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Radiative decay of nondispersive wave packets

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Abstract. – We study the decay of nondispersive electronic wave packets in strongly driven hydrogen atoms, which are coupled to the atomic continuum and to the vacuum modes of the electromagnetic field. A novel implementation of complex scaling allows to derive a master equation that incorporates the interplay of the coherent and the incoherent decay process. Ample numerical data predict a crossover from ionization via chaos-assisted tunneling to radiative decay, at experimentally accessible Rydberg levels.

Introduction. – The closest quantum analogy to the classical concept of a point particle is provided by a wave packet [1]. This term usually refers to the coherent superposition of the stationary eigenstates of a time-independent Hamiltonian forming an initially localized wave function. The energy spacing of the stationary eigenstates is generically unharmonic, which leads to the apparently unavoidable *dispersion* of such wave packets [2].

However, it has been shown recently that in nonlinear time-periodic systems wave packets can exist that *do not disperse*, because they are periodic eigenstates of the time-dependent problem [3]-[8]. They faithfully trace the corresponding classical trajectory and maintain their localization in phase space, except for some transient modulation in the case of a nonuniform acceleration of the wave packet along its path. In a more general perspective, these nondispersive wave packets are the quantum analog of a classical nonlinear resonance in a periodically driven Hamiltonian system [3]-[5]. They can be considered as direct generalizations of (stationary) eigenstates associated with elliptic islands in the classical phase space of nonlinear, autonomous Hamiltonian systems [9].

An experimentally accessible system where nondispersive wave packets can be observed is a hydrogenic Rydberg atom subjected to a monochromatic microwave field [10], [11]. In a quantum calculation one finds [5], [7], [8] eigenstates of the atom dressed by the driving field, which are perfectly localized on the principal nonlinear resonance created by the resonant coupling of the microwave field to the fundamental harmonic of the atomic Kepler motion. As a consequence of the field-induced coherent coupling of Rydberg states to the atomic continuum, these dressed states acquire a *finite lifetime* [5], [7], [8]. Nonetheless, it has been shown that the width (the inverse of the lifetime) of the wave packet eigenstates is extremely small and decreases on the average with increasing n [7]. The ionization process can be viewed as dominated by tunneling through the classical invariant tori which shield the elliptic island from the surrounding "chaotic sea" [7]. The rates should *exponentially* tend to zero as the

semiclassical limit $\hbar \sim 1/n \to 0$, n the principal quantum number, is approached [9]. This dependence on n is distinct from classical leakage mechanisms which exhibit classical scaling laws (*i.e. power laws*) in n.

Due to their almost eternal lifetimes the nondispersive wave packets are very interesting objects in the diagnostics of complicated atomic and molecular dynamics [12], [13]. They even encouraged speculations about the observability of extremely slow classical processes like Arnold diffusion in real quantum systems [14]. However, the discussion in the literature neglected any *decoherence effects* so far, which originate from the unavoidable coupling to the environment ([1]). Though, the lifetimes predicted for the wave packet eigenstates are comparable to the radiative lifetimes of Rydberg states [7], [8]. Hence, it is still an open question to what extent they will be observable in a laboratory experiment.

In the present letter, we want to elucidate the role of spontaneous emission. For this purpose, we introduce a novel implementation of complex scaling [16] which allows for the simultaneous description of the exact coherent coupling between the atom and the dressing field, together with a perturbative description of the coupling to the electromagnetic vacuum. The resulting master equation provides a *general* framework for the incoherent coupling of *decaying* states to a Markovian environment and is not restricted to the particular application we choose.

Theory. – For the sake of simplicity, we restrict ourselves to Rydberg atoms confined to one spatial dimension, driven by a linearly polarized microwave field. If we set aside for a moment the coupling to the quantized electromagnetic field, the periodically driven Rydberg electron is described by the Hamiltonian

$$H_{\rm sys}(t) = \frac{p^2}{2} - \frac{1}{x} + F\,x\cos(\omega t) \qquad (x>0), \tag{1}$$

i.e. we treat the driving microwave as a classical field in dipole approximation, neglect relativistic effects, assume an infinite nuclear mass, and employ atomic units.

