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DYNAMICS OF POPULATIONS IN HYDROGEN ATOM UNDER THE ACTION OF ULTRA-SHORT LASER PULSE

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We present the results of the research of population dynamics in a hydrogen atom driven by a ultra-short laser pulse. The possibility to produce a wave packet of Rydberg states with high values of orbital and magnetic quantum numbers is studied numerically depending on the pulse duration and intensity. To describe the atom-field interaction three models were compared, namely, the one with discrete spectrum states only, the one with resonance bound states and continuum, and the one with non-resonance discrete spectrum states having high values of orbital and magnetic quantum numbers also taken into account. The wave packet formation is simulated by direct numerical solution of the Schrödinger equation using a large number of basis states both of discrete and continuous spectrum. The continuous spectrum is approximated by a large finite number of states separated by a small wavenumber interval. The role of continuum is shown to be principal in the population dynamics and the wave packed formation. Figs. 9. Ref.: 13 titles.

Key words: hydrogen-like atom, Rydberg states, continuum states, wave packet, laser excitation, ultra-short pulse, population dynamics, numerical modeling.

As a result of superposition of Rydberg states in a hydrogen-like atom with large values of quantum numbers in all degrees of freedom, the formation of a localized quasistable wave packet, moving along a classical Kepler orbit at a great distance from the nucleus (thousands of atomic units), is possible. Such states represent the classical limit of atom, and their dynamics exhibits both classical and quantum properties. For such wave packets the uncertainty product of coordinate and momentum is near to minimal, therefore, they are a good approximation of coherent states.

The possibility to create wave packets with various values of orbital and magnetic quantum numbers and different degree of localization was confirmed in numerous publications and has attracted attention of many researchers. Various ways of theoretical description of such localized states [1] and methods of their experimental production [2-6] have been proposed.

The development of new approaches to the localized wave packets production and the analysis of their properties are of interest for studying the correspondence principle in the classical limit of atom, for understanding the relation between the orbits of classically chaotic systems and the motion of a quantum wave packet, and for quantum control of the behavior of Rydberg electron [7].

The formation of Rydberg states superposition can be attained via the interaction of atom in the ground state with a short strong laser pulse [8-11]. In this case a large number of Rydberg energy levels around the central resonance energy level is populated simultaneously.

Since for dipole transitions the selection rule $\Delta l = \pm 1$ is valid, and in the ground state $l = 0$, such a packet inevitably includes the Rydberg states with low values of orbital and magnetic quantum numbers. As shown in [8-11], it is localized in radial variable and oscillates between the classical turning points. It is possible that intense ultra-short pulses can produce packets of high-*L* states due to non-resonance transitions between the closely-spaced excited states and the transitions via the continuum states [12].

In this paper the possibility to create a wave packet of Rydberg states of a hydrogen atom with various values of orbital and magnetic quantum numbers using an ultra-short high-intensity laser pulse is studied by means of direct numerical solution of the Schrödinger equation. The key point is that the transitions between the bound and continuum states are taken into account. By means of numerical modeling, the population dynamics and the shape of the resulting wave packet are investigated.

1. Excitation of transitions by a laser pulse. In several papers [8-11], as well as in our earlier work the process of atom interaction with laser pulse was considered on the basis of the simplified model without taking continuum states into account. Since the Rydberg energy levels are close to the ionization limit, and the considered pulses possess high power and small duration (and, hence, large spectral width), such account is obviously necessary.

First we consider the model without continuum states, in which transitions occur only between the ground state and Rydberg states $n = 20 \div 40$. Consider the field to be circularly polarized, then the

Rydberg levels with $l = m = 1$ will be populated via one photon transitions. The model wave function of the system can be written as

$$\Psi(\mathbf{r},t) = \sum_{n=20}^{40} C_{n1}(t) R_{n1} Y_{11} + C_{10}(t) R_{10} Y_{00}\,, \quad (1)$$

where $C_{10}(t)$ and $C_{n1}(t)$ are the probability amplitudes of the ground state and Rydberg states; R_{01} and R_{n1} are the radial eigenfunctions of the ground and Rydberg states; Y_{00} и Y_{11} are the spherical harmonics.

