TERAHERTZ TECHNIQUE FOR RESEARCH OF WAVES OF BURNING AND A DETONATION WITH USE OF FREE ELECTRON LASER

A.A. Vasiliev^{1, 4}, E.I. Palchikov^{1, 4}, V.V. Kubarev^{2, 4}, E.N. Chesnokov³, P.V. Koshlyakov³, A.V. Dolgikh^{1, 4} and I.Yu. Krasnikov^{1, 4}

1 Lavrentyev Institute of Hydrodynamics, Siberian Branch, Russian Academy of Sciences, Novosibirsk, 630090 Russia

2 Budker Institute of Nuclear Physics, Siberian Branch, Russian Academy of Sciences, Novosibirsk, 630090 Russia

3 Voevodskii Institute of Chemical Kinetics and Combustion, Siberian Branch, Russian Academy of Sciences, Novosibirsk, 630090 Russia

4 Novosibirsk National Research State University, Novosibirsk, 630090 Russia e-mail: palchikov@hydro.nsc.ru

Abstract

Schemes, experimental methods, and results of registration of wave of burning of an oxygen-hydrogen mixture by means of terahertz radiation are described. Experiments in the range of terahertz wavelengths from 115 to 200 microns are made. Researches were conducted at the lines of absorption of water vapor and OH radical. At the same time registration was conducted in the optical range of wavelengths from 0.4 to 0.9 microns. Experiments of visualization in the terahertz range of absorption in the stationary mode of burning of oxygen-hydrogen mix are made. The results obtained with optical detectors, pyroelectric detectors and Schottky barrier detectors in a dynamic experiment with the flame propagating along a pipe are presented.

Introduction

Any shared system of gas-dynamics and chemical-kinetics equations that describe wave propagation in chemically active media is unstable. The obtainable combustion and detonation waves and their structures are essentially not one-dimensional [1].

The spatial nonuniformity of the front considerably complicates studies of the kinetics of chemical reactions during propagating waves of combustion and detonation and hinders the search for an answer to the question, What is the contour of the reaction zone in such a structure, what concentration of what substances exist in various areas of a zone of reaction and at different stages of process? Knowledge of the detailed dynamics of chemical processes in combustion and detonation waves enables us to create better–quality models for their description and to model (calculate) more accurately the nonstationary processes of transitioning from subsonic combustion to supersonic detonation.

Formulation of the Problem

Usual optical methods of registration of the fast-proceeding processes in gases are limited by lengths of waves from ultra-violet to distant infrared range [2, 3, 4]. The main restrictions of spectral characteristics and speed are connected with the radiation source power, speed of rotation of mirrors, number of electrooptical converter channels, CCD channels, and other elements of system. Expansion of spectral range opens new possible arrangements and carrying out new experimental studies.

This work attempts to overcome the existing difficulties in recording the detailed dynamics of reactions beyond a subsonic combustion front of the hydrogenoxygen gas mixtures using the Novosibirsk Free Electron Laser (FEL) [5].

Terahertz wavelengths of 115 to 200 μ m contain a great many lines of the absorption of polar molecules of water and OH radicals. Most initial combustion components are nonpolar and transparent (e.g., O₂, H₂).

A free electron laser has record power and can be tuned to any radiation absorption line in this range, making it possible to separately, by reaction components study the kinetics of combustion and detonation waves in gaseous mixtures. The laser can thus help create unique measuring techniques in the terahertz band.

Thus, by means of the laser in the terahertz range unique techniques of measurement can be created.

Studying the Spatial Distribution of Water Vapor in the Flame

To measure oxyhydrogen flame absorption, we used a cooled burner. The FEL beam was modulated by a mechanical interruptor, passed through the flame, and was incident on a PM–4 pyroelectric detector. The signal from the pyroelectric detector arrived at a synchronous detector, along with the reference signal from the interruptor. The effect of IR radiation from the flame on the pyroelectric detector was

thus excluded. Figure 1 shows that absorption by water vapor on the 166.81 μ m line was ~50–60%.



Fig. 1. Water vapor absorption at the 166.81 µm line.

To record the spatial distribution of absorption by water vapor, we used a matrix of Pyrocam III [6] pyroelectric detectors with a resolution of 124×124 pixels. Figure 2 illustrates the absorption by water vapor at the wavelength of 166.81 µm. We observed the flame of the stoichiometric $2H_2 + O_2$ mixture. The thickness of the burning layer along the FEL beam was 80 mm, and its width was 20 mm. The height of the flame was 5mm.

