Ultrafast thermometry using quantum coherence

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

[1]*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*

The macroscopic parameters of the physical environment surrounding a target system affect the microscopic quantum states of the constituent molecules within the system. For example, the quantum statistics of atomic or molecular states in thermal equilibrium are dictated by a Boltzmann distribution function that is characterized by the temperature of the environment. Usually, an electromagnetic field can strongly interact with those microscopic states locally without affecting the macroscopic property of the system. Thus, laser probes for measuring the population distribution have been very widely used to achieve accurate nonintrusive thermometry. With the advent of new laser sources with shorter pulse durations [up to a few femtoseconds (fs)], along with the availability of high-power lasers and the ability to control accurately the phase properties required for obtaining pulses of arbitrary shape, ultrafast techniques have become indispensable diagnostic tools [1].

The ultra-short pulses have an extremely wide bandwidth for simultaneous coupling of a number of internal states of a target system for a very short time duration (a few to 100 fs), causing multiple coherent excitations that are all in the same phase [2]; e.g., a 532-nm laser with a 100-fs pulse duration has a bandwidth of 350 cm^{-1} that typically couples many molecular states in the target system. Those coherence excitations function in much the same way as a group of classical coupled harmonic oscillators with slightly different characteristic frequencies. The higher the temperature, the greater is the number of oscillators. Once the pulse ceases, the initial coherence begins to randomize. This decoherence process contains the thermometric information that is extracted by coupling a suitable combination of lasers to generate and measure the coherent anti-Stokes Raman scattering signal [3]. However, thermometric measurements become complicated by multiple interfering molecular coherences that are generated by different pairs of photons from the wide bandwidth of the pump beam.

We will focus on revisiting the effect of quantum coherence on thermometry in the ultrafast regime using the ultra-short laser beams. We will discuss the advantages and challenges involved in ultrafast thermometry and present some recent exciting diagnostic results obtained by our group using phase-manipulated shaped pulses [4].

[1] S. Roy, J. R. Gord, and A. K. Patnaik, Prog. Energy Combust. Sci. 36, 280-306 (2010).

[2] M. O. Scully et al., PNAS 99, 10994 (2002).

[3] R. P. Lucht, P. J. Kinnius, S. Roy, and J. R. Gord, J. Chem. Phys. 127, 044316 (2007).

[4] S. Roy et al., Appl. Phys. Lett. 95, 074102 (2009).