Consider the pulse envelope function to have the Gaussian shape, so that the electric field strength is given by

$$E(t) = E_0 \exp[-(2t/T)^2]\sin(\omega t)\,,$$

where T is the pulse duration, E_0 is the pulse amplitude, ω is its carrier frequency. In the example below the latter was chosen to be resonant to the transition from the ground state to the Rydberg state with n = 25.

The numerical solution of the set of differential equations for probability amplitudes has shown, that choosing properly the pulse power and duration, it is possible to reach almost full transfer of atoms from the ground state to the Rydberg states. The population dynamics in such a model is presented in fig. 1.

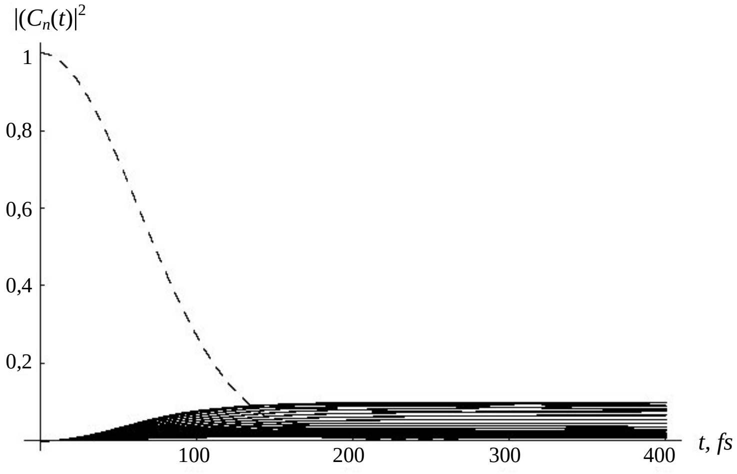

Fig. 1. Population dynamics in the simplified model of atom-light interaction. The ground state population is shown by the dashed line, the populations of Rydberg states by solid lines. The laser field amplitude is $5{,}1\cdot10^7$ *V/cm*, the pulse duration is 600 *fs*

Fig. 1 demonstrates the depopulation of the ground state and distribution of atoms among numerous excited energy levels with $n = 20 \div 40$ and $l = m = 1$.

Simultaneous excitation of the large number of excited states near to ionization limit indicates that it is necessary to consider the transitions to the states of continuous spectrum. Hence, we proceed to a model, including both bound states and a part of continuum adjacent to the ionization limit. As it is commonly done [12], we use the discrete representation of the continuum by a sequence of 40 states with equidistant values of wave number k, starting from $k = 0$, with the step $\Delta k = 0{,}02$ a. u., covering the range of energies from 0 to 0,3 a. u. The wave function of the system can be presented as

$$\Psi(\mathbf{r},t) = \sum_{n=20}^{40} C_{n1}(t) R_{n1} Y_{11} + \\ + \sum_{k} C_{k1}(t) R_{k1} Y_{11} + C_{10}(t) R_{10} Y_{00}\,, \quad (2)$$

where $C_{n1}(t)$ and $C_{k1}(t)$ are the probability amplitude of the corresponding Rydberg states and continuum states.

The numerical computations have shown that the dynamics of populations in the system with the wave function (2) significantly differs from that of the simplified model (1) for the same laser pulse parameters.

In figs 2 and 3 the time dependence of the total population of continuum states and the population of the ground states are plotted versus the time. Approximately a half of all atoms are transferred to the continuum, and a half stays in the ground state. Population oscillations are similar to Rabi oscillations, their high frequency being due to very large field amplitude. The analysis has shown that in weaker fields this frequency decreases. The total population of discrete highly-excited states appears to be insignificant (about 0,2 %). Recall that the pulse parameters are chosen so that in the first simplified model almost all atoms are transferred to Rydberg states.

