Rotational Mechanism of Lasing in Singly Ionized Nitrogen Molecules under Femtosecond mid-IR Pumping

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Abstract: Through high-resolution spectroscopy of lasing on N_2^+ ions at 428nm wavelength, we quantify optical gain and population inversion in the gain medium. We show that molecular rotations are the enabling effect in this lasing process.

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Air lasing is a concept based on the utilization of the common air as a gain medium in a mirrorless impulsive laser source [1]. Various schemes for air lasing are being actively investigated, motivated by its potential to enable efficient single-ended remote sensing schemes [2]. One of the most promising approaches to air lasing is based on pumping by ultra-intense femtosecond laser pulses propagating in the filamentation regime in air [3]. Intensity inside the core of the femtosecond laser filament is of the order of 100 TW/cm^2, which is sufficient to ionize oxygen and nitrogen in the air, producing, in particular, singly-ionized molecular nitrogen ions N_2^+. It has been shown that air filamentation results in optical gain on the UV electronic transitions of N_2^+, at 391 nm and 428 nm emission wavelengths [4]. However, the physical origin of the gain mechanism responsible for those emissions has been highly controversial. Apart from its practical potential, air lasing in N_2^+ presents an intriguing and complex problem in intense AMO physics. Indeed, it is counter-intuitive that optical ionization of the nitrogen molecule appears to preferentially create the nitrogen molecular ion in the upper, not the lower emission state, which is an energetically unfavorable process. In fact, there have been three recent high-profile publications that suggested three different gain scenarios [5-7]. The gain values reported so far have been too low to support spontaneous unseeded lasing. The lasing has been only observed in the forward direction, where it is seeded by the co-propagating harmonic and supercontinuum emissions that accompany filamentation. The lack of solid understanding of the gain mechanism hinders the optimization of the lasing process that could lead to unseeded lasing in the backward direction, which would be of the most practical significance.

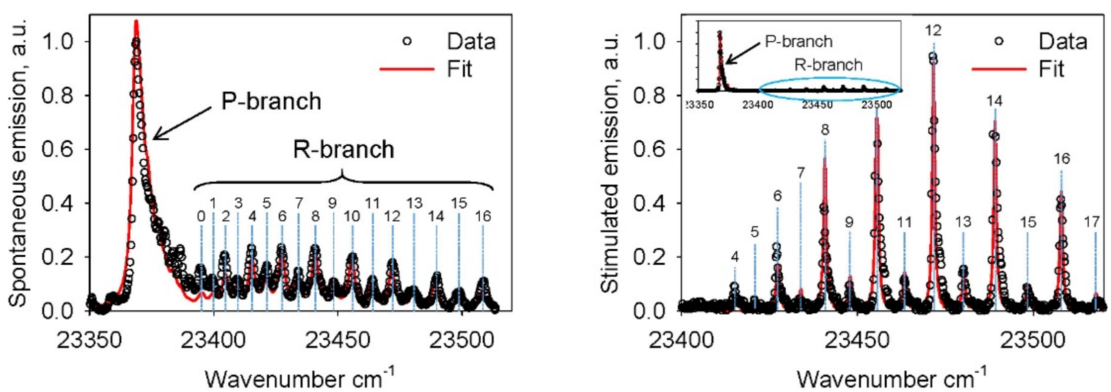

Fig. 1. Left: Experimental data and the fit for spontaneous emission measured from the side. Note a perfect match between the spectral peaks and the tabulated values for different rotational transitions in the R-branch emission, marked with the vertical lines. The corresponding values of the rotational quantum number *J* in the lower emission state are shown next to the individual lines. Spontaneous emission peaks at the *J* value of 6. **Right:** Same for the stimulated emission measured in the forward direction. The R-branch emission, which results from the transitions with the change of the *J* number of +1, is shown. The inset shows the full emission spectrum with both P and R-branch emissions. Stimulated emission peaks at the *J* value of 12, which is different from the peak value for the spontaneous emission. Based on the ratio of the stimulated emission signals on the adjacent transition lines in the R branch, we estimate the peak value of the total gain, in the 1mm-long interaction zone to be about 64, corresponding to the lower-bound estimate for the peak gain per unit length on the R-branch lines of about 40 cm^{-1}.

