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A Raman Laser Radar for Determination of Gaseous Pollution in the Atmosphere

A Raman laser radar device has been designed as an instrument for measuring the gaseous pollution of the atmosphere.

The last achievements in development of high power pulsed lasers rendered a possibility of carrying out spectroscopic investigation of remote physical subjects based on Roman scattering phenomenon [1,2]. These investigations supported by the laser radar technique evolved during the last years - called lidar - find an increasing application in measuring the atmosphere pollution by gaseous chemicals. As a basic source of information the lidar technique employes the Mie and Ravleigh scatterings, for which the frequency of the signal associated with the scattering process is identical with that of emitted laser signal. This is the most advantageous situation from the energetic view point. Only the distribution and concentration of both aerosol and gaseous components of the atmosphere can be concluded from the analysis of this signal without the possibility of determination of its chemical composition. Actually, some attempts are made to measure the absolute concentrations by using several wavelength of the exciting laser.

The chemical properties of the scattering molecules may be extracted by analysing the scattered signals of wavelength different from that of the exciting signal and generated by Raman scattering process [3,4]. All the molecules coexisting in the atmosphere cause the Raman scattering. A typical Raman spectrum of a molecule is presented in Fig. 1. This is a spectrum of nitrogen molecules at 300 K temperature. Here, the horizontal axis presents the differential cross-section of back--scattering. As it is easily seen the Raman spec-



Fig. 1. Raman spectrum of nitrogen molecules at 300 K temperature

trum is composed of three branches: O, Q and S. The branches O and S show a distinct microstructure. The band Q exhibits the rotational structure similar to that of bands O and S but the spacing between the spectral lines is relatively very small. All the lines within the Q band are distinguishable only if applying spectroscopes of very high resolving power. For instance, the band width for CO_2 is equal, to 0.3 cm^{-1} while the linear spread of the bands O and S is ten times as high. The total Raman spectrum width is usually greater than 100 $\rm cm^{-1}$ including all the three bands. Besides, the cross-sections of bands O and S change with the temperature, while band Q is practically insensitive to variation of temperature. Therefore, the detection is usually restricted to the Q band. The last limitation results in reduction

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of the bandwidth, while the effective cross-section is also reduced. The change of cross-section for CO_2 amounts to about 10 % in this case.

The Raman frequencies depend on the species of molecules and on the frequency of the exiting laser; the shift of frequency, however, is a constant value depending only on the molecule species. Fig. 2 shows the Raman shift for the components of atmosphere and the most frequently occuring gaseous pollutions. From Eq. (1) it follows that the backscattered Raman radiation intensity is proportional to the absolute concentration of the substance. However, the results are usually receivable in the form of relative concentration estimated with respect to nitrogen with the same coefficients depending on apparatus constants.

The block diagram and principle of operation of the Raman lidar designated for remote examination of the atmosphere composition and its pollution are presented in Fig. 3. The



Fig. 2. Raman shift for atmospheric components and the most frequently occuring gaseous contaminations

The intensity of the Raman backscattering is given by the expression

$$I_{\text{Ram}} = \left(\frac{d\sigma_{\text{Ram}}}{d\Omega}\right) \frac{N_0(R)T_1T_2}{R^2} \frac{c}{2} \frac{\nu_1}{\nu_2} W, \quad (1)$$

where

W — total energy of the laser pulse,

R – distance from the polluted air,

 $N_0(R)$ – concentration of pollution,

 $d\sigma \operatorname{Ram}/d$ – differential cross-section for Raman scattering,

 T_1 , T_2 – atmospheric transmission for radiation of frequencies v_1 and v_2 .

The received radiation comes from a region of length equal to half of the laser pulse length. In order to simplify the calculation the expression $N_0(R)$ and R^2 are usually assumed to be constant for definite distances. laser beam, passes first the collimating telescope, hits the medium under test and becomes subject to non-selective Rayleigh and Mie scattering as well as scattering on gaseous components of the atmosphere with the Raman--shifted frequencies v_1, \ldots, v_n . These spectral components are simultaneously detected by spectrum analyzer applied to a filtering device and a sensitive detector system.

