Abstract Submitted
for the DAMOP15 Meeting of
The American Physical Society

Sign inversion of nonlinear optical response of nitrogen upon ionization[1] MARYAM TARAZKAR, Department of Chemistry and Center for Advanced Photonics Research, Temple University, DMITRI ROMANOV, Department of Physics and Center for Advanced Photonics Research, Temple University, ROBERT LEVIS, Department of Chemistry and Center for Advanced Photonics Research, Temple University — We report hyperpolarizability calculations for the nitrogen radical cation at neutral and relaxed geometries in the static and dynamic non-resonant regime, using multi-configurational self-consistent field (MCSCF) response theory. The results were compared with those computed using density functional theory (DFT). The open-shell electronic system of nitrogen radical cation was found to exhibit negative second-order optical nonlinearity. The drastic change in the magnitude and sign of the hyperpolarizability coefficient $\gamma^{(2)}$ from the neutral nitrogen molecule to radical cation indicates an enhanced role of excitations in the polarization response of ion as compared with the neutral molecule. The second-order optical properties of nitrogen radical cation have been also calculated as a function of bond length starting with the neutral molecular geometry (S_0 minimum) and stretching the N-N triple bond, reaching the ionic D_0 relaxed geometry, and all the way toward dissociation limit. The results obtained provide the potential for controlling optical properties of laser filament wake channels.

[1]This work was supported through the Air Force Office of Scientific Research, MURI Grant FA9550-10-1-0561.

Dmitri Romanov
Temple University

Date submitted: 02 Feb 2015 Electronic form version 1.4