Thermal Non-Equilibrium in a Femtosecond Laser Filament using CARS

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Femtosecond (fs) lasers have become commonly used in optical diagnostics mostly due to the advantages provided by the high intensity allowing for nonlinear effects without the need of high energy per pulse, and short pulses allowing for dynamics measurements with high temporal resolution. In a fs-laser induced filament, the plasma column generated by the fs laser pulse in the focal region can extend well beyond the Rayleigh range due to the interplay between the positive contribution of self-focusing and negative contribution of the plasma to the nonlinear refractive index of refraction [1]. This talk will present measurements of thermo-dynamical non-equilibrium and its dynamics inside a fs filament using Coherent Anti-Stokes Raman Scattering (CARS). For more than a half century, CARS has been successfully used to measure the temperature in gases in plasmas by identifying the Maxwell-Boltzmann distribution of the vibrational and rotational modes of the constituent molecules. Hybrid fs/ps CARS [2] allows for probing ro-vibrational population distributions and inferring the vibrational and rotational temperatures in a single laser shot. This capability offers not only identification of thermodynamic non-equilibrium in the system, but it also enables monitoring the dynamics of the of vibrational and rotational temperature with high temporal resolution. Here we present non-equilibrium vibrational and rotational temperature measurements inside the fs filament performed using hybrid fs/ps CARS [3]. To circumvent the ambiguity due to the CARS signal generation from both hot and cold regions surrounding the filament, the N2 v1 $\rightarrow$ v0 peak which has contributions from both the hot and cold regions was ignored, and only the higher vibrational levels were used for temperature estimates, since they are obtained only from the hot region. The temporal dynamics of vibrational temperature was also obtained by varying the delay between the filament-generating laser and the CARS probe laser. Under highly non-equilibrium conditions, typical of hypersonic flows, low temperature plasmas, or laser filaments, an overpopulation of the higher vibrationally excited levels can occur, such that the non-equilibrium takes places not only between the rotational (and hence translational) and vibrational modes, but also between the vibrational modes which no longer follow a Maxwell-Boltzmann distribution. Our measurements indicate that significant non-equilibrium non-Boltzmann distribution is observed in fs laser induced filaments. In such cases, we can no longer assign a classical thermodynamics-based temperature, and the gas properties are more accurately captured by the distribution of its quantized energy levels. Since the temperature cannot be defined unambiguously, the non-equilibrium condition is better characterized by the population distribution instead of temperature. References [1] S. L. Chin, Femtosecond Laser Filamentation Induced Phenomena and Applications (Springer, 2020). [2] D. Pestov, R. K. Murawski, G. O. Ariunbold, X. Wang, M. Chi, A. V. Sokolov, V. Sautenkov, Y. Rostovtsev, A. Dogariu, Y. Huang, and M. O. Scully, "Optimizing the Laser Pulse Configuration for Coherent Raman Spectroscopy," Science 316, 265 (2007). [3] R. Rosser, V. Blanchard, G. Urdaneta Rincon, A. Yalin, and A. Dogariu, "Non-equilibrium temperature dynamics in a femtosecond filament using Hybrid Coherent Anti-Stokes Raman Scattering," Opt. Lett. 50, 6638 (2025).