One can see that the regions of cooled water vapor are on the edges of the burner and higher, closer to the center. In close proximity to the burner's surface, the hot water vapor on the line in use are quite transparent, but small fluctuations of about 1 mm are directly observable in the combustion zone. The interference fringes in the upper right corner are not related to the flame but are an artifact of the recording track.



Fig. 2. Spatial distribution of the intensity of FEL radiation transmitted through the burner flame. Dark areas correspond to high concentrations of water vapor.

Dynamic Experiment with a Propagating Flame

For high-speed shooting of detonation processes the ideal registering system there would be a fast video camera of terahertz range. However, only slow terahertz cameras with a frequency of frames up to 50 Hz are created now.

The choice of the single-channel terahertz fast detectors operating at the room temperature not wide, they are pyroelectric detectors or Schottky diode detectors [7].

Schematics of our dynamic propagating–flame experiments are shown in Fig. 3. A polypropylene glass 155 mm tall with an inner diameter of 39 mm, with opened bottom, was filled with the stoichiometric $2H_2 + O_2$ mixture. The optical path of the laser beam was filled with dried air to prevent losses as it traveled. The beam was transmitted at half the glass's height. The mixture was kindled by an electric spark in the lower part of the vessel. Using the pyroelectric detector and the Schottky barrier diode detector, we measured the level of the signal that passed through the channel with the flame. Synchronous detection was used with the Schottky barrier detector. The reference signal corresponding to the bunch repetition frequency was taken from the acceleration track. The integration time constant during synchronous detection was $0.3 \,\mu$ s.



Fig. 3. Diagram of our dynamic experiment with (a) a pyroelectric detector and (b) a Schottky diode detector.

The data obtained with the PM–4 pyroelectric detector are shown in Fig. 4. The signal from the pyroelectric detector was processed with allowance for the inertia of this particular detector and the recording channel (the load resistance).

Because of the capacitive character of the pyroelectric detector, the initial signal was of an integral nature with a certain characteristic integration time. The integral equation for it was:

$$f^{*}(t) = \int_{0}^{\infty} f(t-t') \cdot A(t') dt'$$
 (1)

where $f^{*}(t)$ is the measured signal, f(t) is the initial signal, and A(t) is the instrument function of the measuring track.

To restore the initial signal, we used deconvolution with the instrument function of the recording system. The instrument function of the recording channel was measured with a single nanosecond pulse from a CO_2 laser. The instrument function in the first approximation was a critically short step with an exponential decay whose measured fall time is e times, 21 ms. In calculating the initial signal, we replaced integral equation (1) with a system of linear equations that be easily reduced to a triangular form.

The results from the simple differentiation of the photodetector signal and the results from deconvolution with allowance for the instrument function shown in Fig. 4 are almost identical. The only difference is that the transmission minimum during deconvolution occurs at the 50th ms rather than at the 30th ms characteristic of differentiation.



Fig. 4. Data obtained with the pyroelectric detector after simple differentiation and after deconvolution with allowance for the instrument function.

Figure 5 shows a signal passing through the flame front in the stoichiometric $2H_2 + O_2$ mixture, registered by the superfast Schottky barrier diode-based detector. The laser wavelength was 167 μ m.

The results obtained with various sensors are comparable in their characteristic times and the general form of the signals.

Five microseconds after ignition, blooming occurred for $\sim 10-15$ ms because of the heating of water vapors in the channel and their displacement. Absorption then grew due to cooling and the formation of water vapor as a result of burning with the transmission minimum at the 50th ms. According to the time interval between the ignition and the arrival of the combustion front, the velocity of the combustion wave was 15 m·s⁻¹, or 1.5 times higher than the velocity of the laminar flame for the stoichiometric mixture under atmospheric pressure, indicating a certain turbulization of the mixture in the glass under study.

Thus, when using pyroelectric detectors in our measurements, additional processing of signals is required because initial terahertz signals contain both high-frequency, and low-frequency harmonicas which are distorted by the detector of this type. Therefore for these measurements we used the high-speed terahertz detector on the basis of the diode with a Schottky-barrier, which transferred a power signal without distortions. Speed of the detector allowed to resolve separate bunch pulses of FEL, following with a frequency of 5.6 MHz and, respectively, to use synchronous detection of these pulses. The constant of integration time was 0.2 ms.



Fig. 5. Data obtained with the Schottky barrier detector.

