Ultrafast saturation of Raman coherence

Anil Patnaik[1, 2], Sukesh Roy[3], and James Gord[1]

[1]*Aerospace Systems Directorate, Air Force Research Laboratory, Wright-Patterson AFB, OH 45433 USA*

[2]*Department of Physics, Wright State University, Dayton, OH 45435 USA*

[3]*Spectral Energies, LLC, 5100 Springfield Street, Ste. 301, Dayton, OH 45431 USA*

A strong laser field can drive an atomic or molecular transition to saturation when the associated Rabi frequency exceeds the decays and dephasing associated with the transitions of interest. The dynamics of saturation exhibit typical Rabi oscillations at a rate that is proportional to the square root of the intensity of the driving field. If the pulse duration of the driving field is longer than the Rabi period of oscillation, steady-state saturation can be obtained. However, if the pulse duration is shorter than the Rabi period and the decay timescale, the saturation dynamics become more complex. By increasing the intensity of the driving laser to such an extent that the Rabi period becomes shorter than the pulse duration, an ultrafast non-equilibrium saturation of the transition can be achieved.

The same reasoning applies to Raman coherence, where a pair of pump and Stokes fields can generate the steady-state saturation if the two-photon Rabi frequency is higher than the decay and dephasing rates. This has been shown in the regime of nanosecond-pulse-based excitation [1]. However, if the coupling lasers are short-duration pulses on the order of femtoseconds (fs), this laser can couple multiple rotational states simultaneously in addition to its role as pump laser [2]. We observed that the saturation dynamics are then determined primarily by the saturation of the rotational Raman coherence. Also, the Raman coherence may exhibit saturation-like behavior if a strong probe resonantly couples the Raman-excited state to another electronic transition in the long-pulse regime [3] – of course in the non-equilibrium sense. Note that the saturation thus observed is on a time scale of the vibrational period of molecules and is orders of magnitude faster than state-of-art electronic switches. A very intuitive understanding of the saturation in these ultrashort-pulse regimes is obtained by calculating the pulse area associated with the pulse. This study gives predictive capability for the intensity threshold to avoid Raman and CARS saturation, which is very important for ultrafast spectroscopic measurements.

[1] A. K. Patnaik, S. Roy, J. R. Gord, R. P. Lucht, and T. B. Settersten, J. Chem. Phys. 130, 214304 (2009).

[2] A. K. Patnaik, S. Roy, and J. R. Gord, Phys. Rev. A 87, 043801 (2013).

[3] A. K. Patnaik, J. R. Gord, and S. Roy (in preparation, 2013).