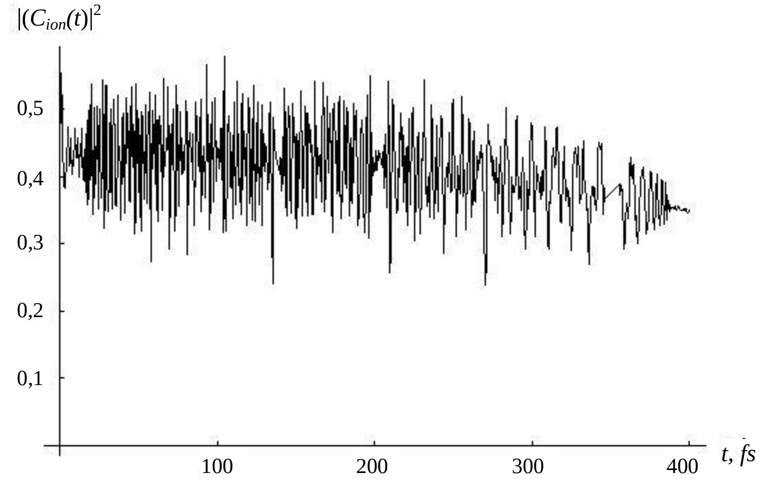

Fig. 2. The total population of the continuum states versus time in the second model. $E_0 = 5{,}1 \cdot 10^7$ *V/cm*, the pulse duration is 600 *fs*

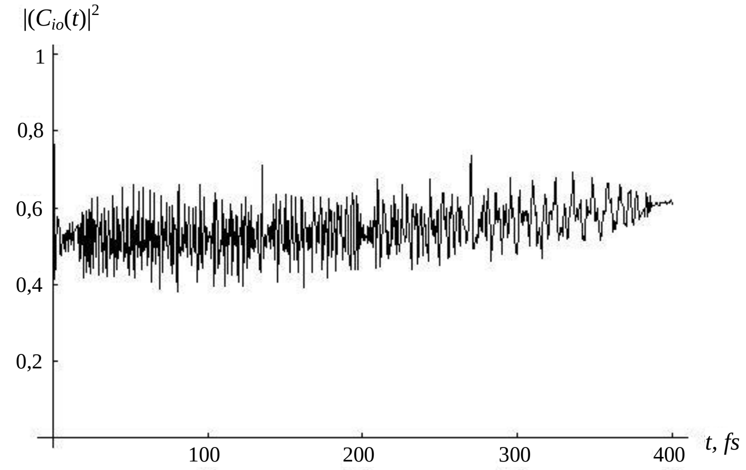

Fig. 3. The population of the ground state versus time in the second model. $E_0 = 5{,}1 \cdot 10^7$ *V/cm*, the pulse duration is 600 *fs*

With the continuum taken into account such field amplitude appear to be too large, and instead of the Rydberg states population the single-photon ionization dominates. The analysis has shown that the most intense Rydberg states population occurs at essentially smaller field amplitudes (fig. 4). In this most favorable case their total population reaches about 1 %.

One can conclude that the simplified model without continuous spectrum is not valid for the description of Rydberg states population in a real atom.

2. Populating the states with high values of orbital and magnetic quantum numbers. The next step in improving our model is connected with including the greater number of states with various values n, l and m into consideration. Let l take the values from 1 to 10 for each continuum energy level. Thus, 400 basis functions will be used for the continuum description. Let us also include the states with the principal quantum number n from 3 to 40 and the orbital quantum number l from 1 to 10 into the basis set of discrete spectrum. The general number of basis states of the discrete spectrum will thus be increased to 328.

The external field is again supposed to be circularly polarized, so that the transitions corresponding to $\Delta l = 1$, $\Delta m = 1$ selection rules are allowed. Therefore, in the considered model for all states $l = m$. The frequencies of the transitions between the low-excited states of the discrete spectrum and the continuum are far enough from the laser carrier frequency, however, as the analysis has shown, they significantly contribute to the atom-field interaction due to rather high values of their dipole moments. Moreover, their presence in the model provides the possibility of cascade processes with the increase of the orbital quantum number and magnetic quantum number by 1 at each step. The total scheme of transitions included in the model is presented in fig. 5.