Dynamics of the front of burning in the optical range

To coordinate results of terahertz experiments on dynamic behavior of waves of burning with physical processes in the optical range, dynamics of the front in a transparent pipe from a polimetilpentene (TPX) with the same diameter (40 mm) and 280 mm long with closed end was investigated (the opposite end from an ignition place, in figure 4 – at the top).

On a sequence of frames (figure 6) can be selected characteristic stages of flame distribution along the pipe: ignition of mix and evolution of the front burning, extending with a speed of 200 m/s; its stop, turbulization and the reverse movement along a pipe (0.6-1 ms) after arrival of the reflected shock wave from the closed end

of the pipe; transition from burning to a detonation and light emission of products on all length of the pipe (1.2-1.4 ms), and brighter area at first is formed in the top part of a pipe and then moves down, with the subsequent afterburning and cooling of products of a detonation (1.6 ms and further, outside the removed record).

Registration of absorption at lines of the radical OH

In figure 7 the modified scheme for dynamic experiments with the extending flame is submitted.

At the same time with a terahertz signal optical flash in the range of lengths of waves $\lambda = 0.4 \div 0.9$ microns, for what the silicon photodiode detector was used, was registered.

In experiment the sealed dural cylindrical channel with a diameter of 40 mm and 1 m long (figure 8) closed at one end by the crushable membrane which was filled from the electrolyzer with stoichiometric $2H_2 + O_2$ mixture through a dehumidifier was used. Filling through drainage entrance realized at the atmospheric pressure by means of the moved piston.



Fig. 6. Dynamics of the front of burning of $2H_2 + O_2$ mixture taken in the optical range.

There are polypropylene windows inserted in the channel transparent for the terahertz radiation and visible light. This allows to view the processes going in a pipe at distance of 300 or 700 mm from an ignition place. Ignition of mixture have been produced by the electric spark in the top part of the tube. The system with the mobile piston completely sealed from external air when filling with mixture. The design of the channel allows to carry out initiation of burning and detonation with various boundary conditions (the pipe end faces opened or closed by membranes or plug), thus it is possible to form and investigate as low-speed processes of burning (laminar and turbulent), and accelerated flames, up to transition of burning to a detonation.



Fig. 7. The scheme of dynamic experiment with the Schottky barrier diode and optical detectors



Fig. 8. a) scheme and general view of the channel with a diameter of 40 mm and 1 m long, which was filled with mix $2H_2 + O_2$; b) the provision of a pipe in installation in an optical path at the exit of the terahertz laser. The oscilloscope was synchronized simultaneously with the ignition voltage

given on a spark plug (a zero timepoint on all oscillograms). For synchronous

research of dynamics of the phenomenon in the optical range the registered light signal through a polypropylene window in the same place where passed terahertz signal was taken. In figure 9,a it is visible (channel 1) that approximately through 1 ms after initiation the front of burning reaches the window which is at distance of 300 mm from a place of ignition and rather smoothly accrues in the range from 1 to 2 ms. It is caused by that the accelerated flame generates the compression waves which are running away from it forward. At a certain stage these waves of compression, outrun up with each other, merge and form the shock front. On the shock front parameters of gas change in steps, unlike smoothly accruing in a compression wave. Then burning forms the shock front (with possible subsequent strengthening of intensity of a shock wave and even transition of burning to a detonation). Two peaks are connected with reflection of shock waves from walls and an end of a pipe. The terahertz signal (channel 2) correlates with a signal in the optical range.

In figure 9,b one can see (channel 1) that through 1.8 ms the sharp front is observed at once. It means that to this area (700 mm from an ignition place) compression waves from the accelerated front of burning have time to create a incident shock wave. And at the distance of 700 mm the detector registers bright flash earlier, than at the distance of 300 mm, it is coordinated with a sequence of frames in figure 6 (a shot from 1.2 ms), there at the distant end from an initiation place flare brightness is also more, than at the neighbor. Existence of several extrema is connected with reflection of shock waves from walls and an end of a vessel. The terahertz signal (channel 2) also has sharper front and decay is longer on time.

Measurement of absorption on the line of the radical OH was obstructed by stray signals from close located lines of absorption of water which become wider at high temperatures and especially – with high pressures (to 10 atm and more) in a reaction zone.

More details about the reasons for selecting of specific lines of absorption on which experiments were made and results of the researches it is possible to get in [8].



