Measurements of flame temperature by femtosecond CARS

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Time-resolved femtosecond coherent anti-Stokes Raman spectroscopy (fs-CARS) is utilized to measure the methane/oxygen/nitrogen flame temperature at atmospheric-pressure. The measurements are performed using the CARS signal of N_2 with 40-fs laser pulses in the first few picoseconds after the initial in-phase excitation. Flame temperatures at 300 and 1 325 K are measured, the experimental results show good agreements with theoretical ones and present a good repeatability.

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Nanosecond coherent anti-Stokes Raman spectroscopy (CARS) has been used for measuring temperature in gas-phase reacting flows for many years[1−3]. The drawbacks of nanosecond CARS are the influences of nonresonant background signals and low repetition rates[4]. With the development of ultra-short laser technology, femtosecond CARS (fs-CARS) was utilized to overcome most of the problems associated with nanosecond CARS recently[5]. Beaud *et al.*[6] firstly used fs-CARS for flame thermometry. Lucht *et al.*[7] furthermore put forward the new principle for measuring temperature in gas-phase reacting flows by fs-CARS. The CARS signal of N_2 is advantageous for measurement of temperature in air-fed reacting flows, because of the high concentration of N_2 throughout the reaction zone. The CARS signal is measured only in the first few picoseconds after the in-phase excitation to ensure the reduction or elimination of influences of nonresonant background and collisional effects[8]. In this letter, we measured the time-resolved fs-CARS signals of N_2 with 40-fs laser pulses, and then simulated the theoretical results with a simple model. The methane/oxygen/nitrogen flame temperatures are extracted from the comparison of theoretical and experimental data points by least-square fit.

A schematic diagram of fs-CARS system is shown in Fig. 1. The laser pulses from a commercial femtosecond laser system (Micra+Legend, Coherent) at 800 nm (2.5 mJ, 1 kHz, and 40 fs) were divided into two parts by a 50% beam splitter. One beam was used as the Stokes beam and the other was used to pump an optical parametric amplifier (OPA, TOPAS, Light Conversion). The output 675-nm beam of OPA was split into two parts by a 50% beam splitter to obtain the pump and probe beams. The frequency difference between the pump pulses and the Stokes pulses matches the Raman resonance frequency of nitrogen ($\nu = 2\,330$ cm^{-1}). The generation of CARS requires temporal and spatial overlap of the pulses in the samples. The relative timing among the different beams was varied by computer controlled delay stages. The three beams were aligned parallel to each other, and overlapped in the folded-BOXCARS beam geometry (4-mm spot diameter, square with 8-mm sides). The three beams were focused on flame using a 300-mm focal lens at room temperature. Premixed methane/oxygen/nitrogen flame at atmospheric pressure is studied. Nitrogen contents are 54%. Flame temperature is 1 325 K, which is measured by thermo couple. This folded-BOXCARS beam geometry ensures that the CARS signal propagates in a direction different from the incoming beams and can therefore be background-free collected. The CARS signal was spectrally dispersed in the monochromator (Omni-λ500, Zolix) and detected by a photo multiplier tube (PMT, PMTH-S1-CR131, Zolix) with a BOXCAR (SR250, Stanford Research Systems).

The theory model is presented in Ref. [7]. Briefly, the theoretical CARS signal as a function of probe delay (τ) is calculated using the following equations:

$$S(\tau) = \int_{-\infty}^{+\infty} I_{\mathrm{pr}}(t-\tau)[P_{\mathrm{res}}(t) + P_{\mathrm{nres}}(t)]^2 \mathrm{d}t, \qquad (1)$$

$$P_{\mathrm{nres}}(t) = \alpha E_{\mathrm{p}}(t) E_{\mathrm{s}}(t), \qquad (2)$$

$$P_{\mathrm{res}}(t) = \beta \left[\int_{-\infty}^{t} E_{\mathrm{p}}(t') E_{\mathrm{s}}(t') \mathrm{d}t' \right] \times \sum_j \left\{ \Delta N_j \left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} \right)_j \cos(\omega_j t) \exp(-\Gamma_j t) \right\}, \qquad (3)$$

where $P_{\mathrm{res}}(t)$ and $P_{\mathrm{nres}}(t)$ are the resonant and nonresonant polarizations, respectively; I_{pr} is the intensity of the probe pulses (Gaussian pulse); $E_{\mathrm{p}}(t)$ and $E_{\mathrm{s}}(t)$ are the time-dependent electric-field amplitudes of the pump and Stokes pulses; α and β are arbitrary scaling factors

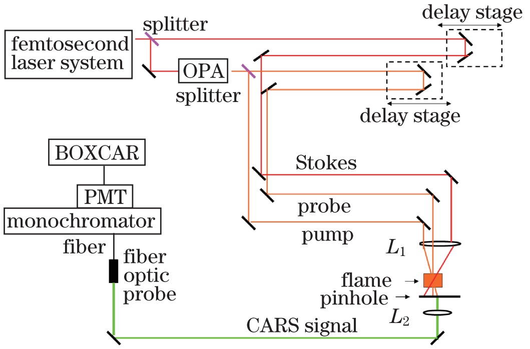

Fig. 1. Schematic diagram of fs-CARS system.

used to match the experimental signal with the theoretical spectrum; j presents different Raman transition; ω_j is the angular frequency; Γ_j is the Raman linewidth; ΔN_j is the difference in population between the excited and ground levels of a Raman transition; $(\mathrm{d}\sigma/\mathrm{d}\Omega)_j$ is the Raman cross section. The parameters for temperature- and pressure-dependent Raman transition are obtained from the Sandia CARS spectral-fitting code[9].

