

## METHOD CARS FOR MEASUREMENT OF HIGH PRESSURE BURNING TEMPERATURE

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### Introduction

Burning of solid propellants in rocket engines occurs at pressure of 4÷8 MPa and temperature of 2500÷3200 K. A thin (approximately 0.1 mm) zone of chemical transformations locating in immediate proximity from a degraded surface moves with the speed of 10÷40 mm/s. Chemically aggressive substances and particulate matter contained in a flow of gaseous reaction products complicate study of flow parameters. The temperature in front of flame is the main of them. With application of optical methods, including Coherent anti-Stokes Raman Scattering of light – CARS, the requirements to measurements on temporal and spatial resolution and absence flow perturbation of a flame zone become feasible.

Both the techniques for excitation and detection of scattered radiation and computer program synthesizing the spectra of researched molecules at elevated temperature and pressure allow the CARS measurements. In this nonlinear four-photon process two pumping laser photons with a frequency  $\omega_1$  and one Stokes photon with a frequency  $\omega_2$  interact with a certain molecules, for example,  $N_2$  and produce coherent radiation with an anti-Stokes frequency  $\omega_3$ . The complete spectrum of probable frequencies of scattered light has the complex form dependent on temperature. The latter can be determined from comparison of an experimental spectrum with a set of those synthesized for different temperatures. Signal detection with a frequency scanning of narrow-band radiation of the dye-laser  $\omega_2$  requires several minutes and can be used to perform measurements in steady flows only [1]. For single-shot generation of entire spectrum with frequencies  $\omega_3$  the broadband radiation of the dye-laser was applied in [2]. We choose the two-frequency CARS, which does not require too high power of lasers. In this case, instantly measured intensities of signal on two fixed frequencies can be used for definition of temperature in the region of crossing beams as well as the form of a complete spectrum CARS [3].

The aim of this paper is the development of a two-frequency CARS method for burning temperature measurement of stoichiometric mixture of ammonium dinitramid with polycapromamide. This model fuel contains nitrogen in exhaust. Time of burning of a one sample is 0.5 s at the pressure of 4.0 MPa in measuring volume.

### CARS spectra calculation

This technique for temperature measurement is based upon the spectral structure of the vibrational ground state band  $0 \rightarrow 1$  ( $\Delta v = +1$ ,  $\Delta J = 0$ ) of nitrogen. Named Q-branch, this band consists of narrow positioned unresolved lines. CARS signal is generated with the following intensity

$$I_3 = \left( \frac{4\pi^2 \omega_3}{c^2} \right)^2 |\chi^{(3)}|^2 I_1^2 I_2$$

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where  $I_1$  and  $I_2$  are the laser and Stokes intensities,  $c$  is the speed of light. The nonlinear cubic susceptibility consists of nonresonant and resonant (real and imaginary) parts [4]

$$\chi^{(3)} = \chi^{NR} + \chi^R = \chi^{NR} + \chi' + i\chi''$$

where

$$\chi^R = \sum_{v,J} \frac{R_{v,J}}{\Omega_{v,J} - (\omega_1 - \omega_2) - i\Gamma_J}$$

with  $\Omega_{v,J}$  – the transition frequency,  $\Gamma_J$  – the half-width of the line. Intensity of transition

$$R_{v,J} = \frac{2N}{\hbar} \left( \frac{c}{\omega_2} \right)^4 \left( \frac{d\sigma}{d\Omega} \right)_{v+1} b_{J,J} \Delta_{v,J}$$

with  $N$  – the numerical density of molecules,  $\hbar$  – the Planck's constant  $\div 2\pi$ ,  $\frac{d\sigma}{d\Omega}$  – the Raman scattering cross section. The Placzek–Teller coefficients equal

$$b_{J,J} = \frac{J(J+1)}{(2J-1)(2J+3)}$$

The population difference of the levels, involved in transition

$$\Delta_{v,J} = n_{v,J} - n_{v+1,J}$$

where

$$n_{v,J} = \frac{1}{Q} g_J (2J+1) \exp\left(-\frac{(G_v + B_v J(J+1))hc}{kT}\right)$$

with  $Q$  – the partition function,  $g_J$  – the statistical weight determined by the nuclear spin,  $G_v$  – the vibrational term,  $B_v$  – the molecular rotational constant,  $k$  – the Boltzmann's constant,  $T$  – the temperature.

