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# Population inversion in laser-driven $N_2^+$

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**Abstract.** The time-dependent population transfer process of $N_2^+$ generated in an intense laser pulse has been investigated using the quasi-stationary Floquet theory by assuming that $N_2^+$ experiences an intense laser pulse with the sudden turn-on. A light-dressed B state is formed with a significant amount of population when pulse is suddenly turned on and is adiabatically transformed to the vibrational ground state ($v = 0$) of the field-free B state when the pulse vanishes. In addition, a part of the population is transferred to the electronically excited A state through one-photon resonance, which also contributes to decreasing the final population in the X state, facilitating the population inversion between the B state and the X state.

## 1 Introduction

A pulsed intense laser field induces ionization of atoms and molecules when the amplitude of the laser field is sufficiently high, so that the generated ions populated in most cases dominantly in the electronic ground state is exposed to the remaining part of the intense laser pulse. This sudden-turn-on of the intense laser field for the ion species created in the intense laser field is expected to induce dynamical processes which could not be realized through the interaction of atoms and molecules with an intense laser pulse whose amplitude increase and decrease smoothly.

Recently, we investigated theoretically a population transfer in a two-level system interacting with an intense laser field with sudden turn-on and demonstrated using quasi-stationary Floquet theory that a population is transferred efficiently from the lower level to the upper level even when the energy gap between the two levels is twice as large as the photon energy of the laser field [1]. We found that, at the very beginning of the interaction ($t = 0$), a significant amount of population is distributed in the upper Floquet state associated with the sudden-turn-on of the laser field and that this upper Floquet state is connected adiabatically to the field-free upper state appearing after the interaction with the laser pulse, resulting in the population transfer to the field-free upper state. This scenario of the population transfer to the excited state is expected to be universal, and therefore, it can be applied to the interpretation of population inversion of any kind of atomic and molecular ions created in a pulsed intense laser field.

It was reported recently that, when 800 nm intense laser pulses were focused in air, coherent emission at 391 nm was generated, showing that the population inversion was achieved between the electronically excited B state and the ground X state in $N_2^+$ created in the laser field [2]. In the present study, in order to investigate the mechanism of the population

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inversion leading to the air lasing, we apply the quasi-stationary Floquet theory to the three lowest lying electronic states, X, A, and B, of $N_2^+$ exposed to an intense laser pulse with the sudden turn-on.

## 2 Method

The electronic ground X state of $N_2^+$ is coupled optically with the electronically excited B state by a light field component parallel to the N-N molecular axis while the X state is coupled optically with the electronically excited A state by a light field component perpendicular to the molecular axis. There is no optical coupling between the A state and the B state because both of them have *ungerade* symmetry. The optical transition between the X state and the B state is off-resonance when the light field is in the near-IR wavelength range.

Using the potential energy curves of the X, A, and B states of $N_2^+$, we first derive the field-free basis set $\{\psi_{\alpha,v}(r)\}$, where $\alpha$ stands for the electronic state, X, A, or B, and $v$ stands for the vibrational quantum number. The time-dependent wave function of $N_2^+$ interacting with the laser field is represented as

$$\Psi(r,t) = \sum_{\alpha=X,A,B} \sum_{v=0}^{N-1} c_{\alpha,v}(t)\psi_{\alpha,v}(r), \tag{1}$$

where $r$ is the internuclear distance, $\left|c_{\alpha,v}(t)\right|^2$ is the population in the $v$-th vibrational state of the electronic state $\alpha$, and $N$ stands for the maximum number of vibrational states included in each electronic state. The same $N$ value is adopted for the three electronic states.

In order to investigate the mechanism of the population transfer between the X and B states, we apply the quasi-stationary Floquet theory [1]. By solving the Floquet eigenequation at each point in time, the time-dependent Floquet states are obtained. Using the time-dependent Floquet states, the wave function of the system can be expressed as

$$\begin{aligned}\Psi(r,t) &= \sum_{q=1}^{3N} k_q e^{-i\varepsilon_q^F t/\hbar}\Phi_q(t) \\ &= \sum_{q=1}^{3N} k_q e^{-i\varepsilon_q^F t/\hbar} \sum_{n=-m}^{m} \sum_{\alpha=X,A,B} \sum_{v=0}^{N-1} \phi_{\alpha v,q}^{(n)}(t)\psi_{\alpha,v}(r)e^{-in\omega t},\end{aligned} \tag{2}$$

where $k_q$ is a coefficient, and $\varepsilon_q^F$ is the eigenenergy for the $q$-th Floquet state $\Phi_q(t)$. In eq. (2), $\phi_{\alpha v,q}^{(n)}$ is the coefficient of the $v$-th vibrational state on the electronic state $\alpha$ shifted by $n$ photons, and $\omega$ is the angular frequency of the laser field.