Complex scaling of (1), by the positive angle θ, together with the Floquet theorem, allows us to determine the exact quasi-energies E_α and widths Γ_α of the dressed states $|\chi_\alpha\rangle$ of the atom in the coherent field. They are given by the complex eigenvalues $E_\nu - i\Gamma_\nu/2$ of the dilated Floquet Hamiltonian $\mathcal{H}^\theta_{\rm sys}$, with discrete, $2\pi/\omega$-periodic and square-integrable eigenstates $|\chi_{\nu\theta}\rangle$ (with Fourier components $|\chi^k_{\nu\theta}\rangle$). $\mathcal{H}^\theta_{\rm sys}$ is obtained from the Floquet Hamiltonian $\mathcal{H}_{\rm sys} = H_{\rm sys}(t) - i\partial_t$ by a non-unitary, complex scaling transformation $R(\theta)$ [17]. The associate left eigenvectors $\overline{\langle\chi_{\nu\theta}|}$ are the complex conjugate of $\langle\chi_{\nu\theta}|$, *i.e.* the transpose of the right eigenvectors. In this basis, the time evolution operator generated by $H_{\rm sys}(t)$ reads, for $t>0$ [17],

$$U_{\rm sys}(t) = \sum_{\nu k_1 k_2} e^{-\Gamma_\nu t/2} e^{-i(E_\nu + k_2\omega)t}\, R(-\theta)|\chi^{k_2}_{\nu\theta}\rangle\overline{\langle\chi^{k_1}_{\nu\theta}|}R(\theta). \tag{2}$$

The adjoint $U^\dagger_{\rm sys}$ is given by the complex conjugate expression [17]. These are needed in our treatment of the coupling of the dressed states to a reservoir,

$$H_{\rm res} = \sum\nolimits_j \omega_j \left(b^\dagger_j b_j + \frac{1}{2}\right) \qquad \text{by} \qquad H_{\rm int} = \sum\nolimits_j x(g_j b_j + g^*_j\,, b^\dagger_j)\,, \tag{3}$$

with a symbolic sum over the continuum of reservoir modes j, the associated creation and annihilation operators $b^\dagger_j$ and b_j, and the coupling constants g_j.

([1]) In a different context, the role of spontaneous emission was studied for the dynamics of radial Rydberg wave packets synchronized with the Rabi oscillation of a strongly driven atomic core [15].

In the product basis spanned by the eigenstates of $\mathcal{H}^{\theta}_{\rm sys}$ and the reservoir Fock states, the evolution $U_0 := U_{\rm sys} \otimes \exp[-iH_{\rm res}t]$ is explicitly known. This suggests to proceed in the interaction picture, to formulate the temporal evolution of the density operator $\hat{\rho}(t)$ generated by the total Hamiltonian $H(t) = H_{\rm sys}(t) + H_{\rm res} + H_{\rm int}$.

At this point, care has to be taken due to the finite widths Γ_ν of the eigenstates of $\mathcal{H}^{\theta}_{\rm sys}$: as a consequence, any square-integrable initial atomic state will leak out to infinity on a finite time scale. Hence, its norm within any finite volume around the nucleus is *not* conserved [18], which explains that $U^{\dagger}_{\rm sys}(t)U_{\rm sys}(t) \neq id$ for $t > 0$ (the expressions for $U_{\rm sys}$ and $U^{\dagger}_{\rm sys}$ only operate on square-integrable states [16], [17]). Consequently, since we want to formulate time-dependent perturbation theory in the basis represented by the $|\chi_{\mu\theta}\rangle$, the density operator in the interaction picture has to be defined by explicitly employing the inverse operators of $U_{\rm sys}$ and $U^{\dagger}_{\rm sys}$, $U^{-1}_{\rm sys}$ and $[U^{\dagger}_{\rm sys}]^{-1}$, respectively. Those are obtained, for $t > 0$, through the identification $U^{-1}_{\rm sys}(t) := U_{\rm sys}(-t)$ and $[U^{\dagger}_{\rm sys}]^{-1}(t) := U^{\dagger}_{\rm sys}(-t)$. The appropriate definition,

$$\tilde{\rho} := U_0^{-1}\, \hat{\rho}\, [U_0^{\dagger}]^{-1}\,, \tag{4}$$

then leads to the equation of motion for the *coherent* evolution of the density operator

$$\frac{\rm d}{{\rm d}t}\tilde{\rho} = -i\,(U_0^{-1} H_{\rm int} U_0\, \tilde{\rho} - \tilde{\rho}\, U_0^{\dagger} H_{\rm int} [U_0^{\dagger}]^{-1}) \tag{5}$$

(which reduces to the usual equation *only* for vanishing Γ_ν).

