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DEVELOPMENT OF FOURIER SPECTROMETERS IN THE SOVIET UNION

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Abstract—The review covers the range of Fourier spectrometers designed and developed in the Central Bureau for Unique Instrumentation of the U.S.S.R. Academy of Sciences. Included are the UFS-02 high resolution instrument, the FS-01 versatile spectrometer, the LSFS-01 laboratory spectrometer for submillimetre range, the AFS-01 automated photoelectric unit and the BFS-01 fast-scan spectrometer. The performances and capabilities of these instruments are illustrated by experimental spectra.

Instruments with high spectral characteristics are very much in demand by practitioners of the spectroscopy of gases and crystals. Performance specifications for their spectrometers are especially severe where it is required to measure faint spectra of weak sources over a wide range of wavelengths and within a short time. In situations where in addition a high resolution of the spectrum was a requirement, the difficulties at one time seemed unsurmountable. However, recent advances in computing technology have opened new avenues for improvement of spectroscopic techniques. In particular, Fourier spectroscopy has been transformed thereby.

This paper is devoted to the development of Fourier spectrometers in the Central Bureau for Unique Instrumentation of the U.S.S.R. Academy of Sciences, Moscow.

The high resolution Fourier spectrometer UFS-02 was developed for investigation of the spectra of atoms, ions and molecules in gaseous or a condensed phase at low temperatures. It resolves lines as close as 0.005 cm^{-1} in the range from 1 to 100 μm [1–3].

The optical and mechanical units of this spectrometer are housed in separate vacuum enclosures whose residual pressure is about 10^{-3} mm Hg. They comprise a source monochromator block, an interferometer, an optical distributing device, a multipath gas cell (vacuum up to 10^{-5} mm Hg) and a reference channel unit.

An interferometer is the key optical unit of any Fourier spectrometer for it controls the spectral characteristics of the instrument. The optical scheme of the interferometer incorporated in the UFS-02 spectrometer is shown in Fig. 1.

In the measurement channel, the collimated light beam launched by the source-monochromator assembly is directed to the beam-splitting part of the beam-splitter-compensator plate. The reflected part of the beam divided by the beam splitter proceeds to a stationary reflector of the cat's eye type. Having passed the reflector and the compensating part of the beam-splitter-compensator, the beam retro-reflected by a plane mirror is again incident on the beam splitter. Here it is mixed with the part of the divided beam that has taken a similar optical path in the other arm of the interferometer, which is equipped with a movable reflector. This interfacing beam is sent from the interferometer to the optical distributing device.

The specific features of this interferometer are listed below.

(1) The cat's eye reflectors sharply reduce the amount of movement needed in the movable reflector mechanism.
(2) The beam splitter and compensator combined in a single plate ensure identity of material, thickness and wedge angle.
(3) The use of auxiliary plane mirrors allows two-fold expansion of optical path difference in preference to the classical arrangement, for the same mechanical motion of the reflector [3, 4].

To cover the entire spectral region, the spectrometer is provided with a set of exchangeable beam splitters of KI grade quartz, CaF_2, KBr and a 6-μm polyethylenterephthalate film.

The system of automatic control and spectral data recording is designed to control the scanning of optical path difference, separate signals from noise, sample and evaluate the measured signal, perform filtering, record into the buffer store, and monitor connections with the computer. The system design takes into account that this spectrometer is based on continuous scanning (movement