The population in the $q$-th Floquet state is obtained as the projection of the wave function of the system, which is obtained by solving the Schrödinger equation numerically, onto the $q$-th Floquet states calculated at time t as

$$\left|k_q(t)\right|^2 = \left|\langle\Psi|\Phi_q\rangle\right|^2. \tag{3}$$

Because the Floquet state is correlated to the corresponding field-free state after the pulse vanishes, we label the Floquet state, which becomes the $i$-th vibrational state on the field-free electronic state $\alpha$, as the $\alpha(v=i)$ Floquet state.

## 3 Results and discussion

By solving the time-dependent Schrödinger equation, the time-dependent populations in the Floquet states of $N_2^+$ were obtained. In the calculation, $N=4$ vibrational states are included in each electronic state, and the maximum dressed photon number is set to be

$m = 6$. In Fig. 1(a), the populations in the seven most populated Floquet states are plotted. In the calculation, the angle $\theta$ between the N-N molecular axis and the polarization direction of the laser field was set to be $\theta = 60°$, so that the coupling strength between the field-free X($v$ = 0) state and the field-free A($v$ = 0) state is comparable to that between the field free X($v$ = 0) state and the field free B($v$ = 0) state.

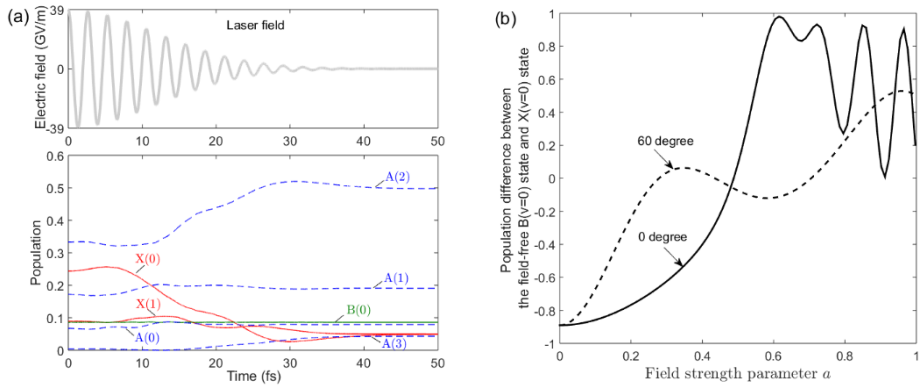


**Fig. 1.** (a) Time-dependent population on the Floquet states at the field intensity of $2 \times 10^{14}$ $\mathrm{Wcm}^{-2}$, and $N_2^+$ ions are aligned so that the angle between the N-N molecular axis and the polarization direction of the laser field is $\theta = 60°$. (b) Final population difference between the field-free B($v$ = 0) state and X($v$ = 0) state, $\Delta P = P(B) - P(X)$, as a function of the laser field strengths obtained as the exact solutions of the Schrödinger equation.

As seen in Fig. 1(a), the B($v$ = 0) Floquet state has 10% population when the field is suddenly turned on, and stays almost constant during the period of interaction with the laser field, which means that the population gained by the B($v$ = 0) Floquet state at $t$ = 0 is transferred adiabatically to the vibrational ground state of the B state. At the same time, the population is also transferred among the Floquet states formed by the non-adiabatic Floquet coupling between the X and A states at the near one-photon resonance. At the end of the laser pulse, the population in the field-free X($v$ = 0) state becomes smaller than that of B($v$ = 0) state, showing that the population inversion is achieved between the X and B states.

In order to investigate the role of A state, we performed the same calculation by setting the alignment angle at $\theta = 0°$. As shown in Fig. 1(b), when $\theta = 0°$, the minimum laser field intensity required for achieving the population inversion between the vibrational ground X state and the vibrational ground B state becomes almost twice as large as that obtained when $\theta = 60°$. This is because, at $\theta = 60°$, an efficient population transfer proceeds from the X state to the A state by one-photon transition, which reduces the population in the X state and facilitates the population inversion between the X state and the B state, resulting in the air lasing at 391 nm. We conclude that the population inversion achieved between the B state and the X state of $N_2^+$ is ascribed to the sudden turn-on of the intense laser pulse as well as to the population transfer from the X state to the A state.

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