Coherence and Decay of Rydberg Wave Packets

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Calculations are presented that show the response of an atom to a picosecond laser pulse which resonantly excites a manifold of Rydberg states. The coherent atomic state that is produced is of the form of a spatially localized wave packet. The motion, decay, and reformation of the wave packet are described and related to the complicated quantum beat pattern that appears in the subsequent spontaneous decay.

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The finite duration of a picosecond laser pulse produces a frequency bandwidth that corresponds to a spectral width of approximately 20 cm^{-1}. When such a pulse is used to excite an atom to high-lying Rydberg levels (principal quantum number $n > 30$), many levels will be simultaneously and coherently excited. If the spatial distribution of the electron wave function at the end of the pulse is calculated, it is found that the short pulse has excited a well-defined wave packet. With the proper choice of pulse length and laser frequency it is possible to generate a minimum-uncertainty packet that oscillates and radiates with many of the characteristics of a classical electron in a Kepler orbit.

Rydberg wave packets offer a particularly interesting example of quantum beats. The rapid time variation of the spontaneous decay can be interpreted in terms of the motion and spreading of the packet. Consequently, the classical motion of the wave packet, as well as the quantum-mechanical spreading, disintegration, and revival of the wave packet, may be studied experimentally by measurement of the quantum beat signal.

Rydberg atomic states have already proven to be a rich source of experiments which expand our understanding of fundamental quantum mechanics. Haroche and co-workers[1] and Meschede, Walther, and Müller[2] have investigated the interaction of a single atom with the electromagnetic field in a high-Q cavity. Kleppner has shown that when Rydberg atoms are placed in a cavity spontaneous emission may be suppressed.[3] Interesting new types of quantum states have been investigated by Clark, Korevaar, and Littman,[4] Cooke and Freeman *et al.*,[5] and Bayfield and Pinnaduwage.[6] As we show below, it is possible, with use of picosecond excitation of Rydberg states, to investigate some of the fundamental questions of electron wave packets and spontaneous emission posed by Schrödinger sixty years ago.[7]

Laser excitation of the Rydberg states is easily described by a semiclassical model in which the interaction-picture amplitude of the ground state is denoted by $a_g(t)$, and the amplitudes of the various excited states by $a_n(t)$. These amplitudes satisfy the equations

$$\dot{a}_g = -\tfrac{1}{2} i \sum_n \Omega_n a_n(t) f(t) \exp(-i\Delta_n t), \tag{1a}$$

$$\dot{a}_n = -\tfrac{1}{2} i \Omega_n a_g(t) f(t) \exp(i\Delta_n t), \tag{1b}$$

where the laser field has a pulse envelope $f(t)$ and the center frequency of the laser is detuned by an amount Δ_n from the transition frequency from the ground state $|g\rangle$ to the Rydberg state $|n\rangle$. We have made the rotating-wave approximation. The coefficients Ω_n are just the Rabi frequencies of the various transitions. For simplicity in our examples we will use the hydrogenic dipole moments and energy-level spacings, but the results are easily extended to more complex atoms. Similarly, for convenience, we will assume that the ground state is an s state.

The Eqs. (1) are easily numerically integrated for a particular case. After investigating a number of examples, we have found that particularly interesting results are obtained when we use a 6–10-psec (FWHM intensity) pulse tuned so that its center frequency resonantly excites $n = 85$. We will denote this average n as $\bar{n}$. This pulse will appreciably excite five to ten levels about $n = 85$. This range of states appreciably excited is denoted Δn. We can then safely limit the sum over Rydberg states in Eq. (1) to $60 \leq n \leq 110$, and regard states $|g\rangle$ through $|n = 59\rangle$ as possible final states for the spontaneous decay following excitation. After the pulse the interaction-picture amplitudes will be constant except for a very slow decay due to spontaneous emission. To a good approximation the time evolution will be just the oscillatory free evolution. The wave function for the Rydberg electron can then be written as

$$\Psi_R(r,t) = \sum_n a_n(t) \exp(-i\omega_n t) u_n(r), \tag{2}$$

where ω_n are the various transition frequencies from the ground state, and $u_n(r)$ are the hydrogenic radial wave functions. Figure 1(a) shows the evolution of the wave packet $r^2|\Psi_R(r,t)|^2$ after the laser pulse. A well-defined wave packet is formed; it moves out to the classical turning point where it narrows. If we cal-

