Ericson fluctuations in an open, deterministic quantum system: theory meets experiment

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We provide numerically exact photoexcitation cross sections of rubidium Rydberg states in crossed, static electric and magnetic fields, in quantitative agreement with recent experimental results. Their spectral backbone underpins a clear transition towards the Ericson regime.

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The experimental characterization and theoretical understanding of complex quantum transport phenomena is of fundamental relevance for many, otherwise rather remote research areas which exploit quantum interference effects for the purpose of an ever improving control over the quantum dynamics of increasingly complicated systems \cite{1, 2, 3, 4, 5, 6, 7, 8}. In such context, “complexity” may arise from many particle interactions, from deterministic chaos, or from disorder, to name just a few of its possible causes. Notwithstanding the many origins of complex quantum dynamics, its qualitative macroscopic signatures are often very similar: an enhanced sensitivity on small changes in some control parameter (be it the boundary conditions in a disordered mesoscopic conductor \cite{3}, the perturbation strength in a strongly driven atomic system \cite{2}, the injection energy in a compound nucleus reaction \cite{4}, or the observation angle in a random laser \cite{11, 12}) enforces a statistical description such as to isolate robust quantities to characterize the underlying physical processes. Surprisingly, many of the resulting predictions are universal in character, i.e., they apply to, on a first glance, rather different classes of physical objects, which only share an increased density of states, and the nonperturbative coupling of their various degrees of freedom.

While erratic fluctuations of some experimental observable under changes of a control parameter come not too surprising in many-particle dynamics or in disordered systems \cite{1, 2, 3, 4, 5, 6, 7, 8}, they still remain rather counterintuitive and for many a cause of discomfort in simple quantum systems with only few degrees of freedom – think of single electron or photon transport across two dimensional billiards \cite{14, 15, 16}, or of the ionization probability of a one electron Rydberg state under external forcing \cite{2, 17}. Here, classically chaotic dynamics substitute for disorder and many-particle interactions, though are expected to generate very similar – if not the same – statistical behaviour, in tantalizing contrast, e.g., to the clock-like regularity of Kepler like Rydberg motion. Hitherto, however, experimental evidence for chaos-induced fluctuations in the coherent quantum transport in low dimensional, strictly deterministic systems is scarce \cite{14}, since bona fide transport measurements require very high spectral resolution in the continuum part of the spectrum, together with the continuous tunability of a suitable control parameter.

In the present Letter, we focus on a paradigmatic example in the realm of atomic physics – the photoexcitation of one electron Rydberg states in the presence of crossed, static electric and magnetic fields. Our contribution is motivated by recent experimental results \cite{18, 19} which probe the atomic spectrum above the field induced ionization saddle, and refines the interpretation of the experiments as the first observation of Ericson fluctuations in a strictly deterministic, open quantum system. Furthermore, this represents the first full-fledged, parameter-free quantum treatment of the truly three dimensional crossed fields problem at experimentally realistic spectral densities.

Ericson fluctuations are a universal statistical feature of strongly coupled, fragmenting quantum systems, first predicted \cite{20} and observed \cite{21} in compound nuclear reactions. They manifest as fluctuations in the excitation cross sections into the regime of highly excited, metastable resonance states, with typical decay widths larger than the average level spacing, such that single maxima in the cross section cannot be identified with single resonances any more, but are rather due to the interference of several of them. In particular, this implies that the typical scale of fluctuations induced by interfering decay channels is smaller than the typical width of individual resonances. In quantum systems with a well-defined classical analog, Ericson fluctuations can be understood as a hallmark of chaotic scattering \cite{22}, somewhat complementary to the exponential or algebraic decay of the survival probability on the time axis \cite{23}. A similarly paradigmatic case of the Ericson scenario as the one to be considered presently arises in the photoexcitation of highly doubly excited states of helium, yet still awaits its full experimental confirmation, due to the extraordinary requirements on the experimental resolution \cite{24}.

Let us start with the Hamiltonian describing the single electron Rydberg dynamics subject to crossed electric and magnetic fields, in atomic units, and assuming an infinite mass of the nucleus:

