Laser Excitation of Radial-Angular Rydberg Wave Packets

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We demonstrate that the excitation of a manifold of Rydberg states by an appropriately short pulse and the angular momentum redistribution by a subsequent properly chosen pulse can produce a radial and angular wave packet. The angular localization results from transitions between the initially excited $\{n, l\}$ manifold and other (higher) l states caused by the second pulse. The geometry of the resulting wave packet is illustrated by specific calculations for metastable $(2s)$ hydrogen as the initial state.

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Although the excitation of radial Rydberg wave packets in atoms is well established and studied [1], angular wave packets—and ideally the combination of both—remain a challenge. It is normally expected that the mixing of angular momenta requires the presence of an additional external field and wave packets of Rydberg atoms in external electric and/or magnetic fields have indeed been studied [2]. We hardly need belabor here the significance of Rydberg wave packets, not only as the link between classical and quantum physics, but also as a fundamental phenomenon of relevance in many contexts [3,4]. But a relatively simple way of creating the proper radial and angular superposition of states has not been discovered as yet.

A scheme for angular wave packets has been proposed by Corless and Stroud [5]. Beginning with the excitation of a single nl Rydberg state—which would of course require a long pulse—they argued on the basis of a Landau-Zener model that, given the large dipole moments (in length gauge) between high-n states, radiation of optical frequency and sufficient intensity will mix the initial nl state with other angular momentum states within the same n manifold. The mixing is to be accomplished by the same pulse that induces the excitation. To the best of our knowledge, the idea has not been implemented experimentally, nor has it been confirmed by more elaborate calculations. Among other issues, mixing l states only within the same n manifold by an optical field, without mixing other n manifolds at the same time, seems to us rather problematic.

It is our purpose in this paper to propose a different scheme, demonstrating at the same time its quantitative validity under realistic conditions. The idea is to begin with the excitation of a radial Rydberg wave packet by a pulse of center frequency and Fourier bandwidth such as to form the desired superposition of Rydberg states around some value $\bar{n}$ of the principal quantum number. Then, with some delay τ to be specified later on, another pulse of longer wavelength is launched. We propose to prove that with the proper choice of wavelength, intensity, and duration of the second pulse, the initial wave packet is changed into one that contains a superposition of several angular momenta, in addition to the superposition of principal quantum numbers, forming thus a wave packet localized radially as well as angularly.

To pin matters down to a full extent, we have chosen to prove the point by working with a well defined scenario, namely, beginning with the $2s$ state of hydrogen as the initial one. There are two main reasons for this choice. First, in hydrogen the calculation can be as accurate as necessary. Second, the wavelength necessary for the excitation of the radial wave packet is in the optical range, which makes our predictions applicable to an experimentally easier context, namely, the excitation of a wave packet by single-photon absorption from the ground state of an alkali atom such as Rb, for example. And, of course, an experiment with $H(2s)$ as the initial state, although more inconvenient, is not out of the question.

With $2s$ as the initial state, we assume a pulse of peak intensity $I_1 = 10^5$ W/cm^2 and frequency ω_1 centered around $\hbar\omega_1 = 3.38$ eV which corresponds to $\bar{n} = 30$. The duration of this pulse is taken to be $\Delta t_1 = T_{\text{Kepler}}/5 = 0.82$ ps with $T_{\text{Kepler}} = 2\pi\bar{n}^3$ (in atomic units). Thus we create a radial Rydberg wave packet in the np series. The second laser pulse is taken to have peak intensity $I_2 = 3 \times 10^9$ W/cm^2 and duration $\Delta t_2 = 2T_{\text{Kepler}}/5 = 1.64$ ps. Its frequency ω_2 is assumed to be such that $\hbar\omega_2 = 0.12$ eV corresponding to the CO_2 laser. The delay $\tau = 4.0$ ps between the two pulses has been chosen so that the maximum of the second pulse is reached when the radial distribution of the initial wave packet is at the inner turning point for the first time after the first laser pulse, ensuring thus more efficient coupling to the mixing pulse than if the wave packet were further away.

