Strong-field control of coherent anti-Stokes Raman scattering in iodine vapor with shaped ultrashort laser pulses

MARTIN BITTER, University of British Columbia, Physics & Astronomy, EVGENY A. SHAPIRO, VALERY MILNER, University of British Columbia, ULTRAFAST COHERENT CONTROL GROUP TEAM — Extensive work has been done to investigate molecular dynamics in weak laser fields. In contrast, our understanding of molecular behavior and the possibilities to control it with strong laser pulses is still limited. In this work, we investigate the process of coherent anti-Stokes Raman scattering (CARS) in iodine vapor for different strong-field regimes. Saturation of the CARS signal with increasing pulse intensities is observed and studied both experimentally and theoretically. We show that it is possible to overcome this saturation by implementing different schemes of coherent control based on the technique of femtosecond pulse shaping. Optimal regimes for enhancing molecular CARS response to strong-field excitation are proposed and demonstrated, paving the way to more efficient nonlinear spectroscopy.