Rydberg States in Laser Fields: Wave Packets and Near-Threshold Phenomena

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1. Introduction

The description of the behavior and motion of Rydberg electrons excited by laser radiation is an essential element in our understanding of multiphoton processes in atoms and molecules [1,2]. Here we consider two aspects of this problem: (i) We discuss generation of Rydberg wave packets by short laser pulses; and their detection in a two-photon experiment with time-delayed pulses [3,4], (ii) We develop a theory of excitation of (autoionizing) Rydberg states which from the outset includes the laser-induced mixing between **all** bound and continuum states of a Rydberg series [5].

When a short laser pulse of duration τ excites electrons from a low-lying atomic level to Rydberg states, wave packets are formed because (i) photon absorption from the initial state to the Rydberg series is localized in space in a volume of the size of the initial state, which is small compared with the spatial extent of Rydberg states; and (ii) provided the laser pulse is so short that its spectral width simultaneously excites many Rydberg levels around some mean energy $E_{\bar{n}} < 0$:

$$\hbar/\tau \gg \frac{1}{2\pi}(E_{\bar{n}+1} - E_{\bar{n}}) \simeq \frac{1}{2\pi}\left.\frac{dE}{dn}\right|_{\bar{n}} = \hbar/T_{E_{\bar{n}}} \quad . \tag{1}$$

Here E_n are Rydberg energies for the principal quantum number n and $T_{E_{\bar{n}}}$ is the classical orbit time corresponding to a particle of energy $E_{\bar{n}}$ moving on a Kepler orbit. According to Equ. (1) wave packets are excited when the pulse duration is short compared with the classical orbit time. In this sense Rydberg wave packets can be interpreted as quantum beats between n-states. In Sec. 2 we give a brief summary of a recently developed semiclassical formalism to describe generation and detection of Rydberg wave packets [3]. In particular, we present a semiclassical analysis of a two-photon process with two time-delayed laser pulses: a first short pulse excites a wave packet whose motion is probed by a second pulse. This provides us with a time and space-sensitive measurement since absorption or induced emission of a probe laser photon can only occur near the inner turning point of the classical Kepler orbit where acceleration of the electron is largest [6]. Thus the two-photon transition probability will show peaks, whenever the time

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delay between the laser pulses in a multiple of a classical orbit time (as long as the wave packet has a well-defined structure).

Our foregoing discussion has emphasized that absorption (emission) of laser photons occurs in a **finite** interaction volume, much smaller than the size of Rydberg orbitals and almost independent of energy. This physical picture is reflected in the fact that the oscillator strength distribution from Rydberg states to other bound or free orbits is a smooth function of energy across the Rydberg threshold. We can make this a basis of a theory of near-threshold behavior of Rydberg states in laser fields. The idea is to solve the problem of interaction of laser radiation with the Rydberg electron above threshold and then to extrapolate this information to the bound Rydberg series [5] using the properties of Coulomb functions [7]. The essential feature of this theory is that it considers the interaction with a Rydberg series as a whole (as opposed to treating the interaction with each Rydberg level separately). Thus from the outset laser-induced mixing between all bound and continuum states of one series is included. Sec. 3 briefly summarizes results obtained for strong field excitation of autoionizing resonances [5].

2. Generation and Detection of Rydberg Wave Packets by Short Laser Pulses

We consider an atom with a single valence electron which is excited by a short pulse at time t_a from an initial state $|i\rangle$ (energy E_i) to Rydberg states. The electric field has an amplitude $\mathcal{E}_a(t)$, mean frequency ω_a and pulse duration τ_a. In perturbation theory we find for the electron wave function at a time $t > t_a + \tau_a$ after the pulse

$$|\psi(t)\rangle = |i\rangle e^{-iE_i t/\hbar} + |\psi_i^{(a)}(t)\rangle \qquad \text{with} \tag{2}$$

$$|\psi_i^{(a)}(t)\rangle = \sum\!\!\!\!\!\!\int_{n,l,m_l} |nlm_l\rangle\, e^{-iE_{nl}t/\hbar}\, \frac{i}{\hbar}\, \mu^{(a)}_{nlm_l,i}\, \tilde{\mathcal{E}}(\delta^{(a)}_{in})\, e^{\delta^{(a)}_{in} t_a} \quad . \tag{3}$$