Technically, the problem turned out to be much more demanding than one might expect at first sight; certainly more so than we anticipated. Thus we adopted a nonperturbative approach to the solution of the time-dependent Schrödinger equation (TDSE) formulated in terms of the

expansion of the time-dependent wave function on a discretized basis for the radial parts of the eigenstates of the bare atom. Decomposing the eigenstates $\Phi_{n,l,m}(r,\theta,\phi)$ of the atom in spherical harmonics $Y_{l,m}(\theta,\phi)$ and radial functions $\chi_{n,l}(r)$ we have

$$\Phi_{n,l,m}(r,\theta,\phi) = \frac{\chi_{n,l}(r)}{r} Y_{l,m}(\theta,\phi) \tag{1}$$

with the standard notation for the quantum numbers (n,l,m). The radial functions and the corresponding energies $E_{n,l}$ are determined through diagonalization of the atomic Hamiltonian on the finite interval $[0, r_{\max}]$ with the fixed boundary conditions $\chi_{n,l}(0) = \chi_{n,l}(r_{\max}) = 0$. For the numerical implementation of the diagonalization, the functions $\chi_{n,l}(r)$ are expanded on a set of B splines of order k constructed in the interval $[0, r_{\max}]$, where the boundary conditions are satisfied automatically by omitting those B splines which have a nonzero value at the boundaries of the interval. The total number of B splines $N_{\rm BS}$ used for the expansion is equal to the number of basis wave functions obtained through diagonalization. The radial part of the dipole moment in velocity gauge $\mu_{n,l,m}^{n',l',m'}$ between the basis wave functions $\Phi_{n,l,m}$ and $\Phi_{n',l',m'}$ is calculated by numerical integration whereas the angular part of $\mu_{n,l,m}^{n',l',m'}$ is known analytically [6]. It is a fundamental feature of this approach that beyond a certain value of n, the states represent a discretized basis for the continuum. Further details concerning the construction of the basis sets and their properties can be found elsewhere [7].

The wave function of the wave packet at any time t is expanded as

$$\Psi_{\rm WP}(r,\theta,\phi,t) = \sum_{n=1}^{N_{\rm BS}} \sum_{l=0}^{n-1} c_{n,l}(t) \Phi_{n,l,0}(r,\theta,\phi). \tag{2}$$

Note that, since the initial state $2s$ has a magnetic quantum number $m = 0$ and the laser pulses are assumed to be linearly polarized along the z axis, $m = 0$ for all components of the wave packet, if the dipole approximation is valid, which it is. The amplitudes $c_{n,l}(t)$ are determined by integrating the TDSE, where the atom-field interactions are described in the velocity gauge, which is computationally much more economical. The envelopes of the amplitudes of the electric fields, which enter the TDSE, are given by

$$f_1(t) = f_{1,\max} \cos^2\!\left(\frac{\pi t}{2.75\Delta t_1}\right) \quad \text{for } t_a := -1.375\Delta t_1 \le t \le 1.375\Delta t_1 =: t_b\,, \tag{3a}$$

$$f_2(t) = f_{2,\max} \cos^2\!\left(\frac{\pi(t-\tau)}{2.75\Delta t_2}\right) \quad \text{for } t_c := \tau - 1.375\Delta t_2 \le t \le \tau + 1.375\Delta t_2 =: t_d\,, \tag{3b}$$

where $f_{j,\max}$ denote the maximum electric field strengths, τ is the delay between the maxima of the two laser pulses, and Δt_j is the full width at half maximum (FWHM) of the intensities $I_j(t) = f_j^2(t)/4$ (in atomic units) (always $j = 1, 2$). The above form of the temporal pulse profiles, which for all practical purposes is a good approximation to the slightly more realistic Gaussian pulse shape, has been chosen in order to have finite limits t_a, t_b, t_c, and t_d leading to unambiguous integration times.

If the two pulses are nonoverlapping, i.e., if $t_c \ge t_b$, the solution of the TDSE with the initial condition $c_{2,0}(t_a) = 1$ can be accomplished in three steps. First, an analytical solution for the amplitudes $c_{n,l}(t)$ is obtained for $t_a \le t \le t_c$, if the exciting laser pulse f_1 is sufficiently weak not to deplete the initial state $2s$ significantly [8], as is always the case in this context. In the rotating wave approximation, which is valid in this first step, the expressions are

$$c_{2,0}(t_c) = \exp\!\left(-\frac{i}{\hbar} E_{2,0}(t_c - t_a)\right), \tag{4a}$$

$$c_{n,1}(t_c) = -\frac{i}{2} \mu_{2,0,0}^{n,1,0} \exp\!\left(-\frac{i}{\hbar} E_{n,1}(t_c - t_a)\right) \int_{t_a}^{t_b} dt' \frac{f_1(t')}{\omega_1} \exp\!\left(-\frac{i}{\hbar}(E_{n,1} - E_{2,0} - \omega_1)t'\right), \tag{4b}$$

$$c_{n,l}(t_c) = 0 \qquad \text{(otherwise)}\,, \tag{4c}$$

which then serve as initial conditions for the numerical integration of the system of differential equations

$$i\hbar \dot{c}_{n,l}(t) = E_{n,l} c_{n,l}(t) + \frac{f_2(t)}{\omega_2} \cos\omega_2 t \sum_{0 \le l' = l \pm 1} \sum_{n' = l'+1}^{N_{\rm BS}} \mu_{n,l,0}^{n',l',0} c_{n',l'}(t) \tag{5}$$

performed for $t_c < t \le t_d$. Finally, for $t > t_d$ no fields are present and the amplitudes evolve freely according to

$$c_{n,l}(t) = c_{n,l}(t_d) \exp\!\left(-\frac{i}{\hbar} E_{n,l}(t - t_d)\right). \tag{6}$$

With the amplitudes $c_{n,l}(t)$ calculated, analysis of $\Psi_{\rm WP}(t)$, given by Eq. (2), provides the spatial distribution of the created wave packet.

