Controlling the Angular Momentum Composition of a Rydberg Electron Wave Packet

J. R. R. Verlet, V. G. Stavros, R. S. Minns, and H. H. Fielding
Department of Chemistry, King's College London, Strand, London WC2R 2LS, United Kingdom
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Sequences of phase-locked laser pulses have been employed to control the orbital angular momentum character of an electron wave packet, which is initially created from a superposition of *s* and *d* Rydberg series. By an intelligent choice of phase, which depends on the excitation energy and the quantum defects, we are able to selectively pump down either all or a fraction of one or other angular momentum component, and by employing multichannel quantum-defect theory we are able to analyze the quantum-state distribution in detail.

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Since the first observations of electron wave packets in atoms [1], and more recently in molecules [2], there has been an increasing interest in controlling the dynamics of these simple systems. Molecular dynamics [3] and even biological systems [4] can be controlled by optimizing laser field parameters such as phase, amplitude, and intensity of the spectral components in a self-learning feedback loop [5]. However, the resulting pulse shapes can be extremely complex and much of the underlying physics cannot be extracted due to this complexity. One is therefore tempted to revisit some of the more simple systems, such as electron wave packets in atoms, where one can fully determine the quantum interfering paths and wave packet dynamics. For example, pulse-shaping techniques have enabled the excitation of arbitrary wave packets [6] and their full amplitude and phase characterization [7]. More intuitive control schemes such as excitation using a train of phase-locked optical pulses [8] has led to the creation of Schrödinger's paradoxical cat [9], and the demonstration of Young's double slit experiment in an atom [10]. This latter experiment is of particular relevance, as ultimately coherent control is based on this simple concept. In this Letter, we extend some of the above-mentioned ideas and explore the dynamics of a wave packet composed of two angular momenta. We demonstrate full control over the time-dependent populations of the different angular momentum states in the superposition using an intuitive sequence of phase-locked pulses.

Consider a wave packet composed of a superposition of two Rydberg series converging to the same ionization limit but with different orbital angular momenta, for example *ns* and *nd* Rydberg states excited from a low-lying *p* state. As a consequence of their difference in angular momentum, they can be expected to have different quantum defects. Such a wave packet may be regarded as being composed of two separate components, both of which are time dependent. After some time, the pathways connecting the two angular components, i.e., the coupling between the initial *p* state and the Rydberg states in the two series, may be expected to be phase mismatched. In this case a second optical pulse could be chosen to be in phase with one component, thus interfering constructively, and out of phase with the other, thus interfering destructively. We can quantify the dynamics of the two components by expressing each wave packet as $\Psi(r, t) = \sum_{nl} a_{nl}\psi_{nl}(r)\exp(-i\omega_{nl}t)$, where $\psi_{nl}(r)$ is the radial wave function of the eigenstate $|nl\rangle$, $\omega_{nl} = -1/2(n - \mu_l)^2$ is its energy and a_{nl} is its amplitude in the superposition. Expanding the energy term as a Taylor's series in $\delta n = n - \bar{n}$ gives rise to a term $\exp[2\pi i(t/t_{cl})\mu_l]$ showing that the wave packet is phase shifted by $2\pi k\mu_l$ after k periods. When there are two Rydberg series, this gives rise to an angular beat with period $t_\mu = t_{cl}/\Delta\mu_l$ [11]. As an example, suppose that the transition probability to each series is the same and that the difference in the quantum defect is $\Delta\mu = 0.5$, the two series will be perfectly interleaved and the energy spacing between adjacent Rydberg states will be half that of a single series. From a frequency perspective, the spacing has halved and so the orbit period will be twice that of a single series. From a time-dependent perspective, the initial return to the core is suppressed as the two Rydberg series differ in quantum defect by half and thus are out of phase at t_{cl}. The wave packet will be observed to return at $t_\mu = t_{cl}/\Delta\mu_l = 2t_{cl}$. As the phases of the two components are mismatched at t_{cl}, a second wave packet launched at this time allows one to select a relative phase so that the interference is constructive with one angular momentum component of the first wave packet but destructive with the other angular momentum component. Removing one of the angular momentum components from the wave packet has the effect of filtering out the oscillation in the angular coordinate to leave only the radial motion.

We demonstrate these ideas in the Xe atom. The experimental setup will be described in detail elsewhere [12] but briefly, Rydberg series converging to the lower spin-orbit ionization limit $Xe^{+2}P_{3/2}$ are excited using a resonantly enhanced two-nanosecond plus one-picosecond excitation process. The narrow bandwidth output of a frequency mixed dye laser (250 nm) is tuned to excite the $6p[1/2]_0$ intermediate state using

two photons. The broad bandwidth ($\sim 15\ \mathrm{cm}^{-1}$) output of an amplified picosecond dye laser (568 nm) then excites a superposition of $nd[1/2]_1$, $nd[3/2]_1$, and $nd[3/2]_1$ series (with a ratio of approximately 1:100:20).