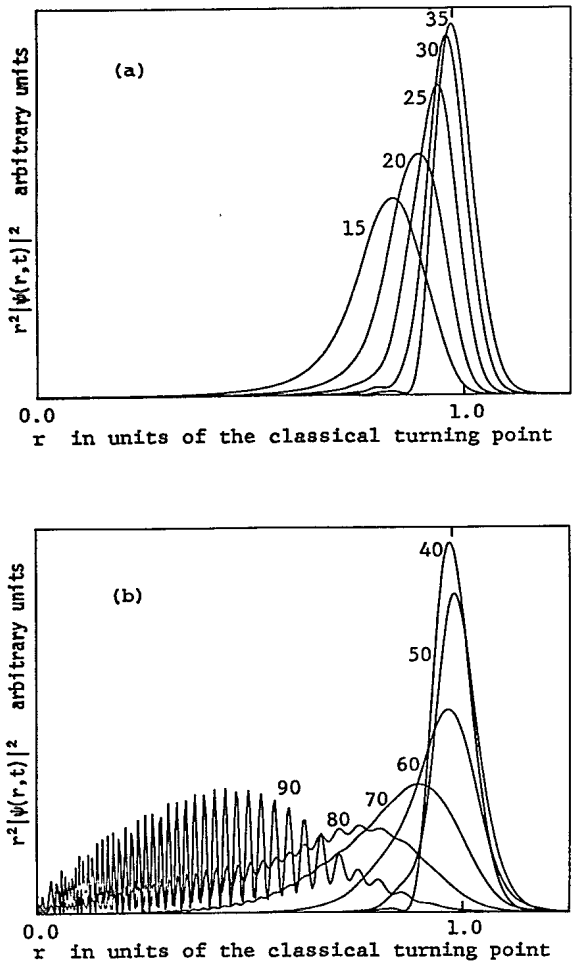

FIG. 1. Rydberg wave packet during its first orbit. In each curve the packet is labeled by the time that has elapsed, in picoseconds, from the center of the laser pulse. A 10-psec pulse (FWHM) was used to excite Rydberg states in the vicinity of $n=85$. The wave packet during the first half of the orbit is shown in (a); in (b) it is shown during the second half.

culate the uncertainty product for the wave packet near the turning point we find that $\Delta r\,\Delta p = 0.53\hbar$. This is only a few percent larger than the minimum allowed by the Heisenberg uncertainty principle. This value for the uncertainty product is calculated with the assumption that the laser pulse envelope is a Gaussian. If instead we take a hyperbolic secant envelope, the product is increased only to $\Delta r\,\Delta p = 0.56\hbar$.

After reaching the turning point, the wave packet reverses its direction and accelerates back toward the nucleus. This is shown in Fig. 1(b). As the packet approaches the nucleus it is dispersed by the strong Coulomb potential. This cycle is then repeated as the wave packet envelops the nucleus and proceeds back toward the classical turning point again. The period of this cycle is just the classical period of an electron in a Kepler orbit with energy corresponding to that of an electron in $|n=85\rangle$. In this case the classical period is 93.4 psec. After a few orbits the wave packet disperses, although as we will see, it later reforms.

If, in our example, we had chosen a longer pulse and tuned the laser so as to produce a larger value of $\bar{n}$, but with Δn unchanged, the wave packet would orbit even more times before dispersing. The reason for this limiting behavior is that the energy levels become nearly equally spaced similar to those of a harmonic oscillator.

Rather than continuing to look at the wave packet for longer times, we will concentrate on a closely related quantity which is more experimentally accessible—the total power radiated by the atom. The radiation after the laser pulse is due entirely to spontaneous emission. Quantum electrodynamics gives a simple analog to the classical Larmor formula:

$$P(t)=\frac{4e^2}{3c^3}\langle\Psi|:\ddot{\mathbf{r}}(t)\cdot\ddot{\mathbf{r}}(t):|\Psi\rangle, \tag{3}$$

where P is the expectation of the power radiated by the atom, Ψ is the atomic wave function at the end of the laser pulse, and $\ddot{\mathbf{r}}$ is the vector acceleration operator for the electron.[8] The colons denote normal ordering.[9]