We can now follow [19] for a perturbative treatment of the reservoir action on the electron, up to second order in the coupling constants. We trace over the reservoir variables, $\tilde{\sigma} := {\rm tr}_{\rm res}\{\tilde{\rho}\}$, since we are only interested in the atomic degrees of freedom. Rotating wave and Markov approximation, together with the neglect of any systematic degeneracy in the Floquet spectrum, yield a differential equation for $\tilde{\sigma}$ which we shall derive and discuss in detail in a separate publication. Back in the Schrödinger picture, $\hat{\sigma} := U_0 \tilde{\sigma} U_0^{\dagger}$, the matrix elements of the system's density operator $\hat{\sigma}_{\alpha\beta} = \overline{\langle \chi_\alpha(t)|}\hat{\sigma}(t)|\chi_\beta(t)\rangle$ obey the final master equation

$$\frac{\rm d}{{\rm d}t}\hat{\sigma}_{\alpha\alpha} = -\,\hat{\sigma}_{\alpha\alpha}\Gamma_\alpha - \,\hat{\sigma}_{\alpha\alpha}\sum\nolimits_{\mu} M_{\alpha\mu} + \sum\nolimits_{\mu} \hat{\sigma}_{\mu\mu} M_{\mu\alpha}, \tag{6}$$

$$\frac{\rm d}{{\rm d}t}\hat{\sigma}_{\alpha\beta} = \left[-i\,(E_\alpha - E_\beta) \,-\, \frac{1}{2}\left(\Gamma_\alpha + \Gamma_\beta + \sum\nolimits_{\eta}\left\{M_{\alpha\eta} + M_{\beta\eta}\right\}\right)\right]\,\hat{\sigma}_{\alpha\beta} \quad (\alpha \neq \beta)\,, \tag{7}$$

with "transition rates" $M_{\mu\nu}$ from $|\chi_\mu\rangle$ to $|\chi_\nu\rangle$. These equations completely describe the temporal evolution of the system in the *discrete* set of *decaying* dressed states $|\chi_\alpha\rangle$ and *exactly* incorporate the coherent coupling to the atomic continuum. The trace of $\hat{\sigma}$ over a complete set of square-integrable states leads to the expression

$$P_{\rm ion}(t) = 1 - \sum \hat{\sigma}_{\nu\nu}(t) \tag{8}$$

for the total ionization probability of the system.

Despite the seemingly intuitive form of eqs. (6)-(9), we emphasize that the $M_{\alpha\mu}$ are in general *complex* quantities, and so are the diagonal elements $\hat{\sigma}_{\alpha\alpha}$. Only for long-lived, "quasi-bound" states as our wave packet eigenstates are these effectively positive and can immediately be interpreted as transition and population probabilities, respectively. In general, for square-integrable initial conditions which involve coherences between different dressed states the imaginary parts of $\hat{\sigma}_{\alpha\alpha}$ are *essential* to obtain positive diagonal elements of $\hat{\sigma}$ with respect to square-integrable states. $P_{\rm ion}(t)$ is *always* a positive quantity since the imaginary parts of the $\hat{\sigma}_{\nu\nu}$ cancel out.

Focusing on the special case of spontaneous emission as the only source of decoherence, the transition rates from $|\chi_\mu\rangle$ to $|\chi_\nu\rangle$ read

$$M_{\mu\nu} = \sum\nolimits_k X^2_{\mu\nu}(k)\; K(|\Delta^k_{\mu\nu}|)\; \frac{1}{2}(1 + \mathrm{sgn}\Delta^k_{\mu\nu}). \tag{9}$$

They involve the (θ-independent) Fourier components of the dipole matrix element, the corresponding level spacings, and the response function of the reservoir at resonant frequency:

$$X_{\mu\nu}(k) := \sum\nolimits_\ell \overline{\langle\chi^\ell_{\mu\theta}|}R(\theta)xR(-\theta)|\chi^{\ell-k}_{\nu\theta}\rangle,$$
$$\Delta^k_{\mu\nu} := E_\mu - E_\nu + k\omega,$$

and

$$K(\Omega) := 2\pi \sum_{m,n,\lambda} \rho^{(\lambda)}_{mn}(\Omega)|g^{(\lambda)}_{mn}(\Omega)|^2\,. \tag{10}$$

