Towards remote air lasing by efficient generation of population inversion in molecular nitrogen ions

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During filamentation of intense femtosecond (fs) laser pulses propagating in air, highly efficient light amplification can be observed at 391 nm, corresponding to the $B^2\Sigma_u^+(\upsilon'=0) \rightarrow X^2\Sigma_g^+(\upsilon''=0)$ transition in the molecular nitrogen ion [1]. We present a lasing-without-inversion (LWI) mechanism [2-4] that is active in this amplification process, under standard conditions where fs laser filamentation leads to self-guiding of light. We show that the LWI process is triggered by the combination of strong-field molecular ionization and molecular alignment, i.e., it arises naturally during the propagation of intense fs laser pulses in the air, using only the natural dynamics of a multi-level quantum system. The amplification process requires no coherence between the excited and the lower electronic states; effectively, the LWI comes "for free". Our pump-probe simulations show [2, 3] that neither electronic nor rotational population inversion in the medium is necessary. Amplification is possible thanks to the coherent rotational dynamics of the ion. The proposed mechanism, which we call *rotational quantum beat lasing without inversion*, is general. The only necessary ingredients are non-negligible population in the upper state and molecular rotations, both unavoidable in intense fs laser fields.

To date, significant light amplification in the air filaments has been observed in the forward direction only. Also the rotational quantum beat lasing without inversion occurs in the forward direction. However, it is highly desirable to induce and control a *bidirectional* cavity-free laser in the open air, since such a system would be of greatest practical use for remote spectroscopy, i.e., remote detection of atmospheric chemical content, e.g., air pollutants or toxic gases, due to its higher efficiency and sensitivity compared to conventional LIDAR (light detection and ranging) techniques based on the *non-directional* backscattering of light.

To this end, we have extended our coherence-based amplification scheme to include a second (control) pulse that generates true electronic population inversion in the molecular nitrogen ions in the filament, thus providing the possibility of backward lasing. Our pump-control scheme [5] takes advantage of the complex laser-induced ro-vibronic molecular dynamics as well as the cross-phase modulation of the control pulse caused by the pump-induced alignment dynamics of the molecular nitrogen ions. In contrast to conventional laser systems, the pump pulse creates by tunnel ionization notable population in the upper, but also in the intermediate and especially in the lower (ro-vibronic) states of the gain medium, making efficient generation of population inversion challenging. We show that the rotation of the molecules can imprint such a shift and sweep on the instantaneous frequency of the control pulse that allows depletion of the lower laser state by efficiently transferring its population to the excited states while suppressing its re-population. Importantly, the proposed air lasing method provides gain windows for specific pump-control time delays without requiring fine tuning of the pulse parameters such as pulse area, peak intensity, or frequency.

References

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