The dynamics of the wave packet are monitored using the optical Ramsey method [13]. The return of the wave packet to its original state after a time $t = \tau$ (i.e., its autocorrelation function) is monitored by exciting two identical, but delayed Rydberg wave packets using a pair of phase-locked picosecond laser pulses and observing interference patterns in the Rydberg state population. For the coherent control experiments a sequence of three phase-locked pulses is required, the first two are the excitation and control pulses, while the third is essentially the detection pulse. The stabilized three-arm interferometer will be described in detail elsewhere [12]. The autocorrelation function is determined by calculating the root-mean-square of the amplitude of the fringes in the Rydberg population, which is measured by applying a delayed (150 ns) pulsed $1.5\ \mathrm{kV\,cm}^{-1}$ electric field to direct field-ionized Xe^+ through a field free time-of-flight tube onto a multichannel plate detector.

We fully characterize the dynamics of this simple control scheme using a modified autocorrelation function where transition amplitudes and energies are determined from 5-channel quantum-defect theory (MQDT):

$$\langle \Psi(0)|\Psi(t)\rangle = \sum_{nl} g(\omega_{nl}) f_{nl}[e^{-i\omega_{nl}t} + e^{-i\omega_{nl}(t+\Delta t+\phi)}], \tag{1}$$

where, $f_{nl} = 2(E_{nl} - E_0)|\sum_\alpha D_\alpha A_\alpha^{(n)}|^2/N^2$ is the transition oscillator strength to state $0|nl\rangle$, and is calculated using the usual MQDT equations for discrete Rydberg levels [14], and $g(\omega_{nl})$ is the excitation profile of the laser field which we assumed to be Gaussian. The 5-channel MQDT comprises $nd[1/2]_1$, $nd[3/2]_1$, and $ns[3/2]_1$ converging to the lower $^2P_{3/2}$ ionization limit and $ns'[1/2]_1$ and $nd'[3/2]_1$ converging to the upper $^2P_{3/2}$ ionization limit. The first term in the square brackets describes the temporal evolution of the first wave packet which is launched at $t = 0$. The second term in square brackets describes the evolution of the second wave packet, which drives the interference between the different angular momentum components and is launched with a delay of $t_2 = \Delta t + \phi$ after the first. ϕ depends on the optical frequency and is discussed below. We may further characterize the dynamics of the interfering wave packets by considering the time-dependent Rydberg state populations following the pulse sequence, which will be described below.

First we observe the dynamics of a single wave packet excited around $n = 30$ (lower trace in Fig. 1) with the corresponding calculated recurrence spectrum (upper trace in Fig. 1). At first sight, the single wave packet spectrum is remarkably hydrogenic. Peaks are observed

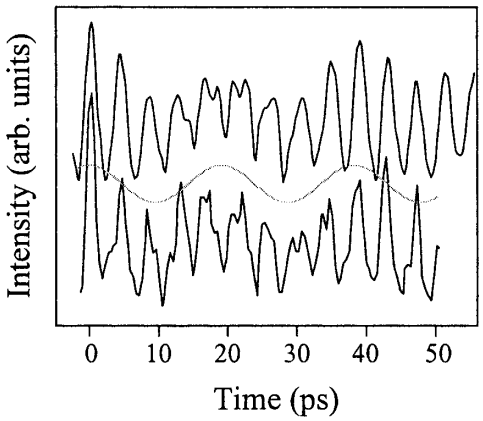

FIG. 1. Dynamics of a single wave packet centered at $n = 30$. The lower trace is the experimental recurrence spectrum and is recorded using the optical Ramsey method. The upper trace is calculated using 5-channel MQDT to determine the transition and amplitudes of the Rydberg states in the superposition. The cosine highlights the angular beat (see text for details).

at multiples of the classical period ($t_{cl} = 2\pi n^3 = 4.1$ ps). The wave packet then disperses owing to the anharmonicity of the Coulomb potential with the appearance of a second order partial revival around 20.5 ps and then a full revival around 41 ps. Superimposed on the hydrogenic spectrum is a slow modulation with a period of about 19 ps. This is the angular beat between the more intense $nd[3/2]_1$ and $ns[3/2]_1$ Rydberg series, $\Delta\mu_l = 0.22$. The first minimum of this slow modulation corresponds to the time when these two angular momentum components of the wave packet are out of phase with one another and occurs at $t_2 = (t_\mu/2) = \frac{1}{2}(t_{cl}/0.22) \approx 2t_{cl}$.

The phase-shift between the two Rydberg series is now exploited in the control scheme. We launch a second wave packet with a delay such that the first wave packet has made two full orbits, coinciding with the minimum in the angular beat. We search for a suitable phase such that the ion signal corresponds to destructive interference with the s states and an enhancement of the d states. The dynamics of the resultant wave packet are monitored following interference with a third phase-locked pulse (the optical Ramsey method) and the results are presented in the lower trace of Fig. 2(a). We then flip the phase, reversing the interference so that we are in phase with the s states and reduce the d-state population, and this spectrum is presented in the lower trace of Fig. 2(b). Note that in this case, rather than observing a wave packet of pure s character the angular oscillation has been enhanced. This occurs because the s and d components are not exactly π out of phase. We can, however, enhance the s/d ratio further from approximately 1:5 to about 4:1. We have explored this feature in detail by investigating the populations of the Rydberg states in the superposition following the pulse pair.