Numerical results. – In our numerical calculations the mode densities $\rho^{(\lambda)}_{mn}$ and coupling constants $g^{(\lambda)}_{mn}$ in (10) are chosen as for a typical waveguide experiment, cf. [19] for details. Note, however, that $K(\Omega)$ is virtually identical to the response function in free space for large values of Ω which dominate the spontaneous decay of the wave packet eigenstates, as we shall see below. For comparison to previously published results [7], we define the amplitude and the frequency of the driving field by $Fn^4 = 0.04442$ and $\omega n^3 = 1$, yielding a fixed phase space structure. Here n indicates the principal quantum number of the Rydberg manifold the wave packet originates from. By numerical diagonalization of $\mathcal{H}^\theta_{\mathrm{sys}}$, we typically obtain 200 Floquet eigenstates per Floquet zone, with about 100 Fourier components each [17]. With these ingredients, (6) has been solved for n between 5 and 130. Initially, at $t = 0$, the wave packet eigenstate was assumed to carry all atomic population.

An inspection of the transition rates involving the wave packet shows that transitions to low-lying atomic states, which remain unaffected by the driving field, dominate the spontaneous decay of the wave packet. Asymptotically, all population which has not been lost through direct ionization to the atomic continuum is accumulated in the atomic ground state. The population of the wave packet can be well described by $\overline{P}_{\mathrm{wp}}(t) = \exp[-(\overline{M} + \Gamma_{\mathrm{wp}})\,t\,]$, with an effective spontaneous rate $\overline{M}$. Figure 1 compares the typical evolution of the wave packet's and of the ground-state population, together with the ionization probability $P_{\mathrm{ion}}(t)$. The figure shows that for dominant spontaneous decay, $\overline{M} \gg \Gamma_{\mathrm{wp}}$, the wave packets are effectively decoupled from the atomic continuum. Most of the population is transferred to (non-ionizing) low-lying atomic states before the coherent coupling to the continuum can induce appreciable ionization.

As already mentioned, the tunneling picture for the decay to the atomic continuum implies that the ionization rate Γ_{wp} decreases *exponentially* (on the average) with n [9]. In contrast, the spontaneous decay rates of unperturbed Rydberg states are known to decrease *algebraically* like n^{-a}, with a between 3 and 5, depending on the quantum numbers of the Rydberg state [20]. Hence, for fixed classical phase space structure and sufficiently large values of n, spontaneous emission should dominate the population loss out of the wave packet eigenstate [9]. This is exactly what is observed in fig. 2, which summarizes the comparison of spontaneous rates $\overline{M}$, resonance widths Γ_{wp}, and total decay rates $\overline{R} = \overline{M} + \Gamma_{\mathrm{wp}}$ extracted from the solution of (6) for the wave packet states over a large range of principal quantum numbers. Indeed, for n between 5 and 93 the coherent coupling to the atomic continuum is generally dominant over the spontaneous decay, except for the occasional appearance of extremely long-lived wave packet states. In this regime, $\overline{R}$ displays qualitatively the same fluctuations as Γ_{wp}, which can be

