An extremely robust strong-field control of atomic coherence

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Abstract: We propose and analyze a scheme for adiabatic creation of maximum coherence of 0.5 with a controllable phase between a ground state and the excited state in a model Λ-structured atom using two short laser pulses. One of the pulses has constant carrier frequency quasi-resonant with transition between an initially empty ground and the excite states. The frequency of the second pulse is swept through the resonance with the adjacent transition between the initially populated ground state and the common excited state of the atom. We demonstrate high degree of robustness of the scheme against variation of parameters of the laser radiation in relatively broad region of values. The proposed scheme may find practical applications in the field of multi-photon ionization, high-order harmonics and Raman sideband generation, as well as in nonlinear wave mixing in coherently prepared media.

OCIS codes: (020.0020) Atomic and molecular physics; (020.1670) Coherent optical effects.

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1. Introduction

Creation of coherent superposition of quantum states plays key role in numerous applications of quantum and nonlinear optics. While generation of coherence between lower (ground) states of an atomic system is an established technique (see e.g [1–4].) less attention, it appears, has been paid to robust creation of coherent superposition between the ground and excited states. The importance of the coherent preparation of atoms in the superposition of the ground state and an excited state for high-order harmonic generation and multi-photon ionization in gases was shown already more than a decade ago [5–9]. In these and similar applications, of high importance is the effective preparation of a (same) coherent superposition of the states in optically thick media. An advantage of the initial preparation of a hydrogen-like model atom in coherent superposition of the states was shown in Ref [10], and a substantial (by many orders of magnitude) enhancement of the emission rate of the high-order harmonics was demonstrated in the case of the initial coherent preparation of the atom compared with the harmonics generated in the presence of the ground state alone. The initial coherent preparation of atomic system plays an important role also in enhancement of the efficiency of multi-photon transfer of populations in the strong-field limit as it is shown in Ref [11], where application of two (or more) subsequent strong pulses instead of a single one appears to be advantageous for optimal control of the multi-photon transfer. In the proposed scheme, the first laser pulse produces initial preparation of the atomic system by creating coherent superposition of the ground and excited states for the consecutive pulse. High-order harmonic generation in He^+-like model ion is analyzed in Ref [12]. when the initial state is prepared as a coherent superposition of the ground state and an excited state and an evident advantage of using such initial preparation of the system for obtaining high conversion efficiency is demonstrated.

As it follows from the above discussion, initial preparation of an atomic system in the coherent superposition of the atomic ground state and an excited state is an important problem to be addressed for optimizing the nonlinear optical processes including multi-photon population transfer and ionization, as well as high-order harmonic generation and nonlinear mixing. If there is a collection of atoms, to obtain a coherent enhancement of a multi-photon process, all of the atoms have to be prepared in a same coherent superposition, preferably with maximum absolute value of 0.5, and with a *same* (arbitrary) *phase* of the coherence.

In this paper, we propose a scheme for creation of a coherent superposition of the ground state and an excited state in a Λ-like atomic system with the maximum value and a controllable phase of the coherence. A distinctive feature of the scheme is an extremely high robustness of the amplitude and the phase of the created coherence against variations of the parameters of the laser pulses and of the resonance conditions. The proposed scheme is based on simultaneous application to the atom of two laser pulses with durations shorter than relaxation times of the atomic system. Each of the pulses is coupled with a corresponding electric-dipole allowed transition in the Λ-like atom. One of the laser pulses acting on the transition between the initially empty ground state and the excited state has constant carrier frequency. The frequency of the second pulse is swept through the resonance with the other, initially populated transition of the Λ-atom, Fig. 1. As it can be seen, the system under consideration resembles the scheme of electromagnetically induced transparency (EIT [13]). It differs from the EIT scheme by using a relatively strong and frequency modulated (chirped) laser pulse instead of a weak probe wave with constant carrier frequency in the EIT scheme. Related papers [14, 15] consider creation of coherence between ground states of Λ-like atoms by varying the speed of the chirp of one of the pulses. The disadvantage of these schemes is their sensitivity to variations of the laser parameters due to using of non-adiabatic couplings between the dressed states.

As we show below, in the proposed scheme the value of the coherence created in the atomic system between the initially empty ground and excited states is close to the maximum

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value (of 0.5) for amplitudes, shapes and durations of the acting laser pulses varying in extremely broad range. Such robustness against variation of the laser pulse parameters makes possible efficient generation of the same (maximum) coherence between the ground and excited states in atoms of an ensemble located in different spatial points covered by the laser beams even if these beams are tightly focused. It is worth mentioning that the phase of the created coherence is same for different atoms in the ensemble. Like the amplitude of the coherence, it is not sensitive to variation of the laser parameters in extremely broad region of values.