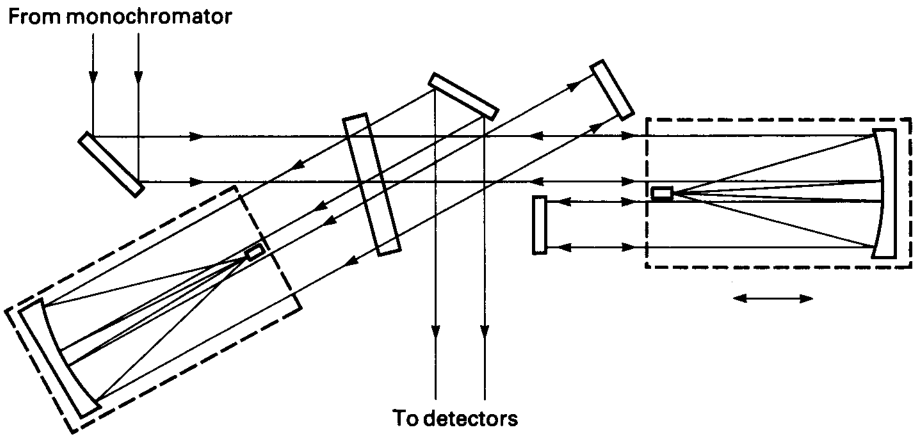


Fig. 1. Optical system of the UFS-02 Fourier spectrometer.

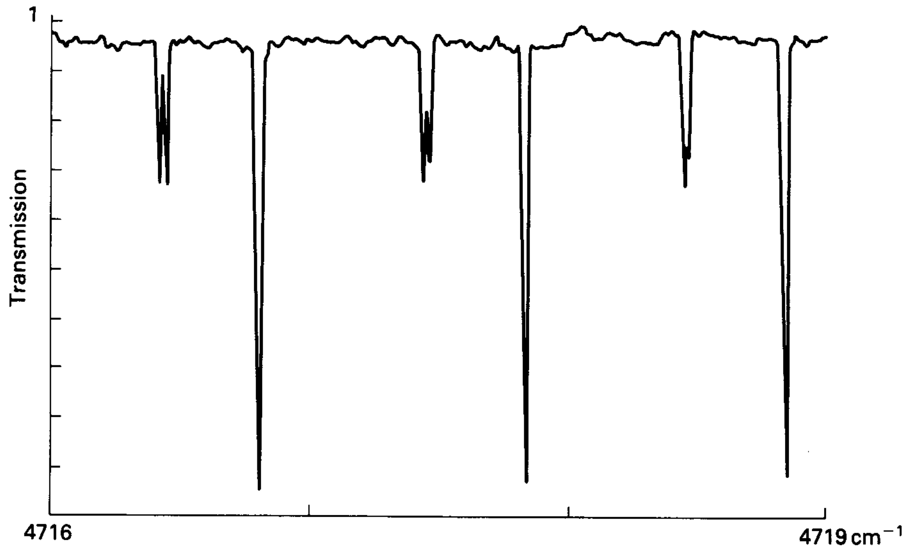


Fig. 2. Absorption spectrum of N_2O.

of a reflector at a constant speed) and accumulation of interferograms (multiple recording of interferograms and their digital addition) for more efficient utilization of experimental time and higher signal/noise ratios in the recovered spectra. Specifically, this system comprises a system for recording measured signals, a block generating a pulse of zero path difference, a module for sensing reference pulses, an automatic control module and a software package based on the UVK SM-3 operating system.

The resolution of the UFS-02 spectrometer was tested with the absorption spectrum of N_2O in the 4700 cm^{-1} range (Fig. 2).

The spectrometer was also used to study absorption spectra of vapourized atoms of Tl and Tm in an inert gas atmosphere, and absorption spectra of rare earth ions in crystals [6]. Gaseous Tl and Tm were obtained by evaporation at 1400–1500 K in special cartridges filled with various inert gases at different pressures and heated externally. Experimental measurements were carried out with the shifts of forbidden lines of Tl and W atoms and for the broadening of these lines as a function of the inert gas pressure (Fig. 3).

It is worth noting that the spectra obtained in Fourier spectrometers are blessed with a convenient scale of wavenumbers. A precise scale of wavenumbers that is established for the whole range with reference to one laser source is a salient feature and advantage of the method (Connes gain).