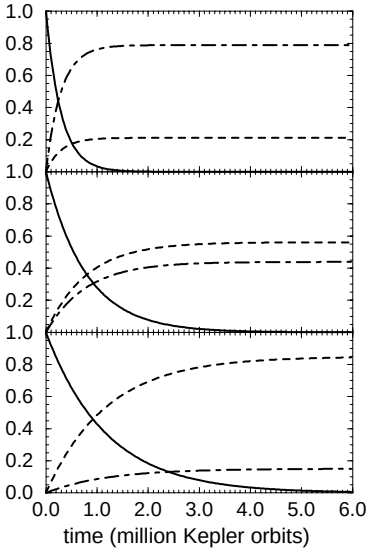

Fig. 1

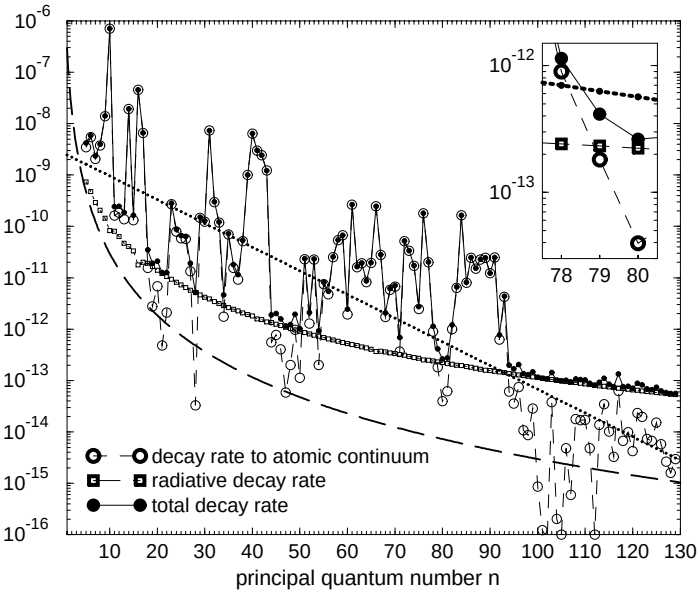

Fig. 2

Fig. 1. – Population of the wave packet eigenstate (solid line) and of the atomic ground state (dashed line), together with the total ionization probability (dash-dotted line), at *fixed* classical phase space structure for principal quantum numbers $n = 78$ (top), 79 (middle), 80 (bottom). As the radiative decay of the wave packet exceeds the decay to the atomic continuum, the total ionization yield is suppressed.

Fig. 2. – Comparison of the decay rates to the atomic continuum $\Gamma_{\rm wp}$, the radiative decay rates $\overline{M}$, and the total decay rates $\overline{R}$ of the nondispersive wave packets, as a function of the principal quantum number n. To fix the classical phase space, the field amplitude and frequency are chosen as $Fn^4 = 0.04442$ and $\omega n^3 = 1.0$, respectively. One observes a crossover from coherent to radiative decay. The dotted line indicates the overall *exponential* decay of $\Gamma_{\rm wp}$ as opposed to the *algebraic* decay of $\overline{M}$ proportional to n^{-3} (1D, squares) or n^{-4} (3D, dashed line). The chaos-induced [7] fluctuations of $\Gamma_{\rm wp}$ are partially amended for intermediate and virtually suppressed for large values of n. The inset shows the different rates yielding the temporal evolution in fig. 1.

attributed to chaos-assisted tunneling [7]. In contrast, for principal quantum numbers $n \geq 94$ spontaneous emission dominates over the coherent continuum coupling and the fluctuations of $\Gamma_{\rm wp}$ are largely suppressed in the dependence of $\overline{R}$ on n.

Our numerical results are obtained for 1D wave packet eigenstates which have their direct 3D analog in wave packets tightly confined to the vicinity of the polarization axis of the driving field and are essentially composed of extremal parabolic states [8]. Whereas this 1D description of the atom is perfectly justified in the Rydberg regime [21], it implies an unrealistic representation of the atomic ground state, which acquires a permanent dipole moment. As a consequence, transitions from an extremal parabolic state to the ground state are overestimated. To compensate for this artifact of the 1D picture, we rescale the radiative transitions to low-lying states by the ratio of the dominant 3D and 1D dipole matrix element (which differs from unity only for low-lying states). The dashed line in fig. 2 shows the resulting estimate for the quasi-1D wave packet of the real atom [8], which scales as n^{-4}, like the emission rate of extremal parabolic states. Since this curve only accounts for transitions within the ladder of extremal parabolic states, it underestimates the actual spontaneous decay rates. Therefore, we predict the crossover from dominant continuum coupling to dominant spontaneous decay to occur in the interval $n_{\rm crit} = 94 \ldots 145$. The upper limit has been obtained from an extrapolation of the dotted and the dashed lines in fig 2.

Summary. – We outlined a novel approach to describe dissipative dynamics on a basis of decaying states, which is generally applicable to the Markovian coupling of a decaying quantum system to an environment. Its application to the case of quasi-1D wave packet eigenstates which interact with the electromagnetic vacuum leads to the prediction of a crossover from dominant continuum coupling to dominant radiative decay, at principal quantum numbers which are accessible in state of the art microwave experiments. This crossover has a clear signature: for $n < n_{\rm crit}$ chaos-induced fluctuations of the total decay rate as a function of n prevail, whereas they are smoothed out for $n > n_{\rm crit}$([2]). An experimental scheme to access the decay rates has been outlined in [22].

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([2]) Qualitatively the same conclusions apply for wave packet eigenstates traveling along low eccentricity and circular trajectories. However, the critical values $n_{\rm crit}$ for the described crossover will be considerably higher, due to the scaling n^{-5} of the spontaneous lifetimes of circular Rydberg states. Hence, spontaneous emission is experimentally irrelevant for circularly driven wave packets, a conclusion which has been drawn independently by I. Bialynicki-Birula and Z. Bialynicka-Birula (preprint submitted to *Phys. Rev. A*, 1997).