\begin{equation}
H = \frac{p^2}{2} + V_{\text{atom}}(r) + \frac{1}{2} B I_z + \frac{1}{8} B^2 (x^2 + y^2) + F x . \quad (1)
\end{equation}
Here, \( B \) and \( F \) are the strength of magnetic and electric field, respectively, and \( \ell \), the angular momentum projection on the magnetic field axis. Note that no uniquely defined one particle potential is available for the bare atomic Hamiltonian with its alkali earth multielectron core, and \( V_{\text{atom}} \) is therefore not given explicitly. However, the deviation of \( V_{\text{atom}} \) from a strictly Coulombic potential in a small but finite volume around the nucleus can be accounted for by the phase shift experienced by the Rydberg electron upon scattering off the multielectron core \([24, 26]\). This phase shift is fixed by the \( \ell \)-dependent quantum defects \( \delta \ell \) of the unperturbed atom, which are precisely determined by spectroscopic data \([27]\). The continuum coupling induced by the electric field (which lowers the ionization threshold by creating a Stark saddle in the \( x \) direction) is incorporated into our theoretical treatment by complex dilation of the Hamiltonian, and the numerical diagonalization of the resulting complex symmetric matrix (represented in a real Sturmian basis) immediately yields the full spectral structure (eigenenergies \( E_j \), decay rates \( \Gamma_j \), and eigenvectors \( |E_j\rangle \)) underlying the experimentally measured photoionization cross section, without adjustable parameters \([24, 26]\). Note that, due to the mixing of all good quantum numbers of the unperturbed dynamics (the principal quantum number \( n \), the angular momentum quantum number \( L \), and the angular momentum projection \( M \)) by the crossed external fields, we are dealing with a truly three dimensional problem \([28, 29]\), and the only remaining constants of the motion are the energy \( E \) and parity with respect to the plane defined by the magnetic field axis. Consequently, the complex symmetric eigenvalue problem which we have to solve has a respectable size, with typical (sparse banded) matrix dimensions of \( 300000 \times 40000 \), for the energies and field strengths we shall be dealing with hereafter.

The experiments in \([18, 19]\) probe the energy range from \(-57.08 \text{ cm}^{-1} \) to \(-55.92 \text{ cm}^{-1} \) (corresponding to principal quantum numbers \( n \approx 43 \ldots 45 \) of the bare atom), at fixed electric field strength \( F = 22.4 \text{ kV/m} \), and for three different values of the magnetic field, \( B = 0.9974 \text{ T}, B = 1.49 \text{ T}, \) and \( B = 2.0045 \text{ T} \). The electric field shifts the effective ionization threshold to \(-91.4 \text{ cm}^{-1} \), hence the experimentally probed energy range lies clearly in the continuum part of the spectrum. Invoking the scale invariance of the classical dynamics in a Coulomb potential, the specific choice of \( F \) and \( B = 2.0045 \text{ T} \) is equivalent to the one in \([28]\) (there for the purely Coulombic problem, and for a lower lying energy range, \( n \approx 19 \ldots 22 \), i.e., at much reduced spectral densities), and corresponds to classically chaotic scattering (where electric and magnetic field are of comparable strength, though incompatible symmetry). While the finite size multielectron core of rubidium strictly speaking invalidates such a scaling argument (as well as it blurs a strict quantum-classical analogy, simply due to the absence of a well defined classical one particle analog) \([20]\), it may still serve as an approximate guide into the regime of broken symmetries of the quantum problem \([30]\).

We performed numerical diagonalizations of the complex dilated Hamiltonian \([11]\) precisely for the experimental parameter values, though in a broader energy range, such as to illustrate the emergence of Ericson fluctuations from a smooth continuum background, with increasing Rydberg energies. The photoexcitation cross section \( \sigma(E) \) is readily obtained from the quantum spectrum, via

\[
\sigma(E) = \frac{4\pi(E - E_0)}{ch} \text{Im} \sum_j \frac{D_{j,L=2}^2}{E_j - i\Gamma_j/2 - E},
\]

where \( D_{j,L=2} \) denotes the relative oscillator strength \([27]\) of the transition from the initial state \(|n = 5 \ L = 1 \ M = -1\rangle \) with energy \( E_0 \sim -0.002 \text{ a.u.} \) into the electronic eigenstate \(|E_j\rangle \) with decay rate \( \Gamma_j \), mediated by a single photon linearly polarized along the magnetic field axis (thus selecting the odd parity part of the spectrum). Note that our computational method does not allow for an absolute calibration of the oscillator strengths, since the wave function of \(|n = 5 \ L = 1 \ M = -1\rangle \) is not explicitly known. For the technical details underlying the expression \([14]\), we refer the reader to \([22, 31]\).

Figure \([\text{II}]\) shows the thus obtained photoexcitation spectra, at magnetic and electric field strengths \( B = 2.0045 \text{ T} \) and \( F = 22.4 \text{ kV/m} \), respectively, and in three differ-
local mean level spacing $\Delta = \langle E_j - E_{j-1} \rangle$, in units of the energy range which spans the domain probed in the three plots of Fig. 2 for the same values of $F$ and $B$. The dashed line at $\Gamma_j/\Delta = 1$ separates isolated ($\Gamma_j/\Delta < 1$) from overlapping resonances ($\Gamma_j/\Delta > 1$). Comparison with Fig. 1 shows how broad resonances on the spectral level induce a strongly fluctuating continuum background, in the photoexcitation cross section. The energy range $E = -60.0 \ldots -55.0$ cm$^{-1}$, where resonance overlap is most pronounced in this plot, covers the energy range probed in the experiments reported in [15, 16].