The propagation of the TDSE in the interval $t_c < t \le t_d$ is the most demanding and crucial step. Speed of the

numerical integration and convergence dictate the size of the basis set. First, a sufficiently large energy E_{max} is chosen and only basis functions with energy $E_{n,l} \leq E_{max}$ are included. Second, the angular momentum quantum numbers are restricted to $l \leq l_{max}$ with l_{max} sufficiently large. The parameters E_{max}, l_{max}, r_{max}, and N_{BS}, which determine the basis set used, have to be checked for convergence. The most basic criterion for r_{max} is to be sufficiently large to provide enough bound states building up the desired wave packet after its excitation. In other words, r_{max} has to be at least as large as the spatial extent of the wave packet. With the minimum value of r_{max} thus established, the amplitudes $c_{n,l}(t)$ have to be calculated repeatedly, with increasing r_{max}, N_{BS}, E_{max}, and l_{max}, until the populations in the bound states (after both pulses) remain constant upon further increase of the parameters. Since photoionization of the bound states and couplings through the continuum are not necessarily negligible, convergence in the continuum (positive energy) states has to be checked as well. A particularly valuable test in this respect is the convergence of a number of above threshold ionization (ATI) peaks, since the ATI spectrum is a quantity most sensitive to the basis parameters r_{max} and N_{BS}; even though ATI is of no relevance to this problem. However, this ensures that the influence of the continuum has been taken into account completely.

The parameters actually used for the results shown below are $k = 9$, $r_{max} = 5000$ a.u. $= 265$ nm, $N_{BS} = 500$, $E_{max} = 0.05$ a.u. $= 1.35$ eV, and $l_{max} = 9$. They fulfill all of the convergence tests described above. Note that the relatively small size of r_{max} in relation to the wavelength λ_2 of the second pulse (r_{max} is only 2.5% of $\lambda_2 = 10\,640$ nm) provides further confirmation of the validity of the dipole approximation for the second pulse.

Figure 1 shows the sum of populations of all Rydberg states with principal quantum number n [part (a)] or with angular momentum quantum number l [part (b)]. The full histograms refer to the radial-angular Rydberg wave packet after the second pulse, whereas the dashed histogram shows the population distribution in n of the purely radial Rydberg wave packet excited by the first pulse. The population distribution in n for both the radial and the radial-angular wave packet is localized around $\bar{n} = 30$. For the radial-angular wave packet the width of the n distribution is slightly larger than for the radial wave packet because of the spectral width of the second laser pulse. In addition, the population of states around $\bar{n} = 30$ is smaller in the case of the radial-angular wave packet, because population has been redistributed among the bound states (see $n = 6$ and $n = 10$) and also ionized to the continuum. Quantitatively, the population of the Rydberg wave packet after the first laser pulse is 2.3×10^{-6} whereas it has decayed to 4.8×10^{-7} after the second laser pulse. The population distribution in l clearly demonstrates the redistribution of population among states with higher odd angular momentum quantum numbers l:

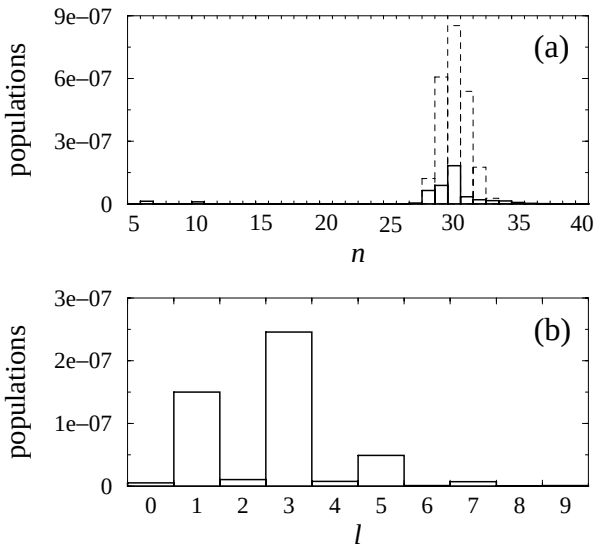

FIG. 1. Sum of populations of all Rydberg states with principal quantum number n (a) or angular momentum quantum number l (b). The dashed histogram refers to the radial wave packet after the first pulse whereas the full histograms refer to the radial-angular wave packet after both pulses.