This feature of the wavenumber scale facilitated the measurements of the positions of Stark components of the levels $4_{I_{15/2,13/2,11/2}}$ of Fr^{3+} in YAG accurate to within 0.03 cm^{-1} (Fig. 4). This

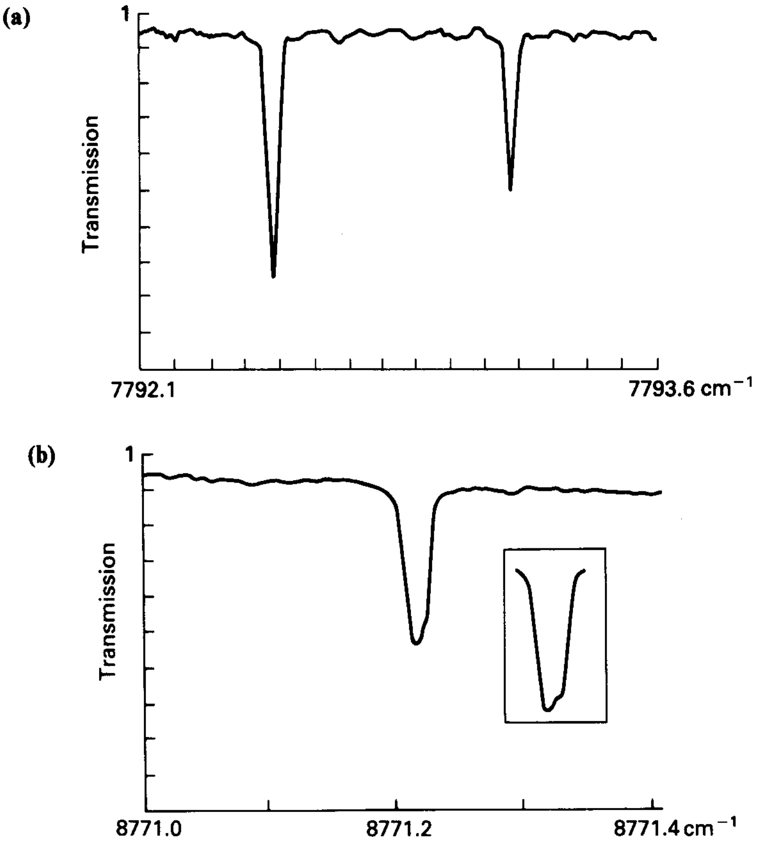


Fig. 3. (a) Absorption spectrum of thallium (Tl) vapor. (b) Absorption spectrum of thulium vapor.

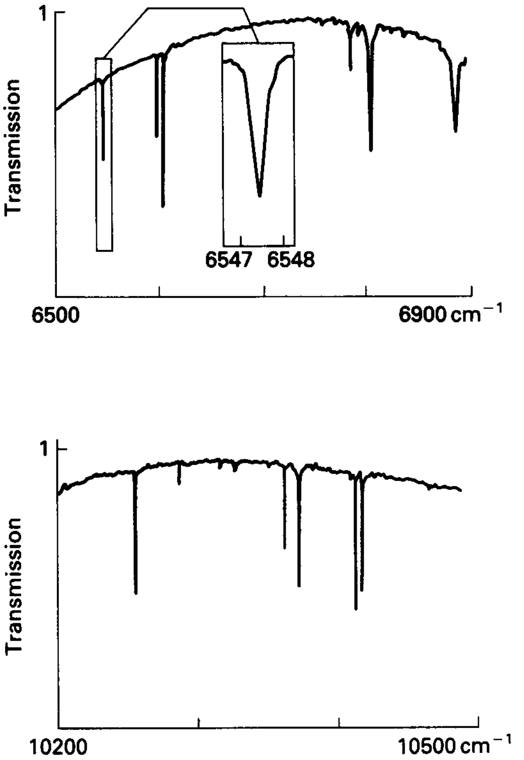


Fig. 4. Absorption spectrum of YAG–Er crystal (0.03%).

