Selective Excitation of Metastable Atomic States by Femto- and Attosecond Laser Pulses

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Abstract

In the present work the possibility to achieve high selective excitation of metastable states of atoms by interaction with short laser pulses with reasonable parameters is demonstrated theoretically. Interactions of hydrogen atom with femto- and attosecond laser pulses are studied with the use of well established close-coupling approach. The parameters of laser pulses which lead to high selective excitation of metastable state of atom together with small ionization probability are calculated with the use of different kinds of optimization procedures.

Keywords:

selective excitation, metastable state, laser control, femtosecond pulse, attosecond pulse

The nonlinear laser spectroscopy provides essentially new possibilities in nuclear, molecular and optical physics on creation and study of selectively excited states of quantum systems (see [1,2]). Development of the two-photon excitation technique [3] made it possible to obtain metastable atoms of hydrogen H ($2S_{1/2}$) in small concentrations. This, however, is enough to carry out precision optical measurements of superfine structure of 2S state [4,5] and other relativistic and radiating effects. Besides, metastable atoms and atomic ions play an important role in excitation- and charge transfer processes even in high-temperature laboratory and astrophysical plasmas (see [6]). Examples above demonstrate the importance of developing the technique for selective excitation of metastable atomic states.

Significant progress in understanding of interactions of atoms and molecules with intense electromagnetic fields results in development of theoretical and experimental methods of control of the quantum systems by laser [7,8]. One of the most natural and flexible approaches in this area is the optimal control theory (OCT). It is based on the idea that the controlling laser pulse should maximize a certain functional so that the variational principle can be used to design the pulse. The procedure leads to a set of equations for optimal laser field, which must be solved iteratively in general. In present these methods have been successfully used in molecular physics.

The main frequencies of optical transitions in the molecules, belongs to the visual and infra-red spectral bands, i.e. this frequencies can be easily generated by modern lasers. The combination of one-photon transitions is the basic mechanism of the quantum control. On the other hand the frequencies of radiational transitions between the low-lying levels of atoms can belong to the vacuum ultraviolet band. Besides the excitation of the metastable atomic states by one-photon absorption is often prohibited by the selection rules. Thus the multiphoton interaction is the basic mechanism for the selective excitation of the atomic states.

Recently [9-12] the processes of multiphoton ionization and excitation of atoms and molecules have been investigated by solving the close-coupling equations without discretization of the continuum. Such effects as the direct ionization, electron rescattering by the atomic core, resonant transitions via an intermediate discrete and continuum states, etc. are essential features of multiphoton laser-atom interactions. Accurate treatment of all these effects requires the exact solution of the quantum equations that describe the dynamics of the system. The close-coupling method is the most suitable for this kind of spectroscopic calculations.

In the present work we calculate the parameters of laser pulses which lead to high selective excitation of metastable ($2S_{1/2}$) state of hydrogen atom together with small ionization probability. The optimization procedures mentioned above are extended to be applied to multiphoton processes. Two- and four-photon selective

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Table 1 Parameters of femtosecond laser pulses with Gaussian-like envelope which leads to high selective excitation of $2S_{1/2}$ metastable state of hydrogen atom.

	Frequency [eV]	Intensity [W/cm^2]	FWHM [fs]	Final population of 2S state [%]	Ionization probability [%]
1	5,1	$1{,}0.10^{13}$	93	96,8	0,16
2	5,1	$1{,}1.10^{13}$	75	93,1	0,3
3	5,1	$1{,}5.10^{13}$	63	97,2	1,3
4	5,1	$2{,}0.10^{13}$	53	94,4	3,4
5	5,1	$3{,}1.10^{13}$	95	89,0	1,0
6	2,55	$4{,}51.10^{13}$	77	12,0	9,5
7	2,55	$4{,}56.10^{13}$	94	12,8	12,0
8	2,55	$9{,}04.10^{13}$	93	13,1	16,2
9	2,55	$1{,}20.10^{14}$	63	13,8	14,3

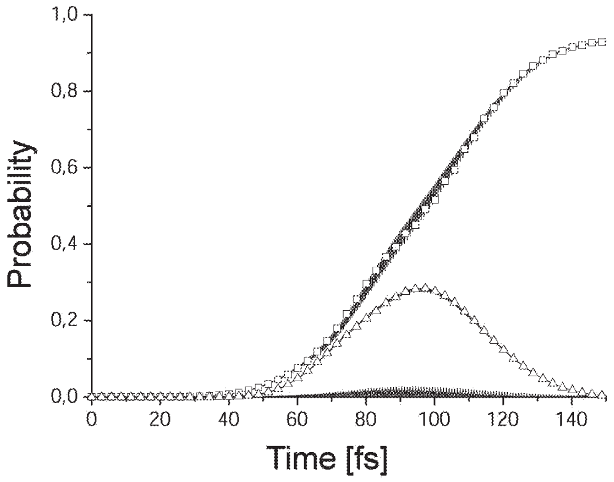

Fig. 1 Time variation of discrete and continuum states populations of hydrogen atom. Triangles: continuum population. Discrete states populations, squares: 2s; no symbol: 2p states. Parameters of femtosecond laser pulse with Gaussian envelope are: frequency 5.1 eV; intensity 1.1.10^{13} W/cm^2; FWHM 75 fs.

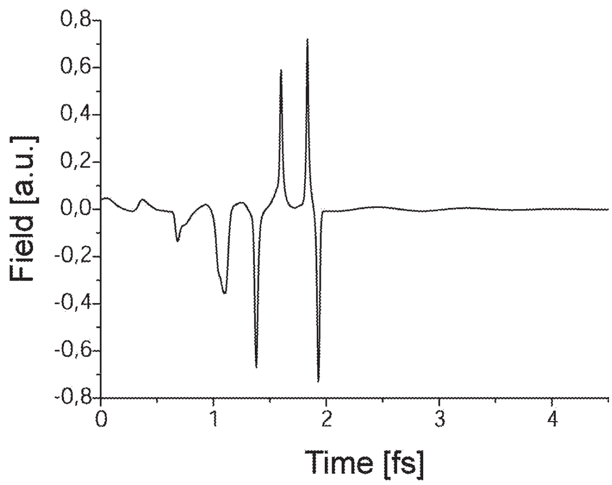

Fig. 2 Optimal field for selective excitation of 2s metastable state obtained with the use of optimization procedure without any restrictions on the laser pulse form for the total interaction duration of 4.5 fs. The final population of 2s state and ionization probability achieved are 68 % and 21 % respectively.

excitation processes are investigated for two cases: (i) the envelope of the laser pulse is fixed to be a Gaussian function and (ii) no restrictions are put on the laser pulse shape. For the interaction time of order of hundred femtoseconds the optimal laser field is found to consist of few femtosecond pulses. It is also found that high selective multiphoton excitation of metastable state with very small ionization probability can be achieved by a single femtosecond pulse with Gaussian envelope. The optimal parameters of these laser pulses are presented in Table 1. The analysis of time variation of atomic states population, like presented in Fig. 1, shows that the Raman type transitions play an important role in the process.

If interaction time reduces to few femtoseconds the optimal laser field transforms into a grid of attosecond pulses, like shown in Fig. 2. Physically this sequence of attosecond pulses affects the atomic electrons as a series of pushes. Each push displaces the atomic electron for a small distance so it can not move far from the atomic core. Thus the photoionization is suppressed during the interaction. The parameters of the sequences of attosecond laser pulses that lead to high final metastable state population are calculated. The final population of 2s metastable state and ionization probability achieved for this kind of controlling field are about 70 % and 15 % respectively.

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