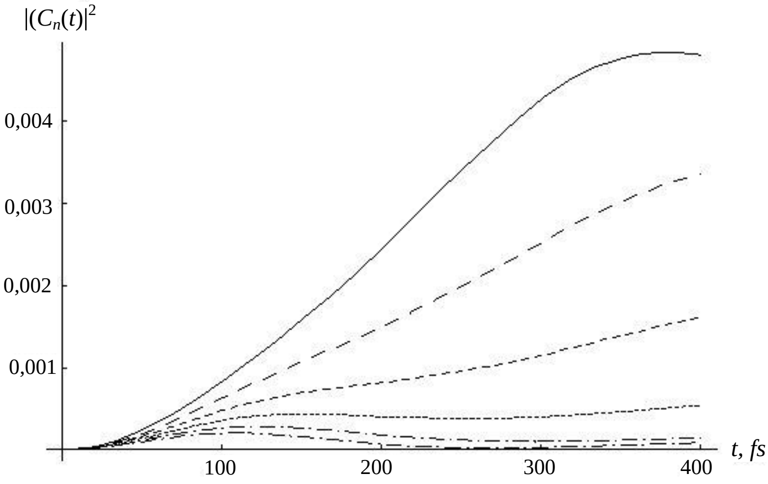

Fig. 4. The populations of Rydberg states with $n = 20 \div 25$ versus time in the second model for the optimal conditions, namely, weaker pulse $E_0 = 4{,}6 \cdot 10^6$ *V/cm* and the pulse duration 600 *fs*

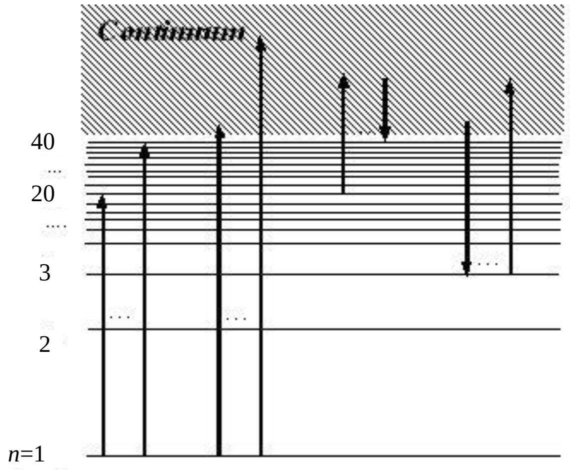

Fig. 5. Scheme of transitions in the model supposing the excitation of the states with high values of angular momentum and its projection under the action of a laser pulse with circular polarization

Thus, the final state of the atom is now presented as a superposition of the states of discrete spectrum and those of the continuum, the total number of basis functions being 728:

$$\Psi(\mathbf{r},t) = \sum_n \sum_{l=m} C_{nl}(t) R_{nl} Y_{l,m=l} + \\ + \sum_k \sum_{l=m} C_{kl}(t) R_{kl} Y_{l,m=l}, \quad (3)$$

The first sum in *Eq* (3) presents the discrete states, the second one includes the continuum states. For the investigation of dynamics of populations the following set of equations for the probability amplitudes was solved numerically:

$$\frac{dC_{10}(t)}{dt} = i\sum_{n=20}^{40} d_{10}^{n1}\exp[i(\tfrac{1}{n^2}-\tfrac{1}{2})t]E(t)C_{n1}(t) + i\sum_k d_{10}^{k1}\exp[i(-E_k-\tfrac{1}{2})t]E(t)C_{k1}(t), \tag{4}$$

$$\frac{dC_{n1}(t)}{dt} = i d_{10}^{n1}\exp[-i(\tfrac{1}{n^2}-\tfrac{1}{2})t]E(t)C_{10}(t) + i\sum_k d_{n1}^{k2}\exp[-i(-\tfrac{1}{n^2}-E_k)t]E(t)C_{k2}(t), \tag{5}$$

$$\begin{aligned}\frac{dC_{nl}(t)}{dt} &= i\sum_k d_{nl}^{kl-1}\exp[-i(-\tfrac{1}{n^2}-E_k)t]E(t)C_{kl-1}(t) + \\ &+ i\sum_k d_{nl}^{kl+1}\exp[i(-\tfrac{1}{n^2}-E_k)t]E(t)C_{kl+1}(t),\end{aligned} \tag{6}$$