Here $|nlm_l\rangle$ are eigenstates of the atomic Hamiltonian H_A for the energy E_{nl}; l and m_l are angular momentum quantum numbers. The dipole operator is denoted by μ, $\delta^{(a)}_{in} = (E_{nl} - E_i - \hbar\omega_a)/\hbar$ are detunings. The spectral density of the pulse is defined by

$$\tilde{\mathcal{E}}(\Delta\omega) = \int_{-\infty}^{+\infty} dt\; \mathcal{E}_a(t)\, e^{i\Delta\omega(t-t_a)}. \tag{4}$$

If the laser pulse $\mathcal{E}_a(t)$ is short in the sense of Equ. (1), $|\psi_a^{(i)}(t)\rangle$ describes a Rydberg wave packet; at least as far as the radial motion is concerned: due to dipole selection rules only few angular momenta contribute in the sum (3), so there is no further localization in the angular variables. Since the spectral density $\tilde{\mathcal{E}}_a$ is concentrated in an energy band $-\hbar/\tau_a \lesssim E - E_i + \hbar\omega_a \lesssim \hbar/\tau_a$ with the

maximum centered around $E_{\bar{n}} = E_i + \hbar\omega_a$, only Rydberg states in this energy interval will contribute in Equ. (3).

We now turn to a discussion of the motion of the wave packet (3). This serves as a guideline for our interpretation of two-photon processes with time-delayed pulses. We expect that the radial center of the wave packet (to the extent it is well defined) follows a classical Kepler orbit. The propagator which governs the time evolution in Equ. (3) for $t \geq t'$ is

$$G^+(\vec{x},t|\vec{x}',t') = e^{-iH_A(t-t')} = -\frac{1}{2\pi i}\int_{-\infty}^{+\infty} dE\, e^{-iE(t-t')} \times \times \sum_{lm} \frac{g_{El}(r,r')}{rr'} Y_{lm}(\hat{\vec{x}})\, Y^*_{lm}(\hat{\vec{x}}') \tag{5}$$

where $g_{El}(r,r')$ is a radial Green's function of the atom with outgoing wave boundary conditions. The key to a semiclassical analysis and physical interpretation is to rewrite this Green's function in the form [8]

$$g_{El}(r,r') = \begin{cases} g^{(s)}_{El}(r,r') & (E > 0) \qquad (6a) \\ g^{(s)}_{El}(r,r') - 2\pi i \dfrac{S(E,l,r)S(E,l,r')}{e^{-2\pi i(\nu+\alpha)} - e^{-2\pi\beta}} & (E < 0,\ \beta \to 0)\ . \qquad (6b) \end{cases}$$

The Green's function for an energy E above threshold ($E > 0$) has been denoted by $g^{(s)}_{El}(r,r')$ where the superscript indicates that it is a smooth Green's function, i.e. free of singularities in E. The explicit mathematical form is not relevant for us at this point. Of course, $g^{(s)}_{El}(r,r')$ is not a physical Green's function below threshold since it diverges for $r_2 < r \to \infty$ with r_2 the outer turning point of the Kepler orbit for the energy $E < 0$. This asymptotic divergence is compensated by the second term in Equ. (6b) where $S(E,l,r)$ is the regular solution of the radial Schrödinger equation (normalized on the energy scale). $S(E,l,r)$ diverges exponentially for $E < 0$ except at the bound state energies $E_n = -Ry/\nu_n^2$ with $\nu_n = n - \alpha$ (n = integer, α = quantum defect) [7]. As expected the second term in Equ. (6b) has poles at these bound state energies. It is the resonant part of the Green's function.

Let us assume that the laser pulse excites a superposition of bound Rydberg states. The propagation of $|\psi^{(a)}_i(t)\rangle$ is then described by

$$g_{El}(r,r') = g^{(s)}_{El}(r,r') - 2\pi i \sum_{m=1}^{\infty} e^{2\pi i(\nu+\alpha)m} S(E,l,r)S(E,l,r') \tag{7}$$

where we have expanded the resonance denominator in Equ. (6b). Substituting Equ. (7) into (5) and (3), and evaluating the resulting integral over the energy (5) in a stationary phase approximation leads to the following interpretation:

(i) For times $t - t_a < T_{E_{\bar{n}}}/2$ only the first term in Equ. (7) contributes. It describes a radial wave packet moving on a Kepler orbit

$$\int_{r_1}^{r(t)} \frac{\mu}{p(r')}\, dr' = t - t_a \qquad (t - t_a > 0) \tag{8}$$

with $p(r)$ the radial momentum of the electron, μ the electron mass and r_1 the inner turning point. We emphasize that - although we have a bound wave packet - its motion is governed by a continuum propagator $g_{El}^{(s)}(r,r')$. This is not surprising because for $t - t_a < T_{E_{\bar{n}}}/2$ does not "know" that its radial motion is bounded, as it did not have time to "see" the potential barrier near the outer turning point r_2. In a two-photon experiment with short pulses (no time delay) and Rydberg levels as intermediate states this corresponds to the absence of intermediate state resonances (**direct** process).

