinite mass of the nucleus,

\[
H = \frac{\mathbf{p}_1^2}{2} + \frac{\mathbf{p}_2^2}{2} - \frac{2}{|\mathbf{r}_1|} - \frac{2}{|\mathbf{r}_2|} + \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|},
\]

(1)

with \( \mathbf{r}_i, \mathbf{p}_i, i = 1, 2 \), the position and momentum in one, two or three dimensional configuration space. Combining group theoretical considerations, complex dilation of the Hamiltonian, and advanced techniques of high performance computing, the energy eigenvalues \( E_k \) together with the associated autoionization rates \( \Gamma_k \) and eigenvectors of this operator can be obtained as complex eigenvalues \( E_k - i\Gamma_k/2 \) of a sparse banded complex symmetric matrix. Whether in the one [16, 17], two [18, 19], or three dimensional [20, 21] case, our approach treats the Coulomb singularities of (1) exactly, as well as the coupling to the atomic continuum, which is induced by the electron-electron interaction term in the Hamiltonian. Hence, the results to be presented hereafter represent precisely the same physical system, with the exclusive distinction lying in the different dimensions of the configuration space accessible to the electrons.

Several approximate descriptions of the classical and quantum dynamics of the 3D helium atoms have been proposed. The most standard ones are the hyperspherical adiabatic approximation [22], Herrick’s algebraic approach [23], and the molecular adiabatic [24] approximation. These approaches make it possible to define some approximate good quantum numbers or classical constants of motion. The relative merits of these various descriptions depend on the states to be described; in several limiting cases, the various approaches are in fact equivalent, see [5] for details. It is important to realize that these approaches can be easily extended to the 2D model of the helium atom, simply by setting one quantum number to 0. In the molecular and hyperspherical adiabatic approaches, the 2D dynamics is obtained by putting the angular momentum \( m \) around the interelectronic axis to 0; similarly, it corresponds to the value \( T = 0 \) for the \((n, N, K, T)\) algebraic classification.

The two arguably most important families of eigenstates of the helium Hamiltonian, which exist in one, two, and three dimensions, represent collinear \( eZe \) and \( Zee \) electronic configu-
rations – with the electrons \(e\) on opposite sides \((eZe)\) resp. on the same side \((Zee)\) of the nucleus. The \(eZe\) configuration corresponds to the maximum value \(K = N - 1\) in the algebraic classification (and, similarly, to \(n_y = 0\) in the molecular adiabatic approach) while \(Zee\) corresponds to the minimum value \(K = 1 - N\), and to \(n_\lambda = 0\), respectively. Both these families are associated with periodic orbits of the classical dynamics of the three body Coulomb problem, and their eigenenergies can be estimated by semiclassical quantization of the classical dynamics \([5]\). However, the dynamical properties of both families are fundamentally different: Whilst the manifold of collinear \(eZe\) configurations is classically unstable, there is a large phase space area supporting regular \(Zee\) motion. Consequently, contributions of a large number of \((unstable)\) periodic orbits have to be summed over in a semiclassical treatment of the \(eZe\) manifold, whilst one single \((stable)\) periodic orbit suffices to estimate the energy levels associated with the \(Zee\) manifold. In the latter case, such semiclassical quantization establishes an immediate link between the fundamental classical frozen planet orbit \([4]\) and series of eigenstates of the quantum problem, which exhibit strong localization of the electronic density along the classical orbit. In the former case, various types of unstable periodic orbits contribute, where the antisymmetric stretch orbit \((1)\) – with the electrons following collinear straight line trajectories with opposite phase, colliding with the nucleus – is the least unstable one and thus enters the abovementioned sum with largest weight \([25]\). Indeed, eigenfunctions of the \(eZe\) manifold therefore tend to be scarred by the antisymmetric stretch orbit at moderate electronic excitations, i.e. they display enhanced electronic density along its trajectory \([5, 7]\). An important point is that both the frozen planet and the collinear \(eZe\) dynamics are fully one dimensional, i.e. are present in the one, two and three dimensional phase spaces. Comparison of the decay rates of the corresponding quantum eigenstates is therefore meaningful.

