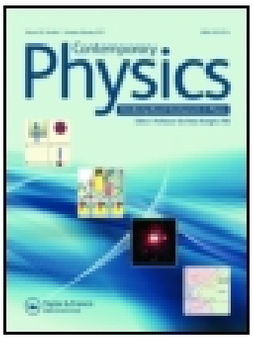

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Laser-induced excitation of electronic Rydberg wave packets

G. ALBER and P. ZOLLER

We give a qualitative review of theoretical ideas and experimental work on laser-induced excitation of atomic Rydberg wave packets. Studying the motion of Rydberg wave packets with the help of short or intense laser pulses provides a bridge between quantum mechanics and the classical concept of the trajectory of an electron and corresponds to the real-time observation of atomic dynamics.

Recent progress in generating short and intense laser pulses has stimulated considerable interest in coherent laser excitation of atoms (Alber and Zoller 1991, Ellinger *et al.* 1991, A. ten Wolde *et al.* 1988, Yeazell *et al.* 1990, Meacher *et al.* 1991, Yeazell and Stroud 1988, Noordam *et al.* 1989, Alber, 1989) and molecules (Bernstein and Zewail 1989). Short laser pulses offer the possibility to excite *coherently* a large number of atomic or molecular energy-eigenstates, thus preparing nonstationary electronic charge distributions. Whereas energy-eigenstates describe stationary electronic states which are delocalized in space, these non-stationary charge distributions represent electronic wave packets whose probability density varies with time and is well localized in space in comparison with the classically accessible region. In contrast to frequency-resolved laser spectroscopy which aims at selective excitation of energy-eigenstates, laser excitation with short pulses makes possible the study of the dynamics of spatially localized electronic wave packets. A well-known example of wave packets in quantum mechanics is the coherent state of the harmonic oscillator. In this contribution we discuss laser-induced excitation of electronic wave packets in Rydberg-atoms (called Rydberg wave packets in the following). This is a subject which has received considerable attention in atomic physics and quantum optics during the last couple of years. A detailed review can be found in Alber and Zoller (1991) and Ellinger *et al.* (1991).

Rydberg atoms are characterized by high principal quantum numbers n of the valence electron. Typically, the size of these atoms is large on the scale given by the Bohr radius a_0: for example, the mean distance $\langle r \rangle$ of a hydrogenic electron in an energy eigenstate with principal quantum numbers $n = 85$ is of the order $\langle r \rangle = 10^4\ a_0$. In general the Rydberg eigenfunctions $R_{nl}(r)Y_{lm}(\theta,\phi)$ with $R_{nl}(r)$ radial eigenfunctions and $Y_{lm}(\theta,\phi)$ spherical harmonics extend over a large region of space (proportional to $n^2 a_0$) for the energy $E_{nl} = -Ry/(n-\mu_l)^2$ with Ry the Rydberg constant and μ_l a quantum defect. In other words, the stationary energy eigenfunctions are delocalized. A Rydberg wave packet consists of a superposition of atomic Rydberg states,

$$\Psi(x,t) = \sum_{n,l,m} a_{nlm} R_{nl}(r) Y_{lm}(\theta,\phi) e^{-iE_{nl}t/\hbar}. \qquad (1)$$

By forming the superposition the wave function becomes localized and non-stationary. Currently, two cases of electronic Rydberg wave packets are discussed in the literature: Radial Rydberg wave packets (ten Wolde *et al.* 1988, Yeazell *et al.* 1990, Meacher *et al.* 1991) are localized only with respect to the radial electronic coordinate r and consist of a coherent superposition of a large number of Rydberg states with different principal quantum numbers n but few values of the angular and magnetic quantum numbers l and m; angular Rydberg wave packets, (Yeazell and Stroud 1988, Noordam *et al.* 1989) on the other hand, are localized with respect to the angular coordinates θ, ϕ, that is, are formed by coherent superpositions of Rydberg states with different values of l and m and fixed n.