The underlying physics of such extreme robustness of the scheme may be explained as follows. The laser pulse with constant carrier frequency acting on the transition between an initially empty ground state and the excited state produces splitting of the excited atomic level into two "dressed" states, which may be conveniently described by symmetric and anti-symmetric superposition of the initially empty ground and excited bare states. The amplitudes of these two bare states are equally weighted in both superposition states since the same laser pulse (with constant carrier frequency) performs "dressing" of the transition. The other laser pulse with chirped frequency produces complete population transfer from the initially populated ground state to the two superposition states. Due to the intrinsic feature of the chirped laser pulse, in the adiabatic regime of interaction the population of the ground state is transferred to that one among these two states, with which the resonance is reached first during the sweep of the laser pulse frequency through the resonance. Independently on which superposition state will be populated as a result of the interaction, a coherent superposition of the initially empty ground state and the excited state with maximum value of the coherence (of 0.5) will be established because the amplitudes of these bare states are equally weighted in both superposition states. A particular superposition state (the symmetric, or anti-symmetric one) will be populated depending on the sign (direction in time) of the frequency chirp. While a same (0.5) value of the coherence will be created for both positive and negative signs of the chirp, the phase of the created coherence will depend on a particular superposition state that is populated.

2. The atom-laser system and the mathematical formalism

We consider interaction of two laser pulses with a three-level atom with Λ-configuration of the working levels, Fig. 1. One of the laser pulses with a constant carrier frequency is acting on the initially empty transition of the Λ-atom. The frequency of the second frequency modulated (chirped) laser pulse coupled with the adjacent transition between the initially populated ground state and the common excited state is swept through the resonance with this transition during the action of the laser pulses.

The atomic wave function $|\psi\rangle$ may be written in the basis of the eigen-states $|n\rangle$ of the unperturbed atomic Hamiltonian as

$$|\psi\rangle = \sum_{n=1}^{3} a_n(t)|n\rangle, \tag{1}$$

where $a_n(t)$ is the complex probability amplitude of the corresponding atomic (bare) state $|n\rangle$, ($n = 1,2,3$).

The electric strength $\vec{E}$ of the laser field is given as

$$\vec{E}(t) = E_1(t)\vec{e}_1 \exp\left[i\int \omega_{L1}(t')dt'\right] + E_2(t)\vec{e}_2 \exp\left[i\omega_{L2}^0 t\right] + c.c., \tag{2}$$

where $E_1(t)$ is the amplitude of the laser pulse with the chirped frequency, and $E_2(t)$ is the amplitude of the pulse with constant carrier frequency. $\vec{e}_{1,2}$ are polarization vectors of the laser pulses. For the sake of simplicity, a linear chirp of the carrier frequency $\omega_{1L}(t) = \omega_{L1}^0 + \beta t$ is assumed for the first laser pulse, β being the speed of the chirp. The atom is supposed to be optically pumped into the state $|1\rangle$ initially.

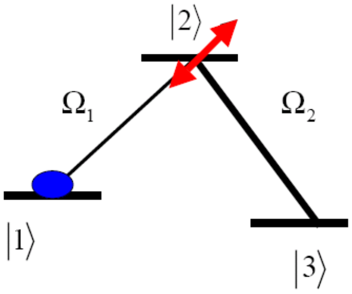

Fig. 1. Schematic of Λ -like atom with two acting laser fields: one with chirped frequency and the other one with constant carrier frequency.

In what follows, we assume that durations of the pulses are much shorter than all relaxation time constants for the atomic system. Under this assumption, evolution of the system is governed by the Schrödinger equation for the state-vector $\underline{a} = (a_1, a_2, a_3)^T$ with the interaction Hamiltonian in the rotating wave approximation,

$$\hat{H} = \hbar \begin{pmatrix} \varepsilon(t) & -\Omega_1(t) & 0 \\ -\Omega_1^*(t) & 0 & -\Omega_2^*(t) \\ 0 & -\Omega_2(t) & \Delta_0 \end{pmatrix}, \tag{3}$$

where the time-dependent detuning of the first (chirped) laser pulse is $\varepsilon(t) = \omega_{L1}(t) - \omega_{12}^0$, and the corresponding (constant in time) detuning for the second pulse is $\Delta_0 = \omega_{L2}^0 - \omega_{13}^0$ with ω_{12}^0 and ω_{13}^0 being the resonance frequencies for the corresponding electric dipole transitions in the Λ -like atom, Fig. 1. $\Omega_1(t) = d_{21}E_1(t)/\hbar$, and $\Omega_2(t) = d_{23}E_2(t)/\hbar$ are the Rabi frequencies with d_{21} and d_{23} being dipole moment elements between the excited and corresponding ground states.