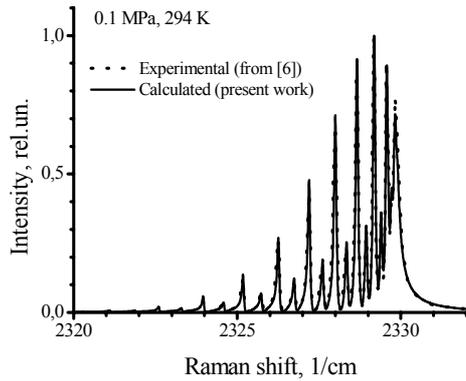


Fig. 1. Spectra of  $N_2$  at pressure of 0.1 MPa and temperature of 294 K.

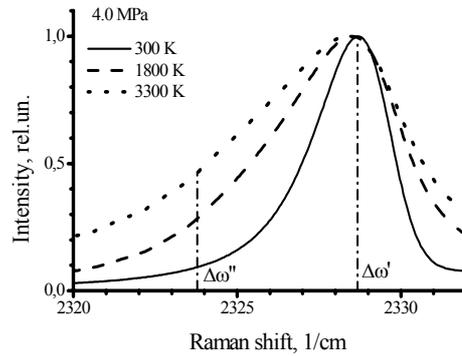


Fig. 2. Calculated spectra of  $N_2$  at pressure of 4.0 MPa and temperatures of 300 K, 1800 K and 3300 K.

As shown in [5], the line interference effect in a 0→1 Q-branch can be taken into account if an imaginary part of susceptibility is used in the form

$$\chi''(J) \sim p \frac{\gamma_J - ((\omega_1 - \omega_2) - \Omega_{v,J})Y_J}{(\Omega_{v,J} - (\omega_1 - \omega_2))^2 + (p\gamma_J)^2}$$

with  $p$  – the pressure,  $\gamma_J$  – the line broadening coefficient,  $Y_J$  – the line mixing coefficient.

Using these formulas one can calculate the shape of N<sub>2</sub> CARS spectrum (Fig. 1 and 2). Here Raman shift  $\Delta\omega$  is the difference from laser's frequency  $\Delta\omega = \omega_1 - \omega_2$ . The experimental spectrum (dot line) from work [6] and calculated spectrum at room conditions are shown in Fig. 1. With a growth in temperature and pressure it becomes smoother and has slight grades (see Fig. 2). In the present method, two narrow frequency CARS signals were generated only. Their Raman shifts  $\Delta\omega' = \omega_1 - \omega_2'$  and  $\Delta\omega'' = \omega_1 - \omega_2''$  are marked by vertical lines in Fig. 2. The ratio of their intensities gave us the flame temperature.

### Measurement technique

A special closed vessel used as the combustion chamber has been filled with background gas at pressure of 4.0 MPa. Four transmitting windows were used for laser diagnostics and for observation. Burning at high temperatures and pressure caused distortions of laser beams because of the large gradient of refraction on flame boundary resulting in either deterioration or disappearance of a signal. Therefore helium was used as a background gas. It has the refraction similar to that of the high-temperature combustion products. The sample of fuel ( $\varnothing 10$  mm, h = 10 mm) was a vertical cylinder in the center of the chamber. The top end was ignited with NiChrome wire. The gas system provided the constant pressure with accuracy of 5%.

For two-frequency excitation of a CARS signals the output of a dye-laser with two narrow lines on Stokes frequencies  $\omega_2'$  and  $\omega_2''$  were used. After its interaction with radiation on the laser frequency  $\omega_1$  a scattered radiation on two anti-Stokes frequencies  $\omega_3' = 2\omega_1 - \omega_2'$  and  $\omega_3'' = 2\omega_1 - \omega_2''$  were generated. For that a part (60%) of doubled output radiation from a Q-switched Nd:YAG laser (532 nm, 70 mJ,  $2 \cdot 10^{-8}$  s, 10 Hz) pumped a scanned dye-laser ( $\approx 607$  nm, 8 mJ,  $1.5 \cdot 10^{-8}$  s). This dye-laser beam and the residual part (40%) of solid-state laser radiation split into two beams were focused (Fig.3) by the lens ( $f = 390$  mm) and reduced together, forming the measuring volume ( $0.15 \times 0.15 \times 6$  mm<sup>3</sup>). The CARS beam, arising in the measuring volume entered the entrance slit of a spectrograph ( $f = 820$  mm). In its exit slit the multichannel analyzer, controlled by the computer, was installed. It included the optical amplifier and photodiode array ( $0.15 \times 25$  mm, 1024 pixels). The contribution of nonresonant susceptibility into a signal was suppressed by adjustment of both polarizations in pumping beams ( $\omega_1 - 0^\circ$ ,  $\omega_2 - +60^\circ$ ) and analyzer ( $\omega_3 - -60^\circ$ ) in anti-Stokes beam.