Studies of Rydberg wave packets are motivated by a number of interesting physical questions (Alber and Zoller 1991, Ellinger *et al.* 1991). Because of their large size and large quantum numbers, Rydberg atoms are interesting physical objects on the borderline between classical and quantum mechanics. On one hand a Rydberg wave packet exhibits classical features: as long as the wave packet is well localized its centre moves

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according to the laws of classical mechanics on a Kepler orbit. Thus, Rydberg wave packets are a way of studying the *dynamics* of an excited atomic electron in a Coulomb potential. On the other hand, a Rydberg wave packet also shows characteristic quantum features. An example is the spreading of the wave packet with time: an initially localized wave packet will, in general, spread with time because the non-equidistant energy level spacing of the Rydberg states will lead to a dispersion (similar to the spreading of a free Gaussian wave packet); for very long times, however, a revival of the wave packet can be observed which is related to the discreteness of the quantized energy levels (see below). A second example of a quantum effect is the possibility of wave packets to interfere with each other. Finally, an interesting aspect is concerned with the destruction of quantum coherence (destruction of the phase relationship) between wave packets due to their coupling to fluctuations introduced by coupling to an environment.

Let us now turn to the question of how short laser pulses excite Rydberg wave packets (Alber and Zoller 1991, Ellinger *et al.* 1991). Consider an atom which is excited from its ground state $|i\rangle$ by a short laser pulse of duration τ to Rydberg $|nlm\rangle$ states close to the photoionization threshold (figure 1). We denote the mean principal quantum numbers and energy of the excited atomic states by $\bar{n}$ and $E_{\bar{n}l}$, respectively. If the spectral width of the laser pulse $\hbar/\tau$, is much larger than the mean energy separation between adjacent Rydberg states $\Delta E_{\bar{n}l}$, many Rydberg states are excited coherently. This is equivalent to the condition that the pulse duration τ is much less than the mean classical orbit time T_{cl} of the excited valence electron: for hydrogen we have

$$T_{cl} = 2\pi\hbar/\Delta E_{\bar{n}l} = \pi\bar{n}^3\hbar/Ry. \tag{2}$$

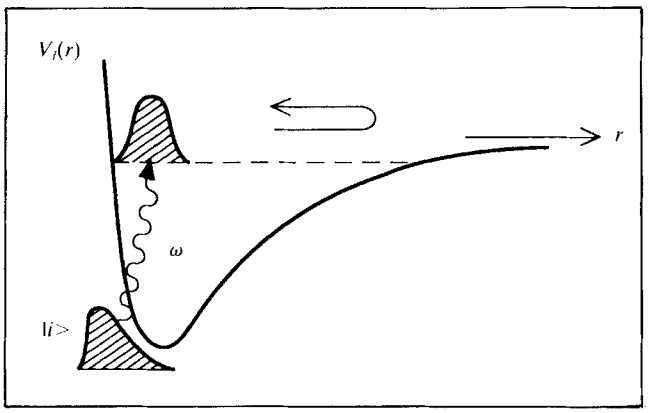

Figure 1. Excitation of a radial Rydberg wave packet by a short laser pulse. Initially the electron is in the ground state $|i\rangle$. A short laser pulse excites a superposition of Rydberg states. This corresponds to a wave packet moving in the effective radial atomic potential (given by the Coulomb potential and the centrifugal barrier).

Furthermore, the atomic ground state $|i\rangle$ is localized in a region of a few Bohr radii around the atomic nucleus which is small in comparison with the size of the Rydberg states. This implies the spatial localization of the laser-induced excitation process to a small region around the atomic nucleus of the extension of the initial atomic state. This laser-induced excitation process is therefore localized in *space* and *time*, that is an electronic wave packet as a superposition of different n-states is generated. Note, however, due to dipole selection rules ($l \to l \pm 1$) in such a laser excitation process only a few angular momentum eigenstates can be excited so that the angular coordinates of the Rydberg electron are still delocalized in a quantum mechanical sense. This corresponds to a radial Rydberg wave packet. Figure 1 is a schematic representation of this excitation process: we have plotted the effective radial potential as seen by the Rydberg electron, consisting of a centrifugal barrier near $r \to 0$ and a Coulomb tail for large r. The excited Rydberg wave packet will move between the two classical turning points.