In this paper, we report the results of a spectroscopic study of stimulated emission in N_2^+ ions at 428 nm wavelength. The measurements of high-resolution rotational spectra of N_2^+ have been reported previously [8], but not in the connection with the gain mechanism in the N_2^+ ions. In our experiments, we pump pure nitrogen gas at 4 torr of pressure by tightly focused 60 fs laser pulses with 2 mJ of energy at 1.5 μm wavelength. The measured spectra are shown in Figure 1, for the cases of side (spontaneous) and forward (stimulated) emissions. The stimulated emission is self-seeded, in the forward direction, by a forward-propagating, spectrally broadened third harmonic of the intense pump beam. Using a knife-edge technique, we estimated the intensity of the pump beam in the interaction zone at about 600 TW/cm^2, which is above the saturated intensity for a complete single ionization of nitrogen molecules. Our key observation is that the spontaneous emission peaks at the value of the rotational quantum number J of about 6, while the stimulated emission peaks at a higher value of 12, thus enabling the rotational gain mechanism discussed below. Note that none of the recent papers [5-7] discussing various possible gain mechanisms in this system took the rotational degree of freedom into account, missing the key physical ingredient of this problem.

We fit the measured spontaneous emission spectrum, taking into account the selection rules and the degeneracy of different rotational transitions involved. To fit the data, we use a room-temperature Boltzmann distribution for the rotational subsystem in the upper emission state, which we shift, by an adjustable value, in the space of the rotational quantum number J. The best fit matches the experimental data nearly perfectly. After having determined the rotational distribution in the upper emission state, we fit the spectrum of stimulated emission, by varying the ratio of the total populations in the upper and lower electronic states of N_2^+, as well as the shift of the rotational distribution in the lower emission state, which is also assumed to be Boltzmann distribution at room temperature. The best fit is obtained for the case with net population inversion, with the ratio of populations in the upper and the lower lasing levels of 1.35. However, an additional gain mechanism based on the rotational excitation of the N_2^+ ions produces an essential contribution to the overall gain. This rotational gain mechanism is illustrated in Figure 2. It is enabled by the difference of the rotational distributions in the upper and lower emission levels that are shifted with respect to each other in J space, due to different polarizabilities of the N_2^+ molecular ions in the upper and lower emission levels. Then, in agreement with our experimental observations, the spontaneous emission peaks at the maximum of the rotational distribution in the upper emission level, while the stimulated emission peaks at the maximum of the difference of the rotational distributions in the upper and lower levels. The net population inversion between the upper and lower levels, although it takes place in our experiments, is not required to produce gain which can be entirely due to the rotational mechanism. Our findings suggest the feasibility of optimization of the gain in this system through temporal shaping of the pump pulse that would produce the most dissimilar rotational distributions in the upper and lower lasing levels.

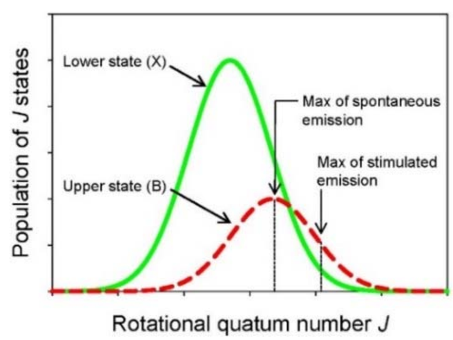

Fig. 2. Illustration of the rotational gain mechanism. If the rotational populations in the upper and lower emission levels are shifted with respect to each other, then spontaneous and stimulated emissions peak at different values of *J*. Spontaneous emission peaks at the maximum of the rotational distribution of the upper emission level, while stimulated emission peaks at the maximum of the difference between the two population distributions. Gain occurs for a sub-set of the rotational states even when the net population inversion between the upper and lower lasing levels is negative.

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