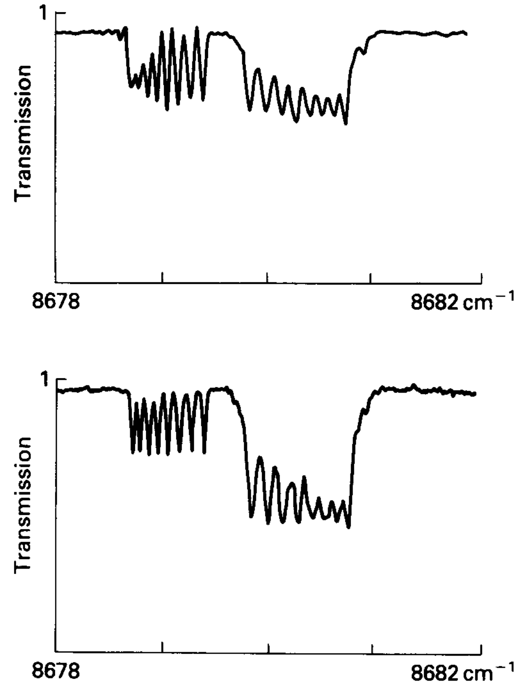


Fig. 5. Segment of transmission spectrum of $LiYF_4$–Ho crystal (1 atomic%) and SNR enhancement.

accuracy was in locating peaks of lines having a halfwidth of 0.3 cm^{-1}. $Y_3Al_5O_{12}$–Er is an effective material for lasers in the range of 3 to 1.5 μm. The above levels participate in lasing, so that the relevant spectroscopic information is of great significance [7].

A nuclear superfine structure was revealed in the optical spectrum of a holmium doped crystal of $LiYF_4$ [2, 8]. Figure 5 shows such a spectrum with a well-resolved superfine structure. This structure may be treated as Zeeman splitting of a degenerate electronic Stark level of Ho^{3+} in the magnetic field of a nucleus, which is quantized in agreement with the projection of the nuclear momentum on the axis of symmetry. The linewidths measured in the spectrum of holmium implanted in the crystalline lattice were not wider than those in spectra of atomic holmium.

In response to a wide range of problems of spectroscopic nature arising in physics and chemistry the Central Bureau for Unique Instrumentation has developed a family of moderate resolution Fourier spectrometers.

The FS-01 Fourier spectrometer [9, 10] is designed for spectral investigations in the range 100 to 5000 cm^{-1}, with a resolution down to 0.1 cm^{-1}. It is operable with light transmitted through (absorbed in), reflected or emitted from specimens in gaseous, liquid or solid state.

The automated photoelectric spectrometer AFS-01 [10] is designed for chemical analysis of electrically active impurities in pure and extra-pure semiconducting materials. It can also be used for investigating the optical properties of solids at low temperatures and in magnetic fields in the spectral range from 25 to 5000 cm^{-1} with a resolution of down to 0.1 cm^{-1}.

The LSFS-01 laboratory submillimetre Fourier spectrometer [10] is designed for verification and adjustment of the submillimetre equipment employed for passive and active diagnostics of plasma and for spectral investigations in the range 3–300 cm^{-1} with a resolution of down to 0.05 cm^{-1} at different polarizations.

The development of these spectrometers was underlain by a number of common principles, and involved a number of common elements, constructions, operating techniques, etc., which have led to largely unified constructions that may be easily readjusted when changing from one problem to another. The common principles include those of fast scanning, and the accumulation of interferograms. Among the common units are a unified module of movable reflector travel and a single system of automatic control and spectral data recording. The basic employer of these units is the FS-01 Fourier spectrometer which will be considered in some detail below.