The time-evolution of the wave packet in the atom can be probed in a two-photon experiment with time delayed short pulses (figure 2 (*a*), (*b*), (Alber and Zoller 1991, Ellinger *et al.* 1991)). In such a pump-probe experiment a first short laser pulse generates a wave packet (see figure 1). The second short laser pulse which is time-delayed by ΔT with respect to the first pulse probes the return of the wave packet to the atomic core region inducing, for example, a transition of the electron to the continuum. Note that a Rydberg electron absorbs optical radiation in the near core region; far from the nucleus the electron moves like a quasi-free particle which does not absorb light. Therefore, for the Rydberg wave packet the probability of being ionized is largest when it returns to its inner classical turning point, that is, if the time delay between pump and probe pulse is a multiple of the mean classical orbit time T_E. In other words, the transition probability of such a pump-probe experiment as a function of the time delay between both laser pulses is expected to exhibit characteristic maxima at multiples of the mean classical orbit time of the excited Rydberg states reflecting the dynamics of the excited Rydberg wave packet in the Coulomb field. Such pump-probe experiments for studying the dynamics of a radial Rydberg wave packet have recently been performed at the FOM-institute in Amsterdam (ten Wolde *et al.* 1988), at the Optical Sciences Center of the University of Rochester (Yeazell *et al.* 1990) and recently in Oxford (Meacher *et al.* 1991). Figure 3 shows the photoionization signal as a function of the time delay between two short optical pulses ($\tau = 15\ ps$) exciting a wave packet in a sodium near $n = 65$, as given by Stroud and coworkers (Yeazell *et al.* 1990). The upper figure is the experimental

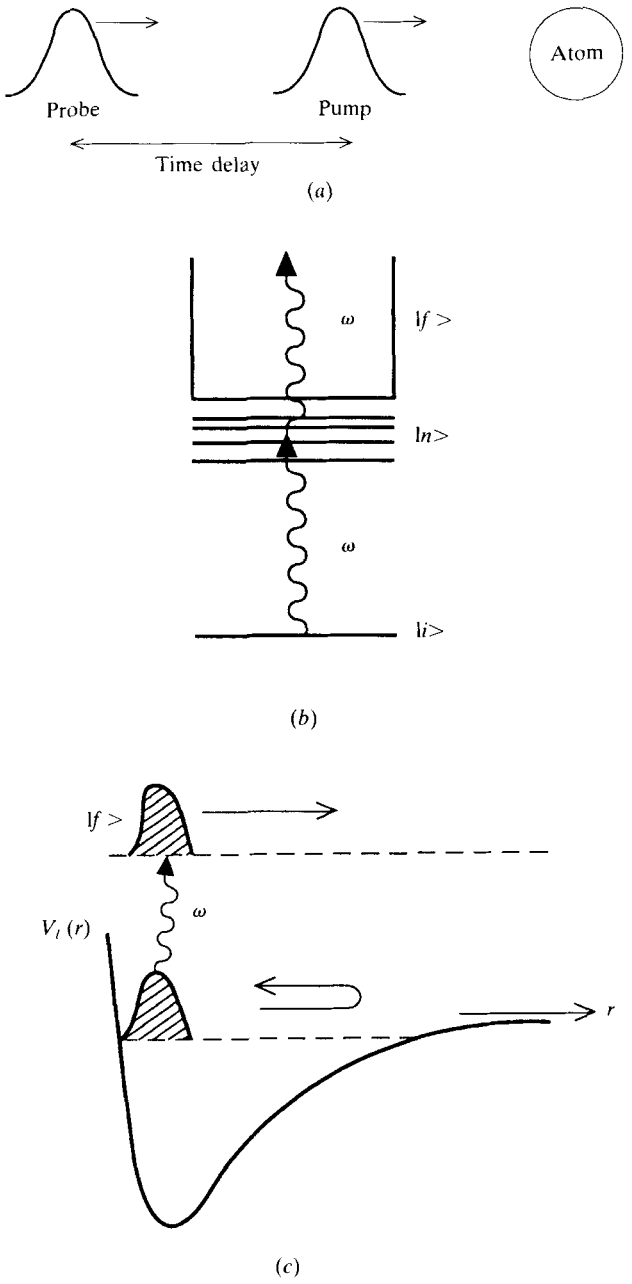

Figure 2. Pump-probe scheme to excite and detect the motion of a radial Rydberg wave packet. (*a*) Time-delayed pump and probe beams exciting the atom (*b*) Atomic excitation scheme: the first (pump) pulse excites the wave packet which is probed by the second time-delayed pulse (*c*) The return of the wave packet to the ion core is probed by the second pulse which ionizes the electron near its inner classical turning point.

result, the lower figure the theoretical prediction. Oscillations at the orbital period indicating the return of the wave packet to the atomic core are clearly evident in these figures. Initially, the wave packet decays in time (spreading of the wave packet), which is related to the anharmonic level spacing of the Rydberg series in equation (1).