(ii) For times $t - t_a > T_{E_{\bar{n}}}/2$ the wave packet is reflected from the outer potential barrier. This periodic motion on a Kepler orbit is described by the second term in Equ. (7)

$$\pm \int_{r_1}^{r(t)} \frac{\mu}{p(r')}\, dr' = t - t_a - m \cdot T_{E_{\bar{n}}} \qquad (m = 1,2,\ldots) \tag{9}$$

with

$$T_E = \frac{\pi\hbar}{Ry} (-E/Ry)^{-3/2} \equiv \frac{\pi\hbar}{Ry} \nu^3 \ . \tag{10}$$

According to Equ. (9) the $m = 1,2,\ldots$ terms in Equ. (7) correspond to the first, second return of the wave packet to its inner turning point. It is possible to observe this periodic motion in a two-photon process with time-delayed pulses: a first pulse at t_a excites the wave packet which is probed at a later time t_b by a second short pulse. As we remarked earlier the transition probability $P_{f \leftarrow i}$ will show peaks at $\Delta t = t_b - t_a = m T_{E_{\bar{n}}}$ $(m = 1,2,\ldots)$ since absorption of the probe photon occurs near the inner turning point of the Kepler orbit. We emphasize the close connection between a resonant two-photon process and the observation of wave packets in a time-delayed pulse experiment (**indirect** or **two-step** process).

In Fig. 1 the radial function of r (in units of the Bohr radius) is shown at time $t - t_a = 1/9$, $2/9$ and $3/9 \times T_{E_{\bar{n}}}$ for a Gaussian laser pulse (duration 8 psec) exciting hydrogen p states around $\bar{n} = 85$ ($T_{E_{\bar{n}}} = 94$ psec). Fig. 2 is a plot of a two-photon Raman transition probability as a function of the time delay $t - t_a$ between the two pulses measured in units of a classical orbit time ($\bar{n} = 85$; $\tau_a = \tau_b = 5$ psec). The dashed line is the semiclassical (stationary phase) approximation which compares well with a calculation based on an explicit summation over n-states (solid line). A sweeping of the laser frequency during the pulse can lead to an initial contraction of the wave packet (Fig. 3). Finally, Fig. 4 shows

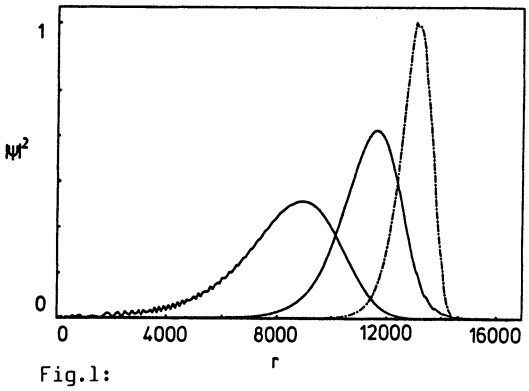

Fig.1:
Radial motion of the wave packet

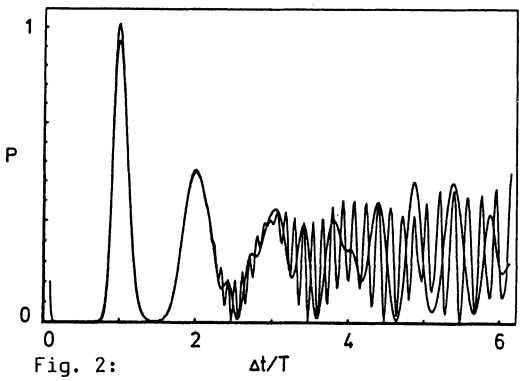

Fig. 2:
Raman transition probability as a function of the time delay between the two pulses in units of the classical orbit time (dashed line = semiclassical approximation)