The optical system of FS-01 is represented schematically in Fig. 6. It comprises individual modules

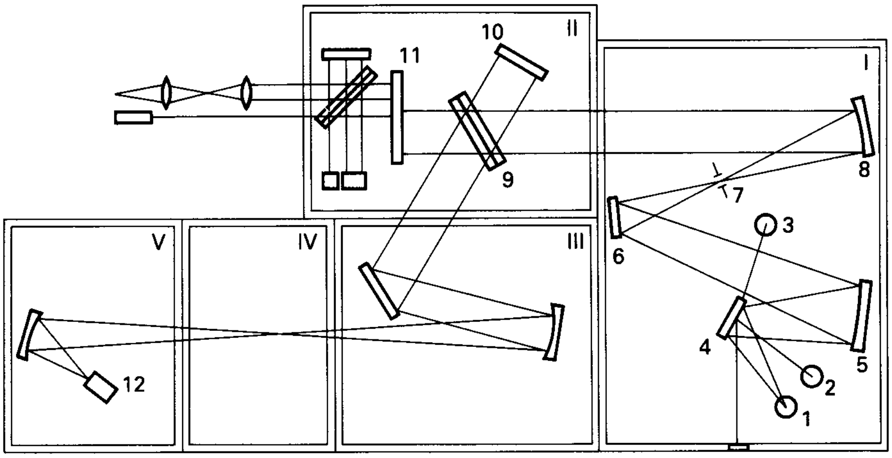


Fig. 6. Optical system of the FS-01 Fourier spectrometer.

encapsulated in vacuum housings. These modules are the illuminating unit (I), interferometer (II), cell compartment (IV) and detector compartment (V).

The illuminating beam module comprises a spherical mirror (5), plane mirrors (4, 6), an iris diaphragm (7), and an off-axis parabolic mirror. These elements form and direct into the interferometer a collimated IR beam emanating from any one of the three sources (1–3).

The interferometer arrangement consists of two separate interferometers with a common drive for the movable mirrors. The principal, or measuring, interferometer (9–11) is set up in the Michelson arrangement with the angle of incidence of rays on the beam splitter equal to 30°.

The mirror travel assembly provides a range of optical path differences up to 10 cm. The rate of scanning can be varied over a wide range so that interferograms may be recorded over the whole spectrum at frequencies matched with the passbands of the IR detectors. The shortest time to scan a spectrum with a resolution of 0.1 cm^{-1} is 2.5 min.

To cover different segments of the spectrum, the instrument is equipped with a set of compensator-beam splitters (9) of grade KI quartz, fluorite (CaF_2), and KBr. The range 25–100 μm is served by a beam splitter made as a 6-μm polyethylenterephthalate film.

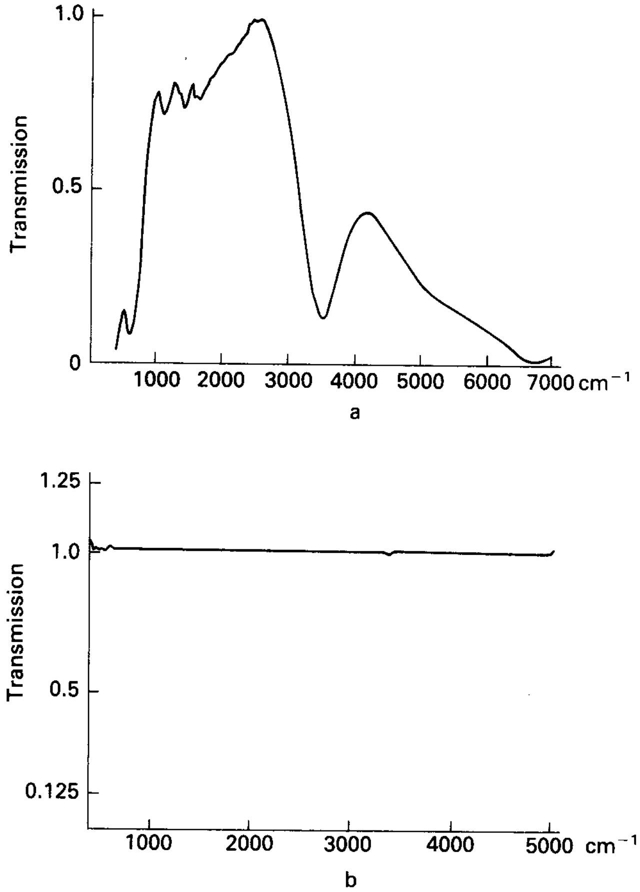
From the principal interferometer the measured beam is directed into the cell compartment (IV) made as a single evacuated module and then to the detector compartment (V) to be received by the appropriate detector (12). The basic set of detectors for the IR range include a pyroelectric detector and an opto-acoustic receiver (OAP-5M) with a polyethylene window.