Different energy ranges, $-70.0 \ldots -65.0$ cm$^{-1}$, $-65.0 \ldots -60.0$ cm$^{-1}$, and $-60.0 \ldots -55.0$ cm$^{-1}$. The latter of these completely covers the experimentally probed energy interval. Clearly, individual resonances can be resolved in the two lower lying spectra, on top of an essentially flat continuum background. In contrast, the experimentally probed energy range is characterized by a strongly fluctuating continuum, with only few narrow structures on top, what immediately suggests the overlapping of an appreciable part of the resonances which contribute to the sum in eq. (4). Inspection of the underlying distribution of resonance widths $\Gamma_j$ along the energy axis, measured in units of the average local level spacing $\Delta = \langle E_j - E_{j-1} \rangle$, indeed comforts this picture, see Fig. 2. The weight of large resonance widths with $\Gamma_j > \Delta$ clearly increases as we probe higher lying energies, and amounts to approx. 65% of all contributing resonances, in the experimentally probed energy range. Many of the structures in $\sigma(E)$ are consequently due to the interference of decay channels through overlapping resonances.

A close comparison of our numerical cross section in the lower panel of Fig. 1 (and of Fig. 3 below) shows close similarity with the experimental signal [18], though no perfect coincidence is achieved. This, however, is anything but surprising, precisely due to the characteristic extreme sensitivity of quantum spectra and cross sections with respect to tiny changes in the boundary conditions, in the regime of classically chaotic dynamics [2, 10, 14, 17]. Therefore, rather than scanning parameter space on a fine mesh, to reproduce the experimentally observed (but fragile!) cross section exactly [22], we calculate the autocorrelation function $C(\Delta E) = \langle \sigma(E + \Delta E) - \langle \sigma \rangle \rangle / \langle \sigma \rangle$ of $\sigma(E)$, which is predicted [22] to have a Lorentzian shape with the characteristic width $\gamma$, $C(\Delta E) \sim 1/(\Delta E^2 + \gamma^2)$, in the regime of strongly overlapping resonances. The latter condition is indeed met by all the three values of the magnetic field employed in the experiments in [18, 19], and Fig. 3 shows the autocorrelation functions, together with the excitation spectra from which they are deduced. $\gamma$ is expected to be a statistically robust quantity, and we verified that its value remains unaffected by the sensitive parameter dependence of $\sigma(E)$ itself, within the error bars indicated. The respective values of the characteristic widths $\gamma$ are in perfect agreement with the experimental values, as listed in table I. In particular, also the nonmonotonous dependence of $\gamma$ on the magnetic field strength $B$ is recovered.

To complete the picture, we also display in Fig. 3 the
excitation spectrum and the associated cross section at a weak magnetic field $B = 0.563$ T (not recorded in [18]), where the classical dynamics of the associated Coulombic problem is near regular, since the electric field dominates the dynamics [28]. In contrast, the present result for rubidium exhibits a very similar structure as for stronger magnetic fields, certainly due to the destruction of the Coulomb symmetry by the multielectron core [33].

In conclusion, we revealed the spectral backbone of experimentally observed fluctuations in the photoexcitation probability of nonhydrogenic rubidium Rydberg states in crossed static electric and magnetic fields – a truly three dimensional, paradigmatic case of microscopic chaotic (half) scattering. By correlating the experimentally available data with the resonance spectrum of the atom in the field (obtained from an accurate numerical treatment without adjustable parameters), and with the evolution of the latter along the energy axis, we theoretically/numerically prove that these experiments indeed successfully entered the regime of Ericson fluctuations, for the first time in a perfectly deterministic, open quantum system.

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| $B$ [T] | $\gamma_{\text{exp}}$ [cm$^{-1}$] | $\gamma_{\text{th}}$ [cm$^{-1}$] |
|--------|-------------------------------|-------------------------------|
| 0.9974 | 0.0083                        | 0.0082 ± 0.0005               |
| 1.49   | 0.0065                        | 0.0062 ± 0.0005               |
| 2.0045 | 0.0081                        | 0.0080 ± 0.0005               |

TABLE I: Comparison of the characteristic correlation decay length $\gamma_{\text{th}}$ deduced from our numerical photoexcitation spectra with the experimental values $\gamma_{\text{exp}}$ reported in [18]. Within the indicated error bars, which are estimated from changes of $\gamma$ under small changes of $B$ and of the electric field strength $F$ (within their experimental uncertainties [18]), the agreement is perfect. In particular, also the nonmonotonous dependence of $\gamma$ on the magnetic field strength $B$ is recovered.