It is convenient to describe the atom-laser interaction in a following new basis:

$$|b_1\rangle = |1\rangle, \quad |b_+\rangle = (|2\rangle + |3\rangle)/\sqrt{2}, \quad |b_-\rangle = (|2\rangle - |3\rangle)/\sqrt{2}, \tag{4}$$

including symmetric $|b_+\rangle$ and anti-symmetric $|b_-\rangle$ superposition states. In the new basis, the atomic wave function has the form $|\psi\rangle = b_1|b_1\rangle + b_+|b_+\rangle + b_-|b_-\rangle$ with states-amplitudes related to the amplitudes in the previous basis (1) as

$$b_1 = a_1, b_+ = (a_2 + a_3)/\sqrt{2}, b_- = (a_2 - a_3)/\sqrt{2}. \tag{5}$$

The Schrödinger equation for the state-vector $\underline{b}$ reads as

$$i\hbar\frac{d}{d}\underline{b} = \hat{H}_b\underline{b} \tag{6}$$

with Hamiltonian $\hat{H}_b$ being the Hamiltonian $\hat{H}$ (Eq. (3) transformed to the new basis,

$$\hat{H}_b = \hbar \begin{pmatrix} \varepsilon(t) & -\Omega_1(t)/\sqrt{2} & -\Omega_1(t)/\sqrt{2} \\ -\Omega_1(t)/\sqrt{2} & \Omega_2(t)-\Delta_0/2 & \Delta_0/2 \\ \Omega_1(t)/\sqrt{2} & \Delta_0/2 & -[\Omega_2(t)+\Delta_0/2] \end{pmatrix}. \tag{7}$$

3. The results of numerical simulations

We proceed with numerical solution of the Schrödinger equation for the amplitudes of bare states with Hamiltonian from Eq. (3) assuming a same Gaussian envelopes of the laser pulses: $f(t) = \exp[-(t/\tau_p)^2]$, $\Omega_{1,2}(t) = f(t)\Omega_{1,2}^{(0)}$ where $\tau_p = \tau_L / \left(\sqrt{2\ln 2}\right)$ with τ_L being the full width at half maximum and $\Omega_{1,2}^{(0)}$ being the peak amplitudes of the pulses. Assuming linear chirp for the carrier frequency of the chirped laser pulse, we have for the detuning of the frequency chirped pulse: $\varepsilon(t) = \omega_{L1}^0 - \omega_{12}^0 + \beta \cdot t$. The population of the bare states of the atom and the absolute value of the created coherence in the field of the laser pulses are presented in Fig. 2

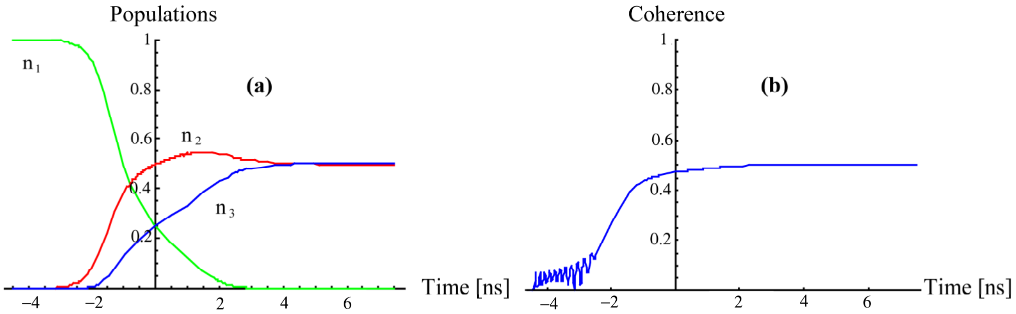

Fig. 2. Dynamics of the states populations in the laser field (a), and absolute value of the coherence created between the states $|2\rangle$ and $|3\rangle$ (b). Parameters applied are: the peak Rabi frequencies $\Omega_1^{(0)} = \Omega_2^{(0)} = 15GHz$, chirp speed $\beta = 5GHz/ns$, detuning of the field with constant carrier frequency $\Delta_0 = 0$, and duration of the pulses $\tau_p = 1.5ns$.

As it is seen from Fig. 2, a maximum value of coherence of 0.5 is created as a result of the interaction. In Fig. 3, the final absolute value of the coherence between the initially empty ground state and excited $|2\rangle$ state is shown by a color map versus the chirp speed and peak Rabi frequency. As it can be seen from this Figure, the value of the created coherence is extremely robust against variation of the peak Rabi frequencies of the pulses and of the speed of the chirp taking into account that both these parameters are given in logarithmic scale.