Figure 7 portrays the transmission spectrum of the FS-01 instrument with beam splitters of KBr, recorded with a resolution of 2 cm^{-1}. The bottom curve shows the reproducibility of this spectrum. The reproducibility of this instrument in transmission measurements is unity, accurate to within 1%. The signal/noise ratio in this spectral range is about 200 on the average and reaches 1500 (with 36 scans) at the best segments.

The high resolution and spectral coverage of this Fourier spectrometer is illustrated by the spectrum of industrial grade methane (Figs 8–11) which contains water and carbon dioxide. This spectrum was recorded with a resolution of 0.1 cm^{-1} over 1 h by accumulating 36 interferograms. The SNR is not worse than 100 over the whole range. It can be seen that a single run yielded the absorption spectrum of methane over the range 1250–1400 cm^{-1}, that of water over the range 1620–1720 cm^{-1}, that of carbon dioxide over the range 2300–2400 cm^{-1}, and that of methane in the range 2800–3200 cm^{-1}.

As for the AFS-01 Fourier spectrometer (Fig. 12), as in other instruments above, its optical system is composed of separate functional modules housed in vacuum casings. The key modules include an illuminating module (I), interferometer arrangement (II), cell compartment (IV), optical distributing arrangement (V) and cryostat (VI).

The optical system of the illuminating beam compartment is built to the same scheme as in the FS-01 instrument. To expand its operating range, the AFS-01 unit is equipped with two additional beam splitters of 12-μm and 25-μm polyethylenterephthalate film.



a

b

Fig. 7. (a) Transmission spectrum of the FS-01 unit with KBr beam splitter. (b) Corresponding spectral reproducibility.

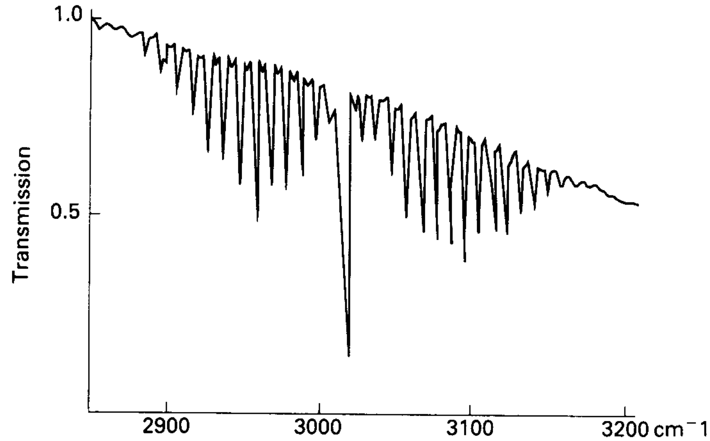


Fig. 8. Absorption spectrum of methane.

Because this instrument has different tasks to do, the optical system in the cell compartment and in the detector compartment differs from that of the FS-01 counterpart.