The collinear antisymmetric stretch orbit (and more generally the \(eZe\) classical dynamics) is stable with respect to transverse bending, though \(unstable\) with respect to perturbations in the axial directions. Hence, already the one dimensional version of \((1)\) accounts for the dominant \((classical)\) decay channel of the \(eZe\) configuration. In the molecular or hyperspherical adiabatic approach, this classical instability manifests itself by large couplings between different adiabatic potential curves, leading to significant avoided crossings in the spectrum: eventually, this will induce rather fast decay of the doubly excited states via autoionization. In contrast, the collinear frozen planet orbit is stable in the transverse as well as in the axial direction, the adiabatic potential curves are only weakly coupled and autoionization of the associated eigenstates of the quantum problem can only occur via \((classically\ forbidden)\) tunneling decay. Whether transverse tunneling is more or less efficient than tunneling along the symmetry axis of the configuration is hard to assess in such a high dimensional classical phase space. However, earlier quantum results on the autoionization rates of 1D and 3D frozen planet states were suggestive of the transverse degree of freedom contributing the dominant tunneling decay \([16, 20, 26]\).

Both families of states are identified in our numerically generated spectra through (a) their energies – which can be rather accurately predicted by semiclassical methods \([5, 25]\) –, (b) the expectation value of the angle between the electrons with respect to the nucleus, in the 2D and 3D problem, and (c) the localization properties of the electronic density in configuration space. In particular, for all states presented here, the wavefunctions are well localized along the respective periodic orbits, as illustrated in Fig. 1.

Figure 2 compares the life times of the collinear antisymmetric stretch eigenstates in one, two, and three dimensions, for principal quantum numbers \(N = 2\ldots 8\), and for singlet as

\[\text{(1)}\text{which is usually named “asymmetric stretch orbit” in the literature, despite its inherent (anti-) symmetry.}\]
Fig. 1 – Contour plot (on a linear scale) of the electronic density of the (triplet) planar frozen planet state (left) and of a (triplet) eigenstate strongly scarred by the antisymmetric stretch orbit (right), in 2D configuration space (spanned by the electrons’ distances \( r_1 \) and \( r_2 \) from the nucleus, in the collinear configurations considered here). Both eigenstates belong to the \( N = 9 \) series. The solid lines depict the associated classical periodic orbits.

Fig. 2 – Autoionization rates of antisymmetric stretch singlet (left) and triplet states (right) of the \( N^{th} \) autoionizing series of the helium spectrum, in 1D (squares), 2D (circles), and 3D (diamonds) configuration space.

well as for triplet symmetry. Apart from unsystematic fluctuations – which we attribute to local avoided crossings with other states, a generic phenomenon in the doubly excited energy range and more generally in any chaotic quantum system – the autoionization rates are of the same order of magnitude in all dimensions, with the 1D states almost always slightly more long lived than those in 2D and 3D. This is consistent with our intuition already alluded to above: since the classical antisymmetric stretch configuration decays along the axial degree of freedom (through coupling between the various adiabatic potential curves), already the 1D model can grasp the dominant fragmentation process, and contributions to the autoionization
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Fig. 3 – Autoionization rates of frozen planet states of helium in 1D (open circles; the exchange energy vanishes in 1D configuration space, since the electrons are effectively distinguishable [26]), 2D (filled squares for singlet, filled diamonds for triplet symmetry), and 3D (open squares for singlet, open diamonds for triplet) configuration space. The much higher rates in two and three dimensions provide a clear proof that autoionization in the longitudinal direction is a slow process, largely dominated by autoionization in the transverse direction, which probes the chaotic domain of phase space.

rates due to transverse tunneling in the 2D and 3D case are but a small correction.

The situation is dramatically different for the classically stable frozen planet configuration, with its 1D, 2D, and 3D decay rates compared in Fig. 3: Here, the 1D decay rates are suppressed by approx. six to seven orders of magnitude as compared to the 2D and 3D rates, the latter being essentially identical, except for unsystematic local differences, once again induced by tiny local avoided crossings in the energy spectrum [19, 26, 27]. Furthermore, the 1D rates vary smoothly with \( N \) and exhibit a clear exponential decay – in agreement with ordinary tunneling in a regular (non-chaotic) 1D system [16,26]. In contrast, the 2D and 3D rates display apparently erratic fluctuations.

