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Population inversion in a strongly driven two-level system at far-off resonance

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[Abstract] We show theoretically that an efficient population transfer can proceed in a two-level system when an intense laser pulse whose photon energy is only around half of the energy separation of the two levels is suddenly turned on. We assume that a two-level system in the ground level starts interacting with an intense laser field suddenly at t = 0 and expand a time-dependent wave function of the two-level system in terms of two time-dependent Floquet states, describing the field-induced mixing between the two field-free levels. At t = 0, a significant amount of population appears in the upper Floquet state, and as time goes on, in response to the decrease in the amplitude of the laser pulse, the time-dependent Floquet states are adiabatically transformed into the field-free levels with their populations being kept constant, resulting in the inverted populations, which may be relevant to the population inversion of N_2^+ created in a laser-induced filament.

[Introduction] Laser induced ionization preferentially occurs at the peak of the laser pulse, leaving generated ions immediately interacting with a strong laser field, which is quite different from the interaction with an entire laser pulse starting from the zero field strength. A recent example is the population inversion observed in the electronically excited B state and the electronic ground X state of N_2^+ [1]. We investigated theoretically the population transition dynamics of the interaction between a two-level system and a sudden turn-on pulse. We found after solving the time-dependent Schrödinger equation that almost 100% population transfer from the ground state to the excited state is possible even when the fundamental frequency of the laser field is far off-resonance, that is, the photon energy is only around a half of the energy separation of the two levels. The required conditions are: (i) the system interacts with a sudden turn-on pulse, corresponding to the creation of the ion species within an intense laser pulse and (ii) the laser field is sufficiently strong.

In order to explain this efficient population transfer, we employ the Floquet formalism. Initially the Floquet theory was developed to describe the dynamics of a system interacting with a laser field with a constant amplitude, where the system is governed by a periodic Hamiltonian and is completely specified by Floquet states and the quasi energies. The Floquet theory was also demonstrated as valid in describing the dynamics of a system interacting with a laser pulse whose envelope varies slowly [2]. We demonstrate that the time-dependent dynamics of the two-level system interacting with a sudden turn-on pulse whose photon energy is only around a half of the energy separation of the two levels is well described by the quasi-stationary Floquet theory. Through the quasi-stationary Floquet states, which adiabatically connects to the field-free states, we find that the sudden turn-on laser pulse distributes a significant amount of population to the upper Floquet state at time t = 0. This population equals the final population in the field-free excited state after the field vanishes.

[Methods] The wave function of the two-level system is governed by the time-dependent Schrödinger equation,

$$i\hbar\frac{\partial}{\partial t}\begin{pmatrix} c_1(t) \\ c_2(t) \end{pmatrix} = \begin{pmatrix} \varepsilon_1 & -\mu E(t) \\ -\mu E(t) & \varepsilon_2 \end{pmatrix}\begin{pmatrix} c_1(t) \\ c_2(t) \end{pmatrix}, \tag{1}$$

where the wave function of the system can be expressed as a superposition of the two field-free states having the energies of ε_1 and ε_2, μ is the transition dipole moment, and $E(t)$ is the laser field being a Gaussian-shape half pulse with the frequency of ω and the half width of 20π. The coefficients $c_1(t)$ and $c_2(t)$ specify the wave function, $|\Psi(t)\rangle = c_1(t)|1\rangle + c_2(t)|2\rangle$. By solving the first order differential equation (1), the time-dependent populations in the two levels can be obtained.

In quasi-stationary Floquet theory, the wave function of the system can be expressed as

$$|\Psi(\tau)\rangle = \sum_{q=1}^{2} k_q e^{-i\varepsilon_q\tau}|\phi_q(\tau)\rangle, \tag{2}$$

using the quasi-stationary Floquet states: $|\phi_q(\tau)\rangle = \sum_{n=-\infty}^{\infty}\sum_{\alpha=1}^{2} \phi_{\alpha,q}^{(n)}(\tau)|\alpha\rangle\, e^{-in\tau}$.

The Floquet states $\phi_\alpha^{(n)}(\tau)$, can be calculated from time-dependent Floquet equation,

$$\sum_{n=-\infty}^{\infty}\sum_{\alpha=1}^{2}\left[(\varepsilon_\alpha - m)\delta_{nm}\delta_{\alpha\beta} + a(\tau)\left(\delta_{m,n+1} + \delta_{m,n-1}\right)\left(\delta_{\alpha,\beta+1} + \delta_{\alpha,\beta-1}\right)\right]\phi_\alpha^{(n)}(\tau) = \varepsilon\phi_\beta^{(m)}(\tau), \tag{3}$$

where $a(\tau)$ stands for the coupling between the two levels.

[Results and Discussion]

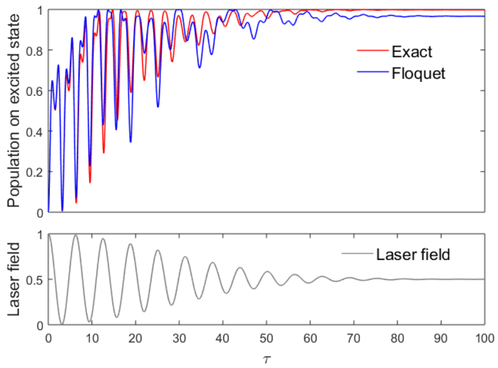

Fig. 1. Time-dependent population in the field-free excited state obtained by solving Eq. (1) numerically and that obtained by the quasi-stationary Floquet theory.

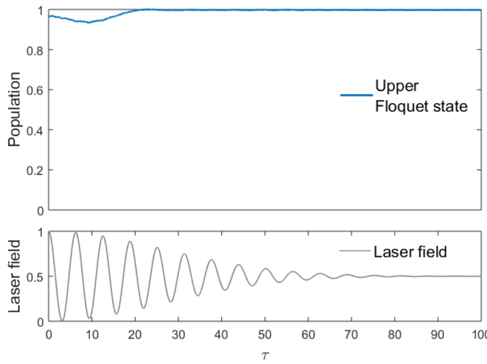

Fig. 2. Time-dependent population in the upper Floquet state.

Numerical results that we obtained after solving the time-dependent Schrödinger equation show that almost 100% population transfer can be achieved when $-\mu E_0/\hbar\omega = 0.768$, and the resultant time-dependent population in the excited state is shown by the red curve in Fig. 1 when $(\varepsilon_2 - \varepsilon_1)/\hbar\omega = 2$. By projecting the wave function of the system obtained numerically onto the upper Floquet state, the time-dependent population on the upper Floquet state is obtained as shown in Fig. 2. Differently from the time-dependent population in the field-free excited state, which is oscillating with a frequency higher than the laser frequency, the population of the upper Floquet state is close to one from the beginning of the interaction, and is kept almost unchanged during the interaction with the remaining laser pulse. By setting the population in the Floquet states to be the same as that at time t = 0, we find that the dynamics of the system is in good agreement with the numerical results as shown in Fig. 1. Thus, we explain the mechanism as follows: for a two-level system interacting with a sudden turn-on pulse whose photon energy is around a half of the energy separation of the two levels, the population in the upper Floquet state can become significantly large from the beginning of the interaction, and this population adiabatically becomes the population in the field-free excited state.

[References]

[1] H. Xu, E. Lötstedt, A. Iwasaki, K. Yamanouchi, *Nat. Commun.* **6**, 8347 (2015).
[2] I. Maruyama, T. Sako, and K. Yamanouchi, *J. Phys. B* **37**, 3919 (2004).