$$\begin{aligned}\frac{dC_{kl}(t)}{dt} &= i\sum_k d_{nl-1}^{kl}\exp[-i(-\tfrac{1}{n^2}-E_k)t]E(t)C_{nl-1}(t) + \\ &+ i\sum_k d_{nl+1}^{kl}\exp[i(-\tfrac{1}{n^2}-E_k)t]E(t)C_{nl+1}(t),\end{aligned} \tag{7}$$

where $d_{nl}^{kl'}$ are the dipole moment matrix elements of the transitions, E_k is the energy of the continuum state for given k. The dipole moment matrix elements for the transitions between the discrete states and continuum states were found by numerical integration, and for the transitions between discrete states - according to well-known formulas.

The investigation of population dynamics was carried out for different pulse durations. The amplitude of the laser field was 3,57·10^8 *V/cm*. At the moment of turning the field on the atoms are supposed to be in the ground state with $n=1$, $l=0$, $m=0$. The action of the pulse with the duration 5 *fs* is represented in figs. 6 and 7.

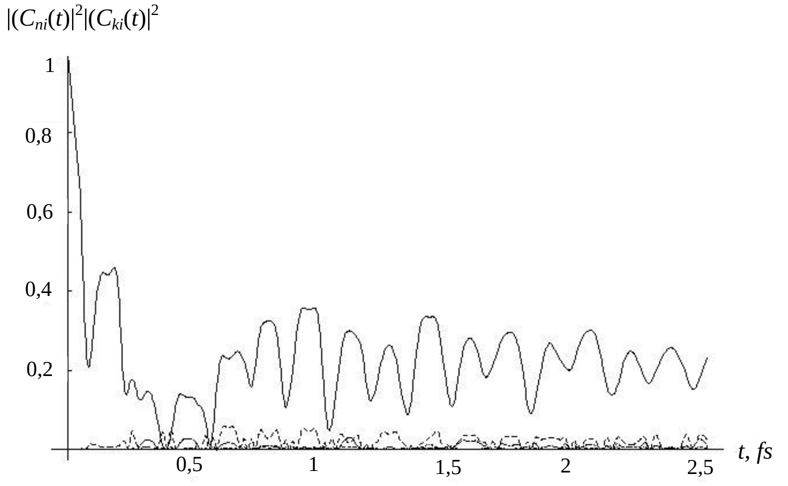

Fig. 6. The population dynamics under the action of a pulse with duration 5 *fs* and the field amplitude E_0=3,57·10^8 *V/cm*. The solid line represents the population of the ground state, the dashed lines – that of four continuum states with the energies closest to zero

Fig. 6 represents the time dependence of the population of the ground state and four continuum states with energies close to zero. One can see that a significant part of atoms goes from the ground state to the continuum states. Then the sequence of up an down transitions between the continuum and discrete levels with various values of n occurs, each of them increasing the orbital and magnetic quantum numbers by 1. Such a process is illustrated by fig. 7, where the population of the levels with $n=3\div7$, $l=2$ is plotted versus time. The oscillations associated with transitions from continuum and back are well seen. At the initial stage the level with $n=3$ is populated maximally, then the population is redistributed between other levels due to transitions via the continuum. The state with $l=m=2$ is populated significantly, that agrees with the results obtained by other authors [12]. A significant part of atoms stays in the ground state.

Similar dynamics of population is observed for a laser pulse by duration 25 *fs*. One can notice that for longer time of the pulse action a greater number of transitions to the continuum and back has time to occur, and, therefore, the states with higher values of the orbital and magnetic quantum numbers are populated. In the end of the pulse action approximately 30 % of atoms are transferred to the continuum, 13 % remain in the ground state. Other atoms are redistributed between the bound states with various values of n, l and m. The population distribution over l is presented in fig. 8.