Dependence of fs-CARS signals on probe delay for different temperatures is shown in Fig. 2. The flame temperatures are 300 and 1 325 K. From Fig. 2, we can see that the two signal traces are approximately the same at the probe delay less than 200 fs. The signal is the maximum value at time zero, which is normalized to a peak intensity of 1 at the probe delay of zero. Dependence of fs-CARS signals on probe delay for temperature 300 K is measured six times. Comparison of theoretical and experimental data points at 300 K is shown in Fig. 3. In Fig. 3 solid lines represent the theoretical best-fit results (based on least-square fit) and symbols represent the experimental data points. The theoretical results are in agreement with the experimental data. In the six measurements, the mean best-fit temperature is 325 K and the random error is ±25 K. The experiments present a good repeatability. The good agreement between theory and experiment and the good repeatability of experiment demonstrate the feasibility of this method. Dependence of fs-CARS signals on probe delay for temperature 1 325 K is shown in Fig. 4. The best fit results based on least-square fit are both 1 050 K (black line in Fig. 4), which have a big gap compared with the real temperature. The reason may be that the CARS intensity for high temperature at large probe delay is far less than the intensity at zero probe delay. The weighted least-square fit is used for obtaining the best-fit results. The weighted factor is $1/S$ (S is the CARS intensity). The best fit results based on weighted least-square fit are 1 300 and 1 200 K (blue line in Fig. 4). We can see that the weighted least-square fit is effective in the high temperature measurement.

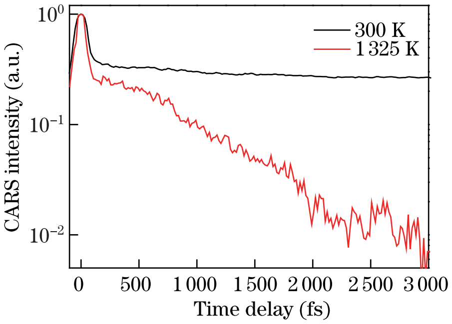

Fig. 2. Dependence of fs-CARS signal on probe delay for different temperatures.

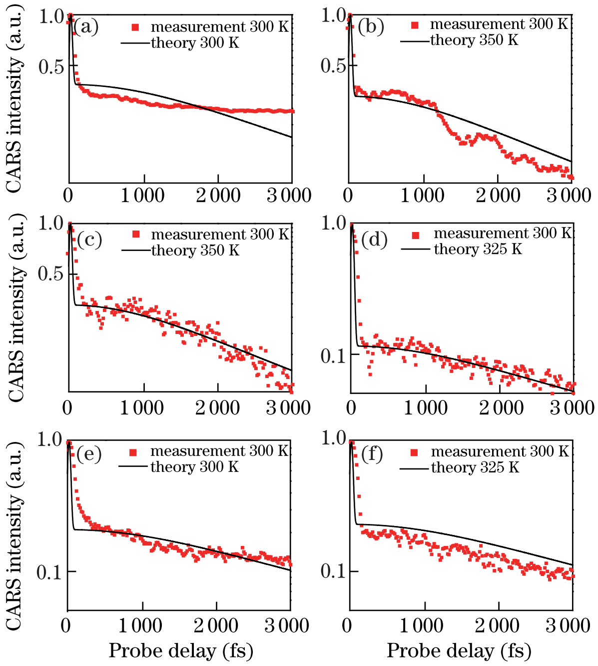

Fig. 3. Comparison of theoretical and experimental results at 300 K (based on least-square fit).

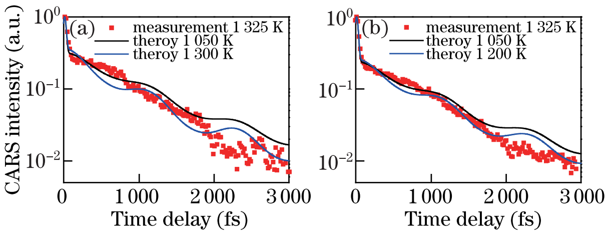

Fig. 4. (Color online) Comparison of theoretical and experimental data points at 1 325 K based on least-square fit (black line) and weighted least-square fit (blue line).

In conclusion, by using the time-resolved fs-CARS, temperatures 300 and 1 325 K are studied in the atmospheric-pressure, methane/oxygen/nitrogen flame. This method shows a potential in flame temperature measurements by fs-CARS.

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