Instead of having population only in states with $l = 1$, as excited by the first pulse, after the second pulse 68% of the population is found in states with $l > 1$. The dominance of odd values of l results from the Raman transitions responsible for the redistribution. (In other words, $l = 3$, for example, is populated via a transition $l - 2$ to $l - 1$ to l, and so on.) The minuscule presence of $l = 0, 2, 4$ reflects the tiny populations in states $n = 6$ and $n = 10$ induced by the second pulse. The effect of the l mixing on the spatial distribution of the wave packet is demonstrated in Fig. 2, where the probability distribution is plotted directly at the end of the second laser pulse when the wave packet happens to be roughly at its outer turning radius. Since, as noted above, the magnetic quantum number $m = 0$, the wave packet is rotationally symmetric with respect to the laser polarization axis. Thus, the probability distribution on the x-z plane, shown in Fig. 2, provides complete information about the wave packet. Clearly, the radial-angular wave packet deviates significantly from a radial one which contains only states with $l = 1$; it is much more localized along the z axis.

On the basis of the experience gained in the course of the calculations, it is our judgment that with somewhat higher intensity and longer duration for the second pulse, an even larger distribution of angular momenta can be populated producing thus further angular localization. Since this would require a longer computational time, without any qualitative change in the basic results, we have chosen not to invest the time at this point. That would certainly be a worthwhile undertaking if an experiment were in sight. As indicated earlier, we have worked in the velocity gauge which has been proven [9] to be

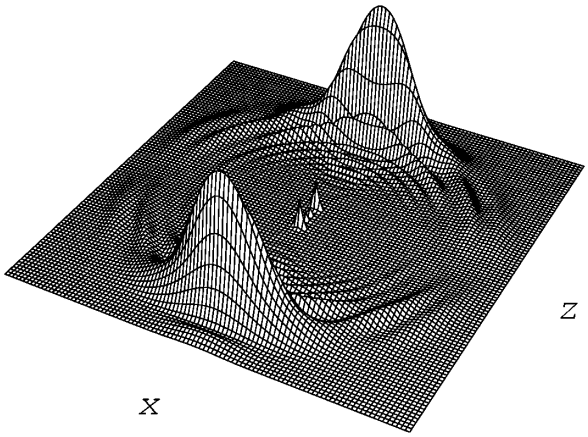

FIG. 2. Probability distribution of the wave packet on the x-z plane (z is the laser polarization direction).

much less demanding in the number of angular momenta necessary for convergence. As dictated by gauge invariance, the results should be identical in the length gauge, but depending on the method and the problem, the necessary computational time may be impractically long.

It is also our judgment that angular localization within only a single n manifold by an optical pulse, as envisioned by Corless and Stroud in [5], is not feasible. First of all, even if somehow one could begin with a single nl state, the mixing pulse would redistribute population among other n states. Our judgment is based on extensive calculations within the dipole approximation that we have performed in the course of this work—which began with the intention to extend the results of Ref. [5]. Second, each time, after having ensured convergence, we ended up with essentially no mixing of angular momenta at all in the case of optical frequencies. It is only at very high intensities ($>10^{15}$ W/cm^2) that we noticed any mixing worth speaking of. But these are intensities several orders of magnitude larger than those envisioned in Ref. [5] with all the complications of strong field interactions. An additional issue complicating the problem further is the complete breakdown of the low order multipole expansion for Rydberg states (say, around $n = 30$) and optical wavelengths [10]. This much more serious issue makes a calculation in the dipole approximation almost irrelevant and suggests that the large dipole moments between states within the same n manifold, invoked in Ref. [5], cannot guarantee angular momentum mixing at optical wavelengths. It must be stressed, on the other hand, that unless a calculation including all multipoles is performed, the question will remain open.

Our scheme, which relies on a much longer wavelength for the mixing pulse, circumvents both of the above difficulties. It also allows the contemplation of further extension of the mixing. If we chose a somewhat shorter wavelength, we could then introduce a sufficient admixture of the quadrupole coupling to produce mixing of the m values as well. This would lead to further angular localization by breaking the symmetry around the z axis. It is thus reasonable to expect that, through the judicious selection of intensity, wavelength, and pulse duration, considerable radial and angular localization of Rydberg wave packets can be achieved.

Finally, our results have, we believe, implications beyond the particular issue of radial-angular wave packets examined here. They show that predictions on the redistribution of population among Rydberg states by laser pulses have to be approached with great caution if they are based on calculations with restricted bases.

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