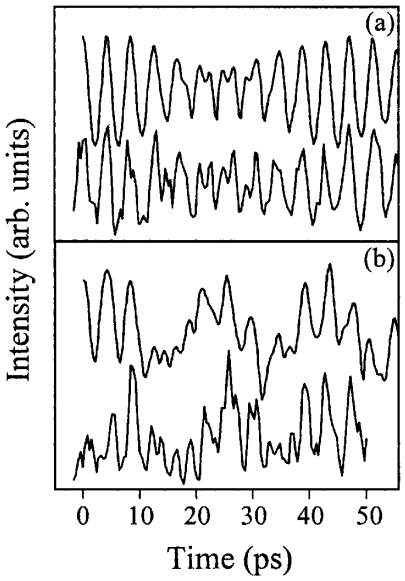

FIG. 2. Wave packet dynamics following a two-pulse sequence with a third pulse as a detection pulse excited under similar conditions as the spectra in Fig. 2. The second pulse is launched at twice the classical period plus a phase shift to enhance the d states but to interfere destructively with the s states (a). Flipping the phase by π reverses the interference to be in-phase with the s states but to reduce the d-state population (b). In each panel the lower trace is the experimental spectrum and the upper trace is the calculation.

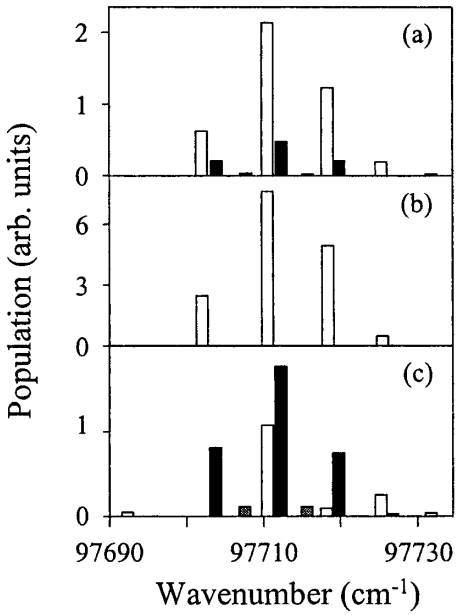

FIG. 3. Calculated populations of the relevant Rydberg states in the superposition following single pulse excitation (a), and two-pulse excitation to either enhance the s states and deplete the d states (b) or vice versa (c). In all panels $ns[1/2]_1$ states are white, $nd[3/2]_1$ are black, and, for completeness, $nd[1/2]_1$ states are gray.

We calculate the multipulse response of the time-dependent populations $|a_{nl}(t)|^2$ [13,15], assuming that the intensity of the laser pulses is sufficiently low that the ground-state amplitude remains virtually unchanged by the pulses. Using the approach employed to determine the population distribution in Schrödinger cat states [15], the state population following a train of two pulses may be written as

$$a_{nl} = -\frac{i}{2}\Omega_{nl} g(\omega_{nl})[1 + e^{i\Delta t_2} e^{i(\omega_n - \omega_{\bar{n}})t_2}], \qquad (2)$$

where Ω_{nl} is the Rabi frequency for the excitation between the intermediate state and a given Rydberg state and contains the transition dipoles calculated using MQDT. Δ is defined $\Delta = \omega_{\bar{n}} - \omega_0$, ω_{nl} is the energy of the Rydberg state excited, ω_0 is the energy of the intermediate state, and $\omega_{\bar{n}}$ is the average energy of the excitation. The population distribution following excitation of a single wave packet is shown in Fig. 3(a). The origin of the angular beat is clear: Radial wave packets are formed from states of the same color and the angular beat is between the white and black states. We now launch a second wave packet at $t_2 = 8.2$ ps and optimize the phase by inspecting the Ramsey fringes of the separate angular momentum components. We then generate the Rydberg state distribution of the wave packet using Eq. (2). In Fig. 3(b) we have pumped down the s states leaving only d states—this corresponds to the wave packet spectrum presented in Fig. 2(a). In Fig. 3(c) we enhance the s/d ratio leaving the two series with similar intensities. This population distribution enhances the angular beat and generates the time-resolved spectrum presented in Fig. 2(b).

The efficiency of this control experiment is ultimately limited by both the system and dispersion. One could find a more suitable system where the phase mismatch is perfect after one or two orbits. Unfortunately, dispersion will limit one from looking at longer times as the paths connecting the individual states of the separate components will become phase shifted.

In conclusion, we have demonstrated how the use of pulse sequencing techniques can be applied to control the angular momentum composition of a Rydberg electron wave packet. Using this approach, we have shown in Xe that a wave packet composed of a mixture of both s- and d-angular momentum states can be modified to remove the s component and leave a wave packet that is localized only in the radial coordinate. Alternatively, reversing the optical phase allows us to enhance the s/d ratio to amplify the angular oscillation. These ideas are very intuitive suggesting that coherent control studies in simple atomic systems help to improve our understanding of the link between optical phase and quantum mechanical phase which will in turn lead to ways of engineering wave packets in more complex systems.

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