This observation carries at least two messages: (i) The 1D frozen planet states are anchored to regular islands in the phase space spanned by the axial degree of freedom [4], what actually manifests in the exponential decrease of the (tunneling) 1D rates with the principal quantum number \( N \) of the inner electron, above \( N = 4 \) [16,19]. Such regular islands express a dynamical stabilization mechanism [28], which here prevents the electrons very efficiently from direct encounter on the axis. Since the Coulomb singularity at the nucleus furthermore establishes an impenetrable potential barrier in the axial direction, there can be no tunneling coupling of 1D frozen planet states to (much less stable) eigenstates of the \( eZe \) manifold, in the axial degree of freedom. The latter can only be mediated by the transverse degree of freedom [29], hence the enhanced decay in 2D and 3D illustrated in Fig. 3. Thus, the dominant decay of (2D-3D) collinear frozen planet states occurs in the transverse direction, whilst the tunneling coupling to the axial decay channel is much weaker. In other words, the angular correlation of the inner and outer electron’s motion is much more fragile than the dynamical stabilization of their relative radial motion. (ii) Since the 2D and 3D rates are essentially identical, for all values of \( N \), our present results of Figs. 2 and 3 also indicate that planar confinement of
the three body Coulomb problem does not imply any severe restrictions for the quantitative description of the quantum fragmentation process. This is plausible, since vanishing total angular momentum $\hat{L} = 0$ strictly confines the classical dynamics of the three-body problem to a fixed plane. Thus, as long as only zero angular momentum configurations are considered, the classical dynamics is the same in two and three dimensions. As a first guess, it is thus not unexpected that the two and three dimensional systems decay similarly by autoionization. Note, however, that the topology of phase space differs in the two cases (a single plane in two dimensions, an infinite manifold in three dimensions), and this could strongly affect transport properties and decay rates, especially when classically forbidden processes such as quantum tunneling come into play. The situation is of course completely different in the one dimensional system, where the classical phase space is only a very restricted subspace of the full two dimensional dynamics. No simple relation is therefore to be expected between the one and three dimensional decay rates.

The present results have been obtained for states belonging to the $\hat{L} = 0$ subspace. For nonvanishing total angular momentum, the classical 3D motion is no longer planar and deviations between 2D and 3D are expected. While the full quantum calculation is not more difficult in 2D for non-zero angular momentum, the complexity increases very rapidly for a 3D quantum calculation (precisely because of the deviation from coplanarity), rendering a repetition of the present calculation practically impossible. However, it is likely that, as soon as the angular quantum number $L$ is smaller than both electrons’ principal quantum number, the planar approximation is a good one, making the 2D model very valuable. Asymptotically, the dynamics of fragmentation is planar, even for non-zero angular momentum.

In the presence, e.g., of an external electromagnetic field, angular momentum is however not conserved. Notwithstanding, even if an external perturbation mixes the angular degrees of freedom of the two-electron dynamics, the angular excitation will in general remain relatively small compared to the radial excitation of the electrons. In such cases, the 2D model should be extremely useful, since the solution of the 2D eigenvalue problem demands less computational effort than in 3D. Whilst this is not crucial for the modeling of the unperturbed system, it is of decisive importance in the presence of a strong external perturbation, what then poses a truly challenging problem in computational atomic physics. Specifically, 2D quantum calculations might provide us with valuable insight on nonsequential double ionization. Indeed, classical arguments [30] suggest that the dominant mechanism for this particular process involves a situation where the nucleus and the two electrons lie on an equilateral triangle in a plane containing the electric field vector. This scenario is truly two dimensional and does not arise within the 1D model of the atom. Therefore, a comparison of accurate quantum calculations in 1D and 2D will elucidate to which extent the mechanism proposed by Ref. [30] contributes to nonsequential double ionization. If this mechanism is dominant, we expect double ionization rates resulting from the 2D quantum calculation to be quantitatively comparable to the “true” double ionization rates of the 3D helium atom.

In conclusion, we have seen that the dimension of configuration space does matter for the quantitative description of the autoionization process of collinear, doubly excited Rydberg states of helium, and that one dimensional models, even with full account of the various Coulomb singularities and atomic continua, can lead to dramatically false predictions for the decay rates. On the other hand, we have also seen that a planar model allows for quantitatively reliable predictions, with promising prospects for future studies of helium under electromagnetic driving.

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