Air lasing by population inversion in N_2^+ induced by strong-field coherent coupling of the X, A and B states

([1]*School of Science, The University of Tokyo*, [2]*Jilin University*) ○Erik Lötstedt,[1] Youyuan Zhang,[1] Toshiaki Ando,[1] Atsushi Iwasaki,[1] Huailiang Xu,[2] Kaoru Yamanouchi[1]

Keywords: air lasing, strong-field excitation, molecular dynamics, numerical simulation

When an intense, femtosecond near-IR laser pulse is focused in air, a short (~ 1 cm) plasma column called a filament is formed by the interplay between self-focusing and strong-field plasma defocusing. From the filament, unidirectional, coherent, and narrow-bandwidth radiation in the visible and UV wavelength range is emitted, which is referred to as air lasing. The emission at 391 nm, corresponding to the $B^2\Sigma_u^+(v' = 0) \rightarrow X^2\Sigma_g^+(v'' = 0)$ transition in N_2^+ (see Fig. 1), has been intensively studied in the past decades, because the air lasing at 391 nm can be produced in a wide range of experimental conditions.

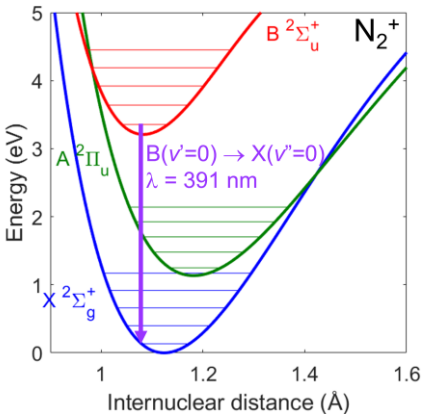

Fig. 1. Potential energy curves of N_2^+.

However, until 2015, the mechanism and the time scale of the formation of the population inversion between $B^2\Sigma_u^+(v' = 0)$ and $X^2\Sigma_g^+(v'' = 0)$ of N_2^+, resulting in the air lasing at 391 nm, had not been understood well. In 2015[1], we revealed both experimentally and theoretically that the abrupt exposure of N_2^+ to the strong laser field, which is referred to as a sudden turn-on mechanism, combined with the population pumping from $X^2\Sigma_g^+(v'' = 0)$ to $A^2\Pi_u(v' = 0)$ of N_2^+, is responsible for the population inversion and that the population inversion is built up on a femtosecond time scale. In the theoretical model, we consider the time-dependent Schrödinger equation (TDSE) for the nuclear wave packets in the $X^2\Sigma_g^+$, $A^2\Pi_u$, and $B^2\Sigma_u^+$ states,

$$i\hbar\frac{\partial}{\partial t}\begin{pmatrix}\psi_X(r,t)\\ \psi_A(r,t)\\ \psi_B(r,t)\end{pmatrix} = \left[T + V - \mathbf{E}(t)\cdot\boldsymbol{\mu}\right]\begin{pmatrix}\psi_X(r,t)\\ \psi_A(r,t)\\ \psi_B(r,t)\end{pmatrix}, \tag{1}$$

where r is the internuclear distance, $\psi_k(r,t)$ is the nuclear wave packet in the electronic state k, T is the kinetic energy operator, V is a 3×3 matrix having the potential energy curves of the $X^2\Sigma_g^+$, $A^2\Pi_u$, and $B^2\Sigma_u^+$ states on the diagonal, $\mathbf{E}(t)$ is the laser field, and $\boldsymbol{\mu}$ is a 3×3 matrix containing the dipole transition matrix elements. By numerically solving the TDSE, we showed that the population inversion can be explained by the following two mechanisms: (i) after ionization, N_2^+ starts interacting immediately with the intense laser pulse, which efficiently induces the population transfer from the $X^2\Sigma_g^+$ state to the $B^2\Sigma_u^+$ state, and (ii) because of the near-resonant $A^2\Pi_u$–$X^2\Sigma_g^+$ coupling, a population is efficiently transferred to the $A^2\Pi_u$ state, which promotes further the population inversion between the $B^2\Sigma_u^+$ state and the $X^2\Sigma_g^+$ state.

In 2019, we conducted pump-probe experiments[2] and showed that the $B^2\Sigma_u^+(v' = 0) \rightarrow X^2\Sigma_g^+(v'' = 0)$ lasing signal at 391 nm oscillates as a function of the pump-probe delay time at the frequencies corresponding to the energy differences between the vibrational states in the $X^2\Sigma_g^+$ state and those in the $A^2\Pi_u$ state, which can be regarded as direct evidence that the mechanism of the excitation and lasing process of N_2^+ is the coherent V-shaped $A^2\Pi_u$– $X^2\Sigma_g^+$–$B^2\Sigma_u^+$ coupling. Recently, we further showed[3] that the rotational coherence created in N_2^+ can be clearly seen in the pump-probe delay-time dependence of the lasing signal at 391 nm. Indeed, the rotational structure of the lasing emission at 391 nm exhibited two maxima in the R-branch emission and the two maxima moved towards the longer wavelength as the delay time increases. By extending our theoretical model by including the rotational degree of freedom of N_2^+, we showed theoretically that the delay-time dependence of the R-branch emission spectrum can be interpreted as a temporal evolution of a rotational wave packet created in the $B^2\Sigma_u^+$ state.

In order to use the air lasing for practical applications such as stand-off spectroscopic measurements and remote sensing, it is prerequisite to increase further the lasing emission intensity. We demonstrated experimentally[4] that the air lasing signal at 391 nm can be enhanced by two orders of magnitude by employing a polarization-gated IR laser pulse having a time-dependent polarization direction. At the peak of the pulse, an aligned N_2^+ ensemble is created by the strong-field ionization, and during the latter half of the laser pulse, the polarization direction is changed so that the population transfer from $X^2\Sigma_g^+$ to $A^2\Pi_u$ is promoted efficiently by the $A^2\Pi_u \leftarrow X^2\Sigma_g^+$ transition whose transition dipole moment is perpendicular to the molecular axis, resulting in the significant increase in the air lasing signal. Furthermore, we revealed[5] that the extent of the population inversion can be increased further by combining the polarization-modulated 800 nm laser pulse with a 1.6 μm IR pulse, which induces vibrational Raman transitions and depletes the population in the $X^2\Sigma_g^+(v = 0)$ state to an almost empty level, leading to a giant enhancement of the lasing signal at 391 nm by 5 orders of magnitude.

References

1) H. Xu, E. Lötstedt, A. Iwasaki, K. Yamanouchi, Nat. Commun. **6**, 8347 (2015).

2) T. Ando, E. Lötstedt, A. Iwasaki, H. Li, Y. Fu, S. Wang, H. Xu, K. Yamanouchi, Phys. Rev. Lett. **123**, 203201 (2019).

3) Y. Zhang, E. Lötstedt, T. Ando, A. Iwasaki, H. Xu, K. Yamanouchi, Phys. Rev. A **104**, 023107 (2021).

4) H. Li, M. Hou, H. Zang, Y. Fu, E. Lötstedt, T. Ando, A. Iwasaki, K. Yamanouchi, H. Xu, Phys. Rev. Lett. **122**, 013202 (2019).

5) H. Li, E. Lötstedt, H. Li, Y. Zhou, N. Dong, L. Deng, P. Lu, T. Ando, A. Iwasaki, Y. Fu, S. Wang, J. Wu, K. Yamanouchi, H. Xu, Phys. Rev. Lett. **125**, 053201 (2020).