Fig. 9. The channel 1 – light radiation signal; the channel 2 – the terahertz signal of FEL registered by the Schottky barrier diode detector which is adjusted to the 119.3 micron (83.8 cm⁻¹) line of the radical OH at distances: a) 300 mm and b) 700 mm from an ignition point

Conclusions

- Schemes for experiments were developed. Results from recording of oxyhydrogen combustion with terahertz radiation have been presented.
- Using a pyroelectric matrix-based detector, the spatial distribution of water vapor was obtained along the cross-section of the oxyhydrogen flame during the stationary burning of a stoichiometric $2H_2 + O_2$ mixture. Radiation absorption from 50 to 80% was measured in different regions of the flame at a wavelength of 167 µm and a burning layer depth of 80 mm.

- Single pyroelectric detectors and superfast Schottky barrier–based photodetectors were used in dynamic experiments with a propagating flame. The dynamics of a $2H_2$ + O_2 mixture's combustion in a cylindrical channel with combustion front velocities of up to 15 m·s⁻¹ was studied. The data obtained with the pyroelectric detectors and Schottky barrier detectors yield similar results for times exceeding 3 ms.
- In the cylindrical channel with a diameter of 40 mm and 1 m long which was filled with the drained mixture $2H_2 + O_2$ researches in different places of the cylindrical canal at $L_1 = 300$ mm and $L_2 = 700$ mm distance from ignition place are conducted. Researches on the line of absorption of the radical of OH are conducted. At the time of several milliseconds correlation between absorption of terahertz radiation, an optical signal and acceleration of the front of a flame with formation of a incident shock wave is obtained. On the line of the radical OH at distance of 300 mm from an initiation place absorption at the time of maxima of brightness of a light signal is about 20%.
- The data on essential absorption of terahertz radiation obtained in this work as products of reaction (H_2O and OH) of oxygen-hydrogen mixture burning can form a basis for creation of more advanced installation which has to consist of several at the same time working Schottky barrier diode detector with the reduced sensitivity to a refraction deflection of terahertz radiation carried along an axis of the cylindrical channel. To see dynamics of difficult structure of the front, the line of fast Schottky barrier diode detectors is also necessary. This line doesn't exist yet and it needs to be created.

REFERENCES

- 1. Ю.Н Денисов, Я.К. Трошин. Пульсирующая и спиновая детонация газовых смесей в трубах // Доклады АН СССР. Т. 125. № 1. С. 110-113. 1959.
- А.С. Дубовик. Фотографическая регистрация быстропротекающих процессов // Издательство: Наука. 466 стр. 1964.
- В.Ф. Климкин, А.Н. Папырин, Р.И. Солоухин. Оптические методы регистрации быстропротекающих процессов. // Изд-во Наука, Сибирское отделение. 205 стр. 1980.
- S. Takeuchi; Y. Ohno; K. Mitsui; M. Arai. High-speed and high-resolution color imaging system combined with multispectral optics: optical layout and feasibility study for combustion analysis. // Proc. SPIE 5580, 26th International Congress on High-Speed Photography and Photonics. p.261-270. 2005.
- G. N. Kulipanov, N.G. Gavrilov, B.A. Knyazev, E.I. Kolobanov, V.V. Kotenkov, V.V. Kubarev, A.N.Matveenko, L.E. Medvedev, S.V. Miginsky, L. A. Mironenko, V. K. Ovchar, V.M. Popik, T.V. Salikova, M.A. Scheglov, S.S. Serednyakov, O.A. Shevchenko, A.N. Skrinsky, V.G. Tcheskidov, N.A. Vinokurov. Research Highlights from the Novosibirsk 400 W average.// Terahertz Science and Technology, Vol.1, No.2, 2008, pp.107-125.
- http://www.ophiropt.com/laser-measurement-instruments/beamprofilers/products/industrial-applications/the-cameras/pyrocam

- Vitaly V. Kubarev, Grigori M. Kazakevitch, Young Uk Jeong, Byung Cheol Lee. Quasi-optical highly sensitive Schottky-barrier detector for a wide-band FIR FEL. Nuclear Instruments and Methods in Physics Research A 507 (2003) 523– 526
- Васильев А.А., Пальчиков Е.И., Кубарев В.В., Чесноков Е.Н., Кошляков П.В., Долгих А.В., Красников И.Ю. Исследование нестационарных волн горения и детонации водородо-кислородной смеси в оптическом и терагерцовом диапазонах. Известия РАН. Физика. 2015, том 79, № 1, с. 196–202.