If the power is time averaged over intervals long compared to a classical orbital period then it is found that the average radiated power decays exponentially as though only the single eigenstate $|\bar{n}\rangle$ had been excited. The spontaneous-decay lifetime of atom, then, is given by the reciprocal of the Einstein A coefficient for the level $|\bar{n}\rangle$. The ratio of the orbital period to the lifetime is of the order of α^3, independent of $\bar{n}$. The exponential decay is negligible in the examples discussed here, so that to a good approximation $\ddot{\mathbf{r}}$ is simply the Coulomb force on the electron divided by the mass of the electron. Finally, the expression for the power may be written

$$P(t)=\frac{4e^2}{3c^3}\left\langle\Psi_R(t)\left|\frac{e^2\mathbf{r}}{mr^3}\cdot\frac{e^2\mathbf{r}}{mr^3}\right|\Psi_R(t)\right\rangle. \tag{4}$$

Here we have explicitly written the acceleration in terms of the Coulomb force, and have put the time dependence back into the wave function. The effect of the normal ordering is to replace Ψ with Ψ_R, as defined in Eq. (2). Thus the radiative power goes as the expectation value on $\Psi_R(t)$ of the square of the acceleration, which peaks sharply when the packet overlaps regions of small r.

In Fig. 2 we have plotted this expectation value of the radiated power for an interval of 3 nsec which is approximately 32 of the 93-psec classical orbital periods. We see that there are three well-defined peaks in the emitted power as the wave packet produces a quantum beat once each classical period when

it passes the perigee of the orbit. After these three beats the packet breaks up, producing a much more complicated beat. After about 2.6 nsec the beat commences again as the wave packet reforms. The complex pattern which occurs between the fundamental-beat recurrences is seen to contain quantum beats at twice, three times, and four times the fundamental beat frequency.

All of this can be understood rather simply. If Eq. (1b) is formally integrated and substituted into Eqs. (2) and (4) it follows that the radiated power is proportional to the absolute square of the quantity

$$\Phi(t) = (1/2\pi)\sum_n \tfrac{1}{2}(\Delta_{n+1}-\Delta_{n-1})\exp(-i\Delta_n t)F(\Delta_n). \tag{5}$$

Here we have denoted by $F(\Delta_n)$ the Fourier transform of the product of the pulse envelope and the amplitude of the ground state. The evolution of the ground state during the pulse is a simple depletion due to pumping. In deriving this expression we have assumed that Δn, the range of states appreciably excited, is much smaller than the average principal quantum number $\bar{n}$. In the example of Fig. 2, $\Delta n = 6$ (FWHM). When Δn is this small in comparison with n it is useful to make a Taylor series expansion of Δn,

$$\Delta_n = \Delta + (n-\bar{n})\Delta' + (1/2!)(n-\bar{n})^2\Delta'' + \ldots, \tag{6}$$

where Δ, Δ', . . . are just Δ_n and its derivatives evaluated at $n = \bar{n}$. If we substitute this expansion into Eq. (5) we find

$$\Phi(t) \simeq (1/2\pi)\Delta' \exp[i(\Delta - \bar{n}\Delta')t]\sum_n \exp(in\Delta' t)F(\Delta_n). \tag{7}$$

Ignoring the initial phase factor, we have a Fourier series that repeats with a fundamental period $T_0 = 2\pi/\Delta'$, exactly the classical orbital period. Furthermore, the Fourier series is approximately the inverse of the Fourier transform $F(\Delta_n)$. So the periodic peaks in $\Phi(t)$ are to a good approximation the same shapes as $f(t)a_g(t)$, i.e., approximately the same as the original laser pulse envelope. The spreading of the packet is determined by the next term in the Taylor series. The packet will reform when this spreading term has no effect, i.e., when $\Delta'' t = 2\pi$. Long-term revivals in the coherence of the atom may be predicted similarly. For example, at $t = \bar{n}^2 T_0$ the second through fourth terms of the Taylor series for $\Delta_n t$ are integer multiples of π; at $t = \bar{n}^3 T_0$ the second through the fifth terms of the series are integer multiples of π. Figure 3 shows the quantum beats at $\bar{n}^3 T_0 = 57\,352.00$ nsec.