The received power P_r of the Raman component of frequency v_r scattered by the medium distant by R gives the general laser radar equation:

$$P_r(v_r, R) = P_0(v_0) \times$$

 $\times t KT(v_0) T(v_r) A_r Y(R) N(R) (d\sigma/d\Omega) R^2, \quad (2)$ where

 $P_0(\nu_0)$ – transmitted pulse power,



Fig. 3. Principle of Raman Lidar operation

t - half time of the laser pulse duration, k - total efficiency of the system,

 $T(r_0)$ – transmission of the atmosphere for radiation of frequency r_0 .

 $T(v_r)$ — transmission of the atmosphere for radiation of frequency v_r ,

 A_r – effective aperture of the receiver,

Y(R) — geometrical factor taking account of an overlap of the transmitting and receiving channels,

N(R) – concentration of the component under test,

 $d\sigma/d\Omega$ — differential cross-section of either the examined atmosphere or pollution due to Raman back-scattering.

The measurement by Raman lidar is reduced to detecting the frequency in the return signal, which corresponds to the Raman-spectrum shift stimulated by the examined substance and the concentration measurement of the substance [5]. At present two measuring methods are used in Raman lidars. The first one applied more frequently consists in adjusting the Raman lidar detecting system to the reception of one or several precisely defined Raman frequencies. Thus, one or more substances contained in the atmosphere are detected simultaneously. In this way the concentrations measurements of nitrogen, oxigen, water vapour, carbon dioxide, sulphur dioxide, nitrogen oxide and others were made. Simultaneously, the profile of the substances distributions were measured.

Another method [6,7] much more difficult in application consists in an analysis of the complete frequency spectrum starting with exciting laser frequency toward longer wavelengths. In this way all the substances included in the atmosphere may be detected. This is essentially a remote Raman spectrophotometer working as a chemical analyzer of the atmosphere.

The results from Raman lidar are usually obtained in the form of relative concentration from which the absolute concentration should be calculated which is expressed as a number of polluting molecules per one million of atmosphere molecules. For this purpose exploited is the fact, that the concentration of the basis components of the atmosphere i.e. nitrogen and oxygen are known and exhibit very high stability. The method of determination of the examined substance concentration consists in referring the examined Raman line intensity to the Raman line of nitrogen or oxygen.

Basing on the methods mentioned above a laboratory measuring setup has been build as presented in Fig. 4. The medium was excited by the neodymium glass laser in the following setup: generator amplifier, and KDP crystal for second harmonic generation. The pulse du-



Fig. 4. Laboratory measuring setup of Raman Lidar

ration was about 20 ns while energy amounted to about 0.7J. Two Newtonian telescopes were used as receiving and transmitting optical systems. The return signal of Raman scattering was displayed on an oscilloscope screen. The experiments carried out were based on the first method of analysis with a set of interference filters. In the course of examination the frequences corresponding to Raman scattering on N_2 , CO_2 and SO_2 molecules were detected by using a single giant pulse.

In order to have a possibility of performing the complete spectrum analysis of atmospheric gases and measuring the profiles of their concentration a new Raman lidar system with a YAG crystal laser of repetition frequency 50 Hz and recording the results in a X - Ystrip chart recorder is currantly under construction.

The Raman laser system may find application for atmospheric contamination analysis as well as atmosphere temperature determination. Independently, an analysis of other difficult media like combustion products, toxic media, and plasma discharge media and so on may also be performed [8].

Radar à laser (de Ramanoff) pour examiner la pollution de l'atmosphère par les gaz

On a construit un radar à laser pour examiner la pollution de l'atmosphère par les gaz. Le milieu est stimulé par le laser sur le verre néodymique dans le système composé d'un générateur à étranglement passif, d'un amplificateur et d'un cristal KDP pour transformation en deuxième harmonique. Les systèmes émetteur et récepteur étaient constitués par des télescopes dans le système de Newton. Lors du travail avec une seule impulsion gigantesque dans le signal retournant on a découvert des fréquences correspondant à la dispersion de Ramanoff par les particules N_2 , CO_2 , SO_2 .

Лазерный рамановский радиолокатор для исследования газовых загрязнений атмосферы

Для исследования газовых загрязнений атмосферы был создан лазерный рамановский радиоллокатор. Среда возбуждается лазером на неодимном стекле в системе, состоящей из генератора с пассивным сужением, усилителя, кристалла KDP для преобразования на вторую гармонику. Перадающую и приёмную системы представляли собой телескопы в системе Нютона. При работе с единичным импульсом гигантом в возвратном сигнале были выявлены частоты, соответствующие рамановскому рассеянию частицами N₂, CO₂, SO₂.

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