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Population inversion in laser-driven N_2^+

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[Abstract] We simulate the time-dependent population transfer process of N_2^+ generated in an intense laser pulse using the quasi-stationary Floquet theory by assuming that N_2^+ experiences an intense laser pulse with sudden turn-on. We find that a light-dressed B state 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 the one-photon resonance, which also contributes to decreasing the final population in the X state. 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.

[Introduction] A pulsed intense laser field induces ionization of atoms and molecules when the amplitude of the laser field is sufficiently high, and the generated ions start being exposed to an intense field immediately. Recently, we demonstrated that this sudden exposure to an intense laser field could transfer a population in the lower level in a two-level system efficiently to the higher level even when the energy gap between the two levels is twice as large as the photon energy of the laser field [1]. This scenario of the population transfer to the excited state is expected to be universal and can be applied to an 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 is generated, showing that the population inversion was achieved between the electronically excited B state and the electronic 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 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.

[Methods] 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 while there is no optical coupling between the A state and the B state because both of them are *ungerade* in symmetry. By solving the Floquet eigenequation [1] at each point in time, the wave function of the system can be expressed by a Floquet basis functions as

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

where r is the internuclear distance, k_q is a coefficient, ε_q^F is the eigenenergy for the q-th Floquet state $\Phi_q(r,t)$. In eq. (1), $\phi_{\alpha v,q}^{(n)}$ is the coefficient of the v-th vibrational state in the electronic state α shifted by n photons, and ω is the angular frequency of the

laser field. The same N value is adopted for the three electronic states.

The population $|k_q(t)|^2$ in the q-th Floquet state is obtained as the projection of the wave function $\Psi(r,t)$ of the system on $\Phi_q(r,t)$. Because a Floquet state is correlated to a corresponding field-free state after the pulse vanishes, we label a Floquet state, which becomes the i-th vibrational state in the field-free electronic state α, as an $\alpha(v=i)$ Floquet state.

[Results and Discussion]

By solving the time-dependent Schrödinger equation, the time-dependent populations in the Floquet states of N_2^+ were obtained. In Fig. 1(a), the populations in the seven most populated Floquet states are plotted. In the calculation, the angle θ between the N-N

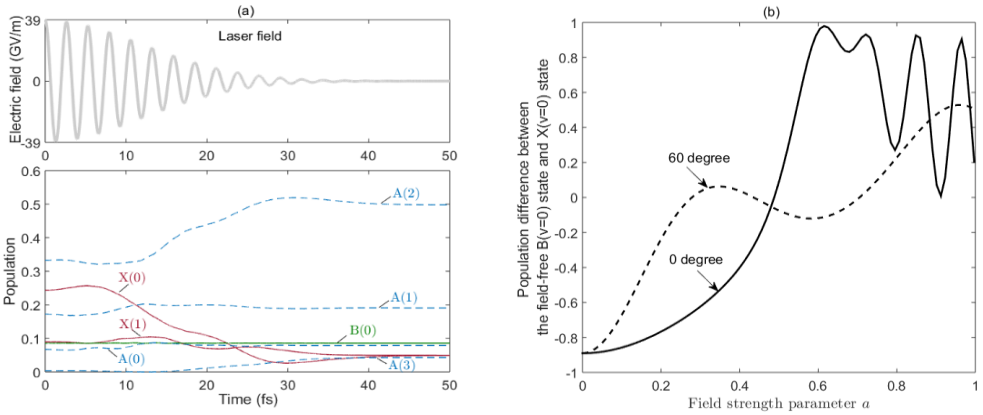

Fig. 1. (a) Time-dependent populations in the Floquet states at the field intensity of 2×10^{14} $\mathrm{Wcm^{-2}}$. The N_2^+ molecular ions are aligned so that the angle between the N-N axis and the field polarization direction becomes $\theta = 60°$. (b) Final population differences between the field-free B($v = 0$) state and the X($v = 0$) state under different field intensities obtained when the Schrödinger equation was solved directly.

molecular axis and the polarization direction of the laser field was set to be $\theta = 60°$. As seen in Fig. 1(a), about 10% of the population is transferred to the B($v = 0$) Floquet state immediately after the field is suddenly turned on, and the population stays almost constant during the interaction with the remaining part of the laser pulse, 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, population inversion between the X and B states is achieved.

We compare the results obtained when $\theta = 0°$ with that obtained when $\theta = 60°$. As shown in Fig. 1(b), when $\theta = 60°$, the minimum laser field intensity required for achieving the population inversion between the vibrational ground X state and the vibrational ground B state is only a half of that required when $\theta = 0°$, showing that the population transfer from the X state to the A state lowers 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.

[References]

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