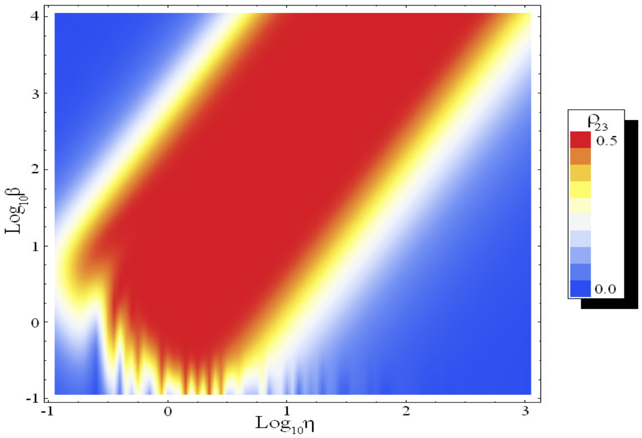

Fig. 3. Color plot of the absolute value of the created coherence versus the speed of the chirp and the peak Rabi frequency $\Omega_2^{(0)}$ of the laser pulse with constant carrier frequency (in logarithmic scale): $\Omega_2^{(0)} = \eta \cdot \Omega_1^{(0)}$ with a fixed value of the peak Rabi frequency of the chirped laser pulse $\Omega_1 = 15GHz$. Other parameters applied are: $\tau_p = 3ns$, $\Delta_0 = 0$.

As it was mentioned in the Introduction, one has to create coherent superposition states with a *same* (arbitrary) phase of the coherence in *all* atoms of the ensemble for efficient coherent enhancement of the processes of multi-photon ionization or high-order harmonic generation. The scheme under consideration allows robust creating coherence with a phase that does not depend on the shape, duration and intensity of the laser pulses or the speed of the chirp in the adiabatic regime of interaction. In the same time, the phase may be controlled by the sign of the frequency chirp: for a positive frequency chirp the created coherence has a phase equal to $\varphi = 0$ radians; and the phase of the coherence $\varphi = \pi$ radians for the opposite direction of the chirp (negative chirp), Fig. 4.

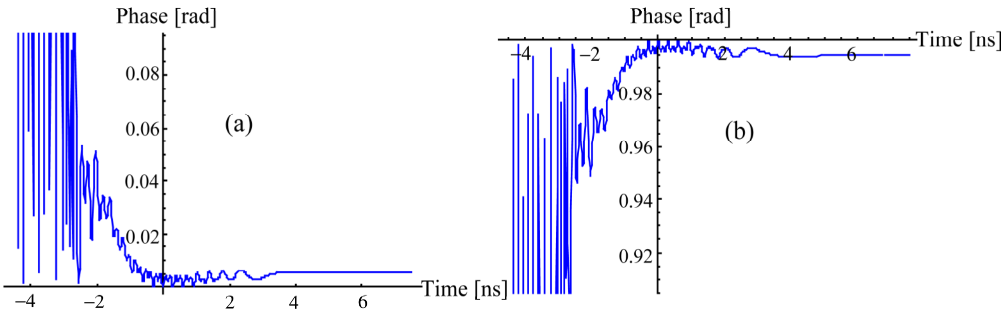

Fig. 4. The phase of the coherence between the initially empty ground and excited states in π - radians. (a)- in the case of positive chirp $\beta = 5GHz / ns$, and (b)- in the case of negative chirp $\beta = -5GHz / ns$. Other parameters applied are the same as in Fig. 2.

Such a behavior of the phase may be explained as follows. The population of the initially populated ground state is transferred to one of the superposition states, either the symmetric state with the coherence having phase of 0 radians, or the anti-symmetric one having coherence with phase equal to π radians.

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3. Conclusions

In conclusion, a scheme for adiabatic creation of maximum coherence of 0.5 between ground and excited states has been proposed and analyzed. The underlying physics of the process is explained on the basis of symmetric and anti-symmetric superposition states. The extreme robustness of the proposed scheme makes it possible to effectively create a same (maximum) coherence between the ground and excited states in an optically thick atomic medium even in the case of tightly focused laser beams. The latter case is especially important when the coherently prepared atomic medium is used for example, for generation of high-order harmonics or multi-photon ionization requiring tight focusing of laser radiation for achieving ultra-high light intensities. In addition, the possibility to robustly control the phase of the created coherence simultaneously for all atoms in an ensemble makes the proposed scheme applicable also in the processes of coherent nonlinear optics including nonlinear wave mixing and anti-Stokes or hyper-Raman scattering. An important application of the scheme may be found in nonlinear microscopy based on coherent anti-Stokes Raman scattering or on third harmonic generation: creation of maximum coherence in the focal point will significantly decrease the applied laser pulse energy resulting in non-destructive analysis, especially important in biology and medicine. Detailed consideration of these applications will be the subject of our upcoming paper. Note, that while the case of single-photon transitions between the atomic states was considered in the proposed scheme, multi-photon transitions as well may be efficiently utilized for creating coherence between a ground state and the excited state in atoms and molecules.

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