As the excitation pulse is shortened from the picosecond regime to the femtosecond regime, we again find that a well-formed Rydberg wave packet is generated. The large bandwidth of these short pulses results in a broad distribution of excited states. In the case of pulses of a few femtoseconds, the distribution of excited states runs from $n = 3$ to well into the continuum of unbound states. In this case $\Delta n > \bar{n}$ and the Taylor series of Eq. (6) does not converge. The resulting Rydberg wave packets do not oscillate, but disintegrate long before reaching the classical turning point. The wave packet of width Δr has associated with it a spread in momentum Δp whose lower bound is given by the uncertainty principle. Narrow wave packets have a large spread in momentum, which is the reason for their rapid disintegration.[10] Figure 4

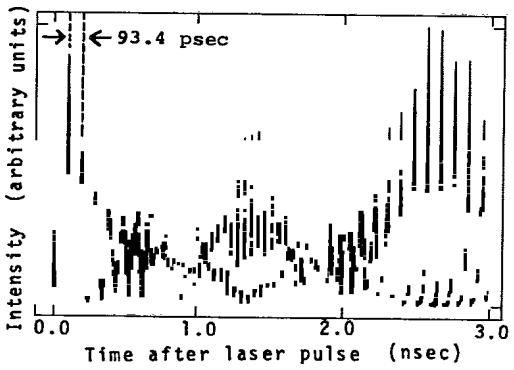

FIG. 2. Intensity of spontaneous radiation. The intensity is modulated by quantum beats. The excitation is as in Fig. 1.

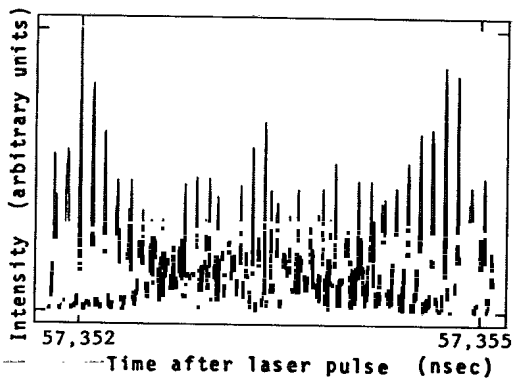

FIG. 3. Approximate revival in the quantum beat pattern. The revival occurs at $\bar{n}^3 T_0 = 57\,352$ nsec after the initial excitation of the atom which is described in Fig. 1. T_0 is the electron's classical orbital period.

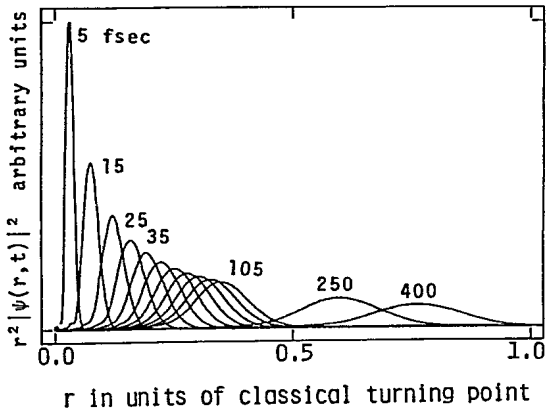

FIG. 4. Rydberg wave packets generated by pulses of various lengths. Here $\bar{n}=25$ and the packets are labeled by the pulse lengths (FWHM) in femtoseconds. The packets are as they appear toward the end of the pulse.

shows the wave packets as they appear toward the end of pulses of various lengths. In the numerical generation of these wave packets, we used hydrogenic eigenfunctions to $n=60$, and used the WKB method to generate as many as 1000 continuum eigenfunctions. Without the continuum states, the packets would be severely deformed. The population excited to these continuum states at the end of the pulse will not remain bound to the nucleus. We have not included multiphoton ionization in these calculations. As the pulse becomes intense enough to invert the hydrogen atom in a period shorter than 5 fs multiphoton ionization becomes significant, and intensities approach those at which above-threshold ionization is believed possible.[11] In the case of alkali-metal atoms for which the excitation frequency is smaller, multiphoton effects will occur at even lower intensities, and correspondingly longer pulses must be used to prevent ionization.

Although these packets do not oscillate, a classical correspondence may be found in the position of the peak of the packet toward the end of the pulse. The position is that of a classical electron that has been abruptly, at $t=0$, given an energy equal to that of the state $|\bar{n}\rangle$, and then allowed to travel freely up the potential well from a starting position near the nucleus. The time $t=0$ is the time at which the center of the excitation pulse reaches the atom.

Classical Kepler mechanics and the quantum theory of atoms have always looked very different, but Rydberg electron wave packets appear to be a way to produce quantum states which behave in many ways like classical states. The time scales and laser parameters make the area attractive for experimental investigation.

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