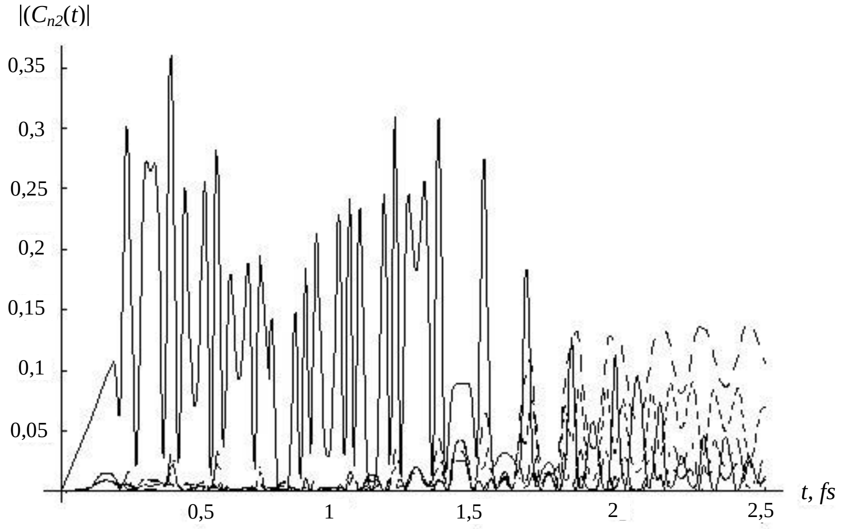

Fig. 7. Population of eigenstates with l=2: solid line - n = 3, dashed line - n = 4, 5, 6, 7. E_0 =3,57·10^8 *V/cm*

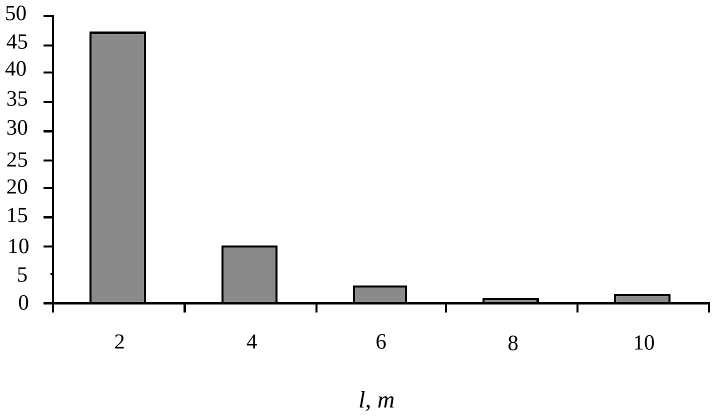

Fig. 8. Population distribution over l and m

In fig. 8 one can find essential contribution of states with high values of orbital and magnetic quantum numbers to the general population of the wave packet formed as result of the pulse action. It is possible to note, that states with odd values of l and m (1, 3, 5, 7, 9) are practically not populated. It can be explained by preservation of parity [13]. Basing on the amplitudes $C_{nl}\,($ the spatial distribution of the probability density is readily found. In fig. 9 such a distribution is shown in the plane $z = 0$. One can see the localization of electron in the radial variable and no localization in the angular variable, which, as mentioned above, is typical for a superposition with prevalence of states with low values of orbital and magnetic quantum numbers.

In conclusion we would like to note that the simplified model and the model with account of continuum yield results that are principally different. Without account of additional discrete levels no acceptable population of Rydberg states can be obtained. At the same time, the essential part of population is transferred to the continuum. The more exact computations using the extended model containing the extended set of discrete states, as well as the states of continuum with various values of orbital and magnetic quantum numbers proved itself to be capable of describing the transfer of population to the states with higher orbital quantum number. The increase of population of states with higher values of orbital and magnetic quantum numbers with increasing pulse duration is observed. It is possible to assume, that the optimization of pulse duration and amplitude allows to reach high values l and m up to the maximal ones. Presumably even-photon mechanism of population of discrete levels via the continuum is confirmed.