The frequencies and intensities of dye-laser radiation were adjusted using

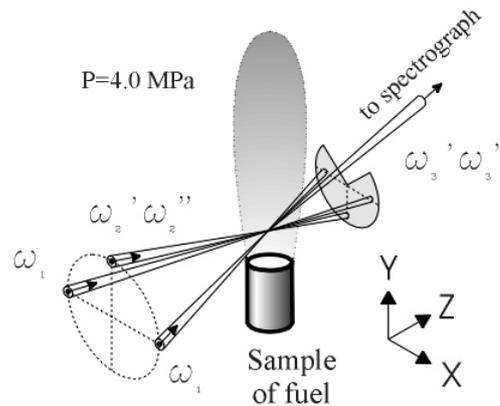


Fig. 3. Measurement geometry.

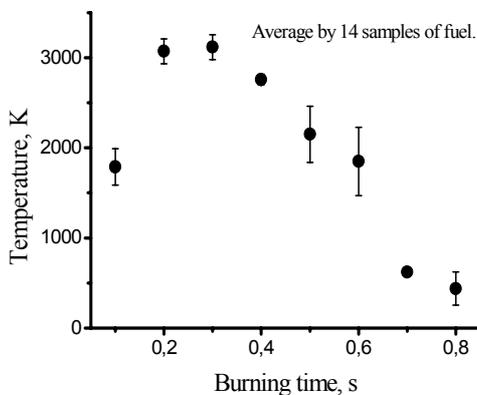


Fig. 4. Measured temperature versus burning time.

frequency (see Fig.2). And, finally, when the dye-laser resonator was tuned to a shorter wave, simultaneously with reduction of intensity of the first line in the laser spectrum, the other line arose. Corresponded to the next transmission peak of EFP, it has Raman shift of frequency  $\Delta\omega'' = 2323.85 \text{ cm}^{-1}$ . The tuning was stopped in that moment, when intensities of both lines of  $\omega_2'$  and  $\omega_2''$  equalized.

### Experimental results

A distance of 1 mm between measuring volume and the top end of sample was adjusted. Previously the spectrum of the dye-laser was detected with accumulation on 25 pulses. The received amplitudes of lines were used further for alignment of diodes sensitivities on two fixed frequencies. After that the "control" two-frequency spectrum CARS was detected at  $T_0=293 \text{ K}$  with the same accumulation. After complete replacement of gas by helium at pressure of 4.0 MPa in the chamber, forty single-shot CARS spectra were detected with repetition of 10 Hz. Within this period of time, ignition and subsequent burning of a sample was performed. In course of the postprocess inspection of experimental data, the beginning of burning was determined with accuracy of 0.1 s by occurrence in spectra of a small background from flame. It was possible to detect up to 5÷6 spectra during the burning.

The technique was approved in experiments for gas temperature measurement ( $1174\pm60 \text{ K}$ ) at the axe of a spiral from 0.15-mm NiChrome wire reeled coil to coil. It was heated with an electrical current up to temperature of  $1273\pm5 \text{ K}$  as indicated by the pyrometer with a disappearing string (length of a spiral was 10 mm, internal diameter was 2.5 mm, pressure was 4.0 MPa, volumetric concentration of nitrogen was 0,02).

The measurements are carried out at burning of fourteen samples. The calculated temperature dependence of the CARS intensities ratio for two certain frequencies was used at the following data processing. In Fig. 4 the average temperatures are given depending on time of sample burning. In these experiments, the statistical error of individual measurement up to 500 K was mainly caused by shot-noise of the photodetector and instability of laser radiation power. The temperature of burning averaged by 9 instant meanings detected within a 0.2÷0.3 s equals to 3097 K ( $\pm 3 \%$ ).

chamber and registration system. The air pressure of 0.1 MPa was increased by addition of helium up to pressure of 4.0 MPa. By tuning of dye-laser wavelength the maximum of anti-Stokes signal was achieved. After that an etalon Fabry-Perot (EFP) with base of 1.03 mm was installed in the resonator of the scanned laser. In such interferometer the difference of frequencies of the neighbour transmission peaks equals to  $4.84 \text{ cm}^{-1}$ . As a consequence the laser linewidth decreased from  $1 \text{ cm}^{-1}$  to  $0.1 \text{ cm}^{-1}$ . To decline the etalon, the maximum signal was achieved again. Thereby determined by EFP transmission peak of N-th order the dye-laser wavelength was adjusted to a maximum of a Q-branch  $\text{N}_2$  with a  $\Delta\omega' = 2328,69 \text{ cm}^{-1}$  Raman shift of

Thus, the technique for local optical measurements of burning temperature in substances containing the nitrogen as a combustion product is developed. It can be applied for creation and tests of solid fuel used in rocket engineering.

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