After a revival time the wave packet will rephase again and will again be localized (see Alber and Zoller 1991; Ellinger *et al.* 1991, A. ten Wolde *et al.* 1988, Yeazell *et al.* 1990, Meacher *et al.* 1991, for references): this corresponds to a scenario of fractional revivals of the wave packet. To understand these revivals we rewrite equation (1) by expanding E_n to second order around the mean principal quantum number $\bar{n}$,

$$|\Psi(t)\rangle = \sum_n |n\rangle e^{-2\pi i(\Delta n t/T_{cl} + \Delta n^2/T_{rev})}, \tag{3}$$

where $\Delta n = n - \bar{n}$ and T_{cl} are the classical oscillation period and T_{rev} denotes the revival time (for simplicity we suppress the quantum numbers l and m). The simplest revival occurs at $t \sim \frac{1}{2}T_{rev}$ where

$$|\Psi(t)\rangle \sim |\Psi(t + \tfrac{1}{2}T_{cl})\rangle_{cl}, \tag{4}$$

with

$$|\Psi(t)\rangle_{cl} = \sum_n |n\rangle e^{-2\pi i \Delta n t/T_{cl}}, \tag{5}$$

a 'classical' (nondecaying) wave packet without the dispersive term. This revivals can be clearly seen on the right of figure 3. Another revival takes place at $t \sim \frac{1}{4}T_{rev}$ where

$$|\Psi(t)\rangle \sim 2^{-1/2}(e^{-i\pi/4}|\Psi(t)\rangle_{cl} + e^{+i\pi/4}|\Psi(t + T_{cl}/2)\rangle_{cl}). \tag{6}$$

Equation (6) represents a non-classical object formed by *two* correlated wave packets which are spatially separated within the classical orbit. An interesting feature of this long-term evolution is the appearance of oscillations with twice the orbital frequency, which can be attributed to this break up of the wave packet into two macroscopic parts. Thus one expects a doubling of the wave packet period which can be seen for intermediate times in figure 3. Very recently, beautiful observations of fractional revivals were reported also by Meacher *et al.* (1991).

Excitation of angular wave packets requires, in addition to a short laser pulse, a dressing of the Rydberg states by an external field to form the superposition of different l,m states. An external field is required because in an unperturbed atom a laser cannot excite simultaneously l-states (for example an s ground state will only couple to p-states due to dipole selection rules). In an external field different l and m-states will be mixed, that is, a laser can excite a coherent superposition of these 'dressed states'. When turning the external field off very slowly each of the dressed states is coupled adiabatically to a particular l,m-level, so that we are left with a

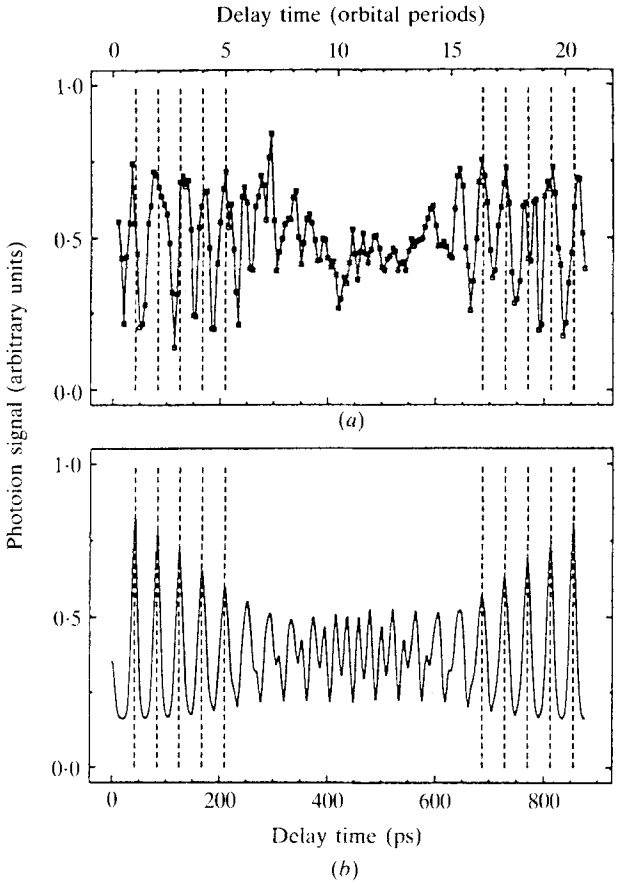

Figure 3. The photoionization signal as a function of the delay between the pump and probe pulse time shows the evolution of the radial wave packet: (*a*) experiment and (*b*) theory (reproduced from ten Wolde *et al.* (1988) with the authors' permission).