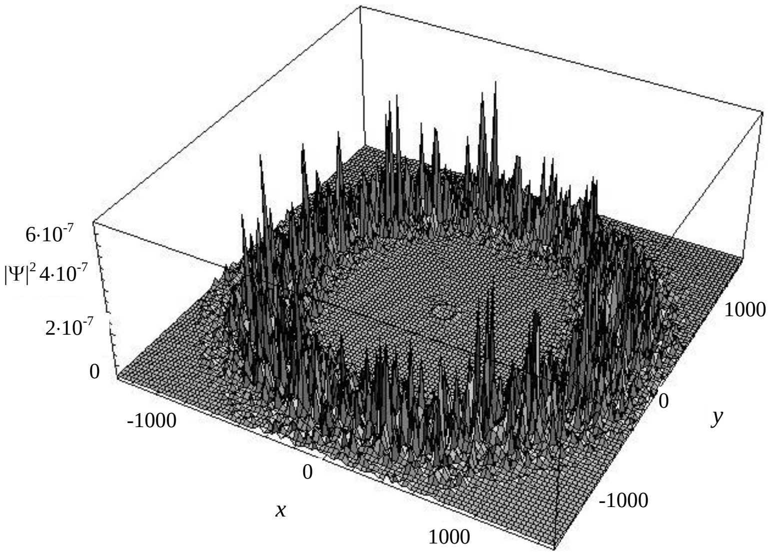

Fig. 9. Distribution of probability density for wave packet formed by pulse with circular polarization with the duration 25 *fs*

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ДИНАМИКА ЗАСЕЛЕННОСТЕЙ В АТОМЕ ВОДОРОДА ПОД ДЕЙСТВИЕМ УЛЬТРАКОРОТКОГО ИМПУЛЬСА ЛАЗЕРНОГО ИЗЛУЧЕНИЯ

В. Л. Дербов, Н. И. Тепер

Представлены результаты исследования динамики заселенностей в атоме водорода под действием ультракороткого лазерного импульса. Численно изучается возможность получения волновых пакетов ридберговских состояний с большими значениями орбитального и магнитного квантовых чисел в зависимости от продолжительности и интенсивности импульса. Для того чтобы описать взаимодействие атома и поля, сравнивались три модели, а именно: модель, включающая только дискретный спектр, модель с резонансными состояниями и непрерывным спектром и модель, учитывающая также нерезонансные состояния дискретного спектра, имеющие большие значения орбитального и магнитного квантовых чисел. Формирование волнового пакета моделируется прямым численным решением уравнения Шрёдингера, используя большое число базисных состояний как дискретного, так и непрерывного спектра. Непрерывный спектр аппроксимируется большим конечным числом состояний, разделенных малыми интервалами волнового числа. Показано, что роль непрерывного спектра является принципиальной в динамике заселенностей и формирования волнового пакета.

Ключевые слова: водородоподобный атом, ридберговские состояния, состояния непрерывного спектра, волновой пакет, лазерное возбуждение, ультракороткий импульс, динамика заселенностей, численное моделирование.

ДИНАМІКА ЗАСЕЛЕНОСТІ В АТОМІ ВОДНЮ ПІД ДІЄЮ УЛЬТРАКОРОТКОГО ІМПУЛЬСУ ЛАЗЕРНОГО ВИПРОМІНЮВАННЯ

В. Л. Дербов, Н. І. Тепер

Репрезентовані результати дослідження динаміки заселеності в атомі водню під дією ультракороткого лазерного імпульсу. Чисельно вивчається можливість одержання хвильових пакетів рідбергівських станів з великими значеннями орбітального та магнітного квантових чисел в залежності від тривалості та інтенсивності імпульсу. Для того щоб описати взаємодію атома і поля, порівнювались три моделі, а саме: модель яка, має в собі тільки дискретний спектр, модель з резонансним станом і неперервним спектром і модель, яка враховує також нерезонансні стани дискретного спектру, що мають великі значення орбітального й магнітного квантових чисел. Формування хвильового пакету моделюється прямим чисельним рішенням рівняння Шредингера, використовуючи велике число базисних станів як дискретного, так і неперервного спектру. Неперервний спектр апроксимується великим кінцевим числом станів, розділених малими інтегралами хвильового числа. Показано, що роль неперервного спектру є принциповою в динаміці заселеностей і формування хвильового пакету.

Ключові слова: воднеподібний атом, рідбергівські стани, стани неперервного спектру, хвильовий пакет, лазерне збудження, ультракороткий імпульс, динаміка заселеностей, чисельне моделювання.

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