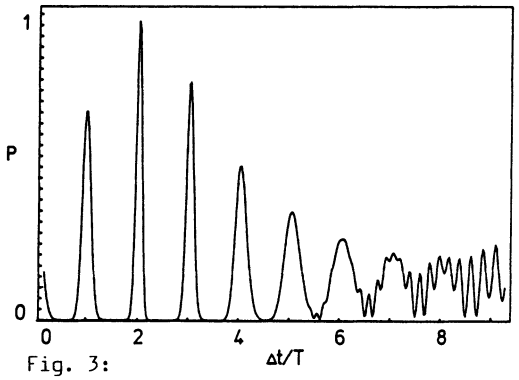

Fig. 3:
Same as Fig. 2 but with a change of the laser frequency during the pulse

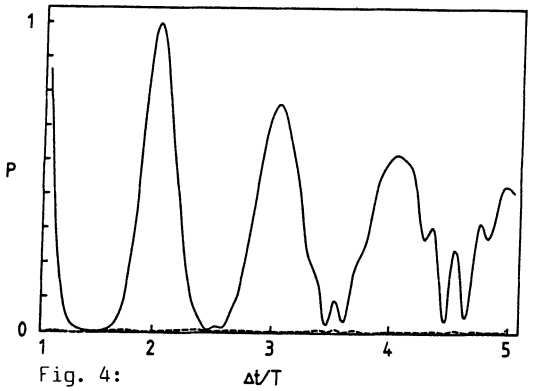

Fig. 4:
Constructive (solid line) and destructive interference (dashed line) between wave packets in a Raman experiment.

interference between two wave packets: the first one is excited at t_a, the second one by a short pulse at time $t_{a'} = t_a + T_{E_{\bar{n}}}$ ($\bar{n} = 85$). It is possible to probe this interference by a third time-delayed pulse at $t_b = t_{a'} + T_{E_{\bar{n}}}$; depending on the relative phase between the first and second pulse the two wave packets interfere constructively (solid line) or destructively (dashed line).

3. Strong Field Excitation of an Autoionizing Rydberg Series Near Threshold

We consider excitation of an autoionizing Rydberg [9] series in a configuration where a first laser excites electron from the groundstate $|g\rangle$ with energy E_g to one of the excited states $|e\rangle$ (energy E_e) by N-photon absorption which is coupled

by a stronglaser of frequency ω_2 to an autoionizing Rydberg series. For weak excitation by the first the ionization is

$$R = \frac{1}{2}\,\Omega_{eg}^2 \quad \mathrm{Im}\left\{\frac{-1}{E_g+N\omega_1-E_e-\Sigma}\right\} \qquad \text{with} \tag{11}$$

$$\Sigma = \langle e|D\,\frac{1}{E_g+N\omega_1+\omega_2-H_A+i\epsilon}\,D|e\rangle \qquad \epsilon \to 0 \tag{12}$$

the resonant part of the complex selfenergy of $|e\rangle$ H_A is the atomic Hamiltonian, D the dipole operator describing interaction with the strong field. For $E_I < E_e+\omega_2$, i.e. in the unstructured continuum (Fig. 5), Σ is a slowly varying function energy; in the autoionizing region $0 < E_1+{}_2 < E_I$. Σ contains the infinite series of Rydberg resonances. To extract this rapid energy dependence we relate the "above" and "below" threshold self-energy. We obtain

$$\Sigma = \begin{cases} \delta\omega_{bg} - i\frac{1}{2}\gamma\,\dfrac{1+q^2r^2}{1+r^2} & \text{(flat continuum)} \\ \delta\omega_{bg} - i\frac{1}{2}\gamma\,(1+\dfrac{(q-i)^2}{x+i}) & \text{(autoionization)}\ . \end{cases} \tag{13}$$

Here $\delta\omega_{bg}$ is a back ground shift in the flat continuum above the second threshold; γ is the ionization width state $|e\rangle$ to the first continuum. The detuning variable is defined as

$$x = \tan\pi\,(\nu_2 + \alpha)/r^2 \tag{14}$$

with the effective quantum number ν_2 given by

$$E_g + N\hbar\omega_1 + \hbar\omega_2 = E_I - Ry/\nu_2^2\ . \tag{15}$$

α is the unperturbed quantum defect of series 2 and r is a measure of the configuration interaction between the two channels; the Fano parameter q is a measure of the interference between direct ionization of $|e\rangle$ to the first continuum and excitation to the autoionizing resonance followed by decay to the continuum. It is of central importance that the parameters γ, q, α and $\delta\omega_{bg}$ are essentially constant along the Rydberg series and across the threshold.