coherent superposition of angular momentum states. The dynamics of this angular wave packet corresponds with the time evolution of the Runge–Lenz vector. In a pure Coulomb potential this vector is a constant of motion and is directed along the major axis of the classical Kepler ellipse. A time evolution of this Runge–Lenz vector reflects the deviation of the potential acting on the excited valence electron from the pure Coulomb field due to core polarization in addition to relativistic effects (analogous to the precision of the perihelion of Mercury). A core polarization potential arises in atoms due to the perturbation of the spherical symmetry of the ion core by the valence electron. Stroud and coworkers (Yeazell and Stroud 1988) reported an angular wave packet experiment where Rydberg states with the same radial quantum number n but different angular quantum numbers l and m are excited in the presence of a radio frequency field which then was turned off adiabatically. Another experiment performed by ten Wolde *et al.* (Noordam *et al.* 1989) studied the time evolution of this Runge–Lenz vector in the presence of a weak, static external electric field in rubidium.

An exciting goal for future experimental work is preparation of a minimum uncertainty wave packet localized in both the radial and angular coordinates (Alber and Zoller 1991, Ellinger *et al.* 1991). This is a quantum state with minimum fluctuations according to the Heinsenberg uncertainty principle. Such a Rydberg wave packet will move along a Kepler ellipse and would come closest to a 'classical atom', still compatible of course with the laws of quantum mechanics.

Another interesting aspect of Rydberg wave packets concerns time-dependent studies of electronic dynamics in systems which are not integrable classically and exhibit classical chaos (Friedrich and Wintgen 1989). An example of such a system which has recently received considerable attention both experimentally and theoretically is the hydrogen atom in a weak external, static magnetic field close to the photoionization threshold (Friedrich and Wintgen 1989). Frequency resolved absorption spectra of this system exhibit an intriguing complexity of spectral lines in this energy regime. In such a system time-resolved studies of the electronic dynamics gives valuable complementary information on the behavior of the system. Figure 4 shows the theoretical prediction for the transition probability in a pump-probe scheme, where the first laser pulse excites the Rydberg

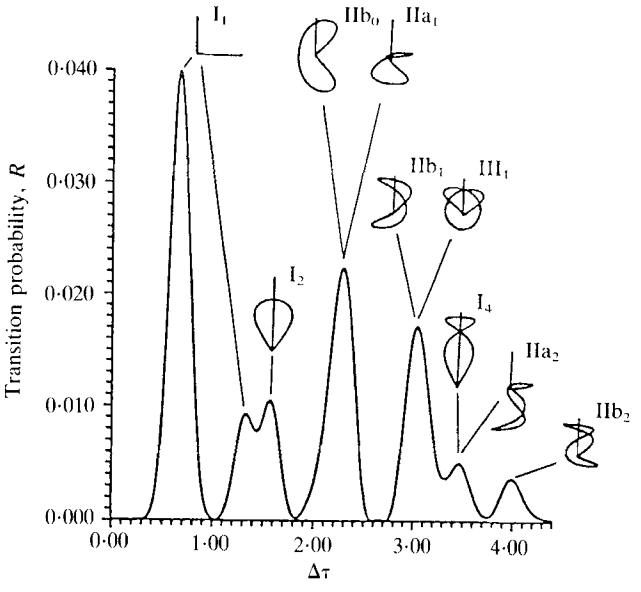

Figure 4. Rydberg atom in magnetic field: the transition probability for a Raman process as a function of the time delay between the two laser pulses. The peaks are associated with closed orbits where the wave packet returns to the ion core. The magnetic field is $B = 0{\cdot}47$ T. Time is measured in units of the Larmour precession time $T = 76$ ps.

wave packet and the second pulse de-excites the electron to some low-lying atomic state (induced Raman process) as a function of the time delay for a Rydberg wave packet which has been prepared in a sodium atom in the presence of a static magnetic field (Alber 1989). Each peak corresponds to the return of a fraction of the excited wave packet to the ionic core region along one of the *closed orbits* which are indicated on top of the each recombination peak. Wave packet experiments in these systems still remain to be done.

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