We emphasize the following features:

(i) Σ and, therefore, R contain n-mixing by the second laser of the infinite number of Rydberg states in terms of a small set of parameters which vary slowly with energy.

(ii) The two-level result (for a single autoionizing resonance) is obtained by expanding the detuning variable x around the Rydberg state n,

$$x \to (E_g + N\hbar\omega_1 + \hbar\omega_2 - E_I + Ry/\nu_2^{(n)2}) \quad / \frac{1}{2}\Gamma \tag{16}$$

with $\Gamma = 4r^2/\nu_2^{(n)3}$ the autoionization width. This result is valid when Γ and the Rabi frequency for $|e\rangle \rightarrow |n\rangle$ (which scales proportional to $\sim\nu_2^{(n)-3/2}$) are both much smaller than the level separation of the Rydberg series. It is remarkable that the generalization from two-level theory to infinite Rydberg series can be obtained by the simple substitution rule (16).

(iii) Σ averaged over the resonance is continuous across threshold.

In Figs. 6a and b the ionization R is plotted as a function of the detuning of the first (weak) laser. At sufficiently low intensities and for well isolated autoionizing resonances the two-level approximation for excitation from $|e\rangle$ to the autoionizing state is valid. In this case a doublet splitting (AC-Starksplitting) of the ionization signal as a function of detuning of the first laser is observed

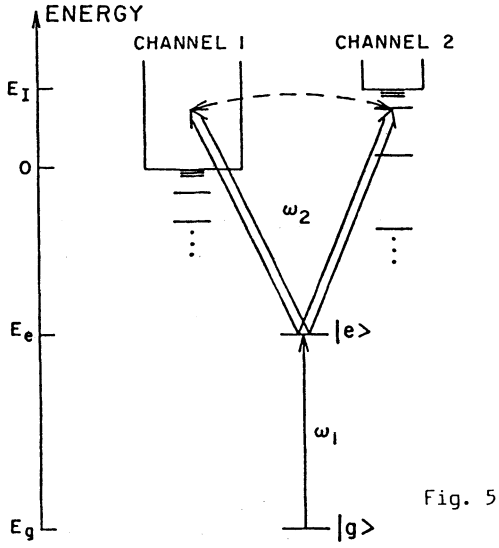

Fig. 5: Atomic configuration

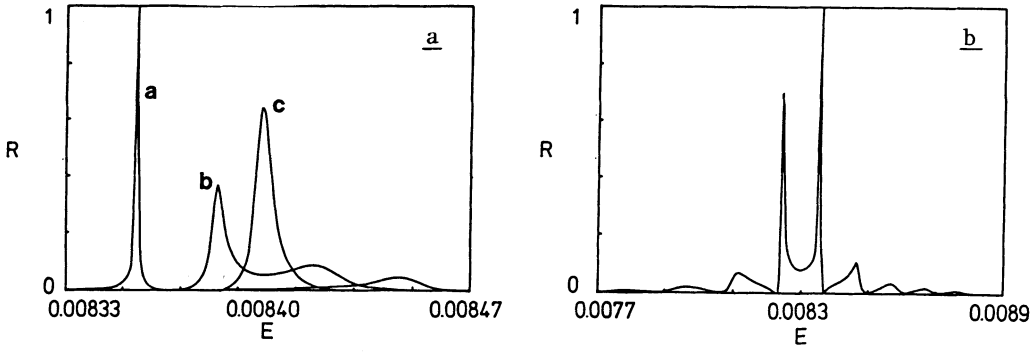

Fig. 6: Ionization rate R as a function of the detuning of the first (weak) laser (AC-Starksplitting of autoionizing resonances). E denotes the final state energy in Rydbergs. (a) The two-level approximation is valid. (b) AC-Starksplitting of overlapping autoionizing resonances.

(Fig. 6a). The asymmetry of the spectrum is a consequence of the interference between direct ionization to the continuum and the indirect (resonant) transition via the autoionizing resonance [9]. In Fig. 6b the ionization rate is plotted at high intensities and for overlapping autoionizing resonances. Laser-induced mixing of many n-states of the Rydberg series is observed.

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