The optical distributing system forms and directs the light beam onto a detector or into the photoconductivity cryostat (VI). The IR detectors are the OAP-5M opto-acoustic detector with a polyethylene window, and the PM-5 pyroelectric detector. The measurement channel beam is launched into the cryostat through a conical lightguide made of stainless steel and falls on the specimen placed on a sapphire substrate in a copper holder. The holder along with the specimen,

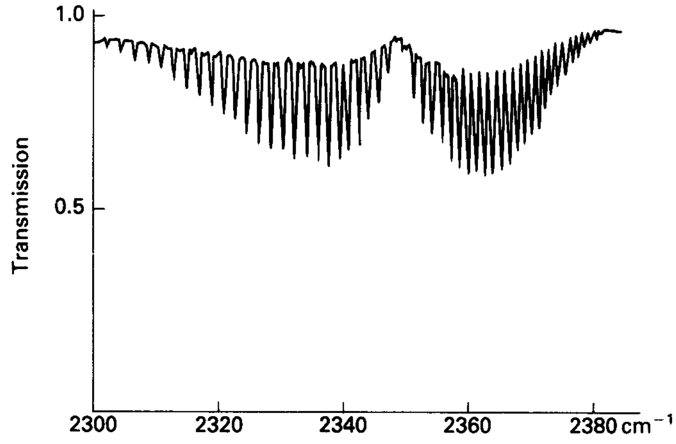


Fig. 9. Absorption spectrum of CO_2.

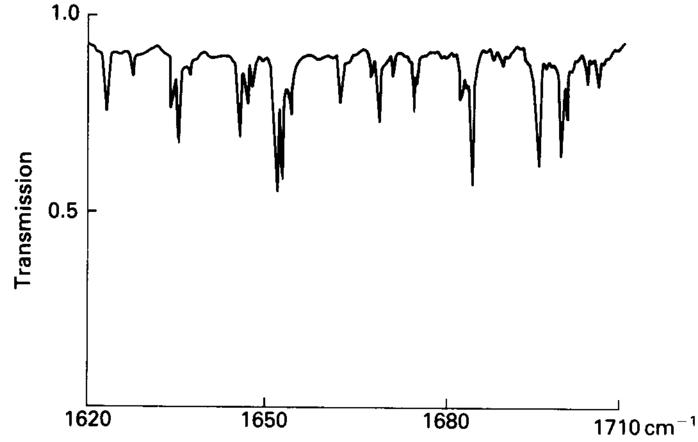


Fig. 10. Absorption spectrum of water.

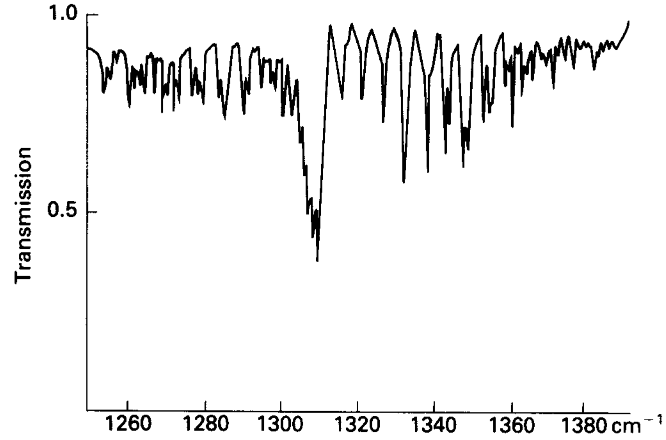


Fig. 11. Absorption spectrum of methane.

a Hall cell, a resistor heater, and resistance thermometers is placed inside a chamber filled with helium, which is inside a helium cryostat with a superconducting magnet. The temperature of the specimen varies from 4.2 to 120 K and can be maintained accurately within 0.5 K.

The alloy $Hg_{1-x}Cd_xTe$ ($x = 0.2$) with its forbidden zone width, $E_g = 100$ (800 cm^{-1}) is a promising material for IR photodetectors in the range 8–14 μm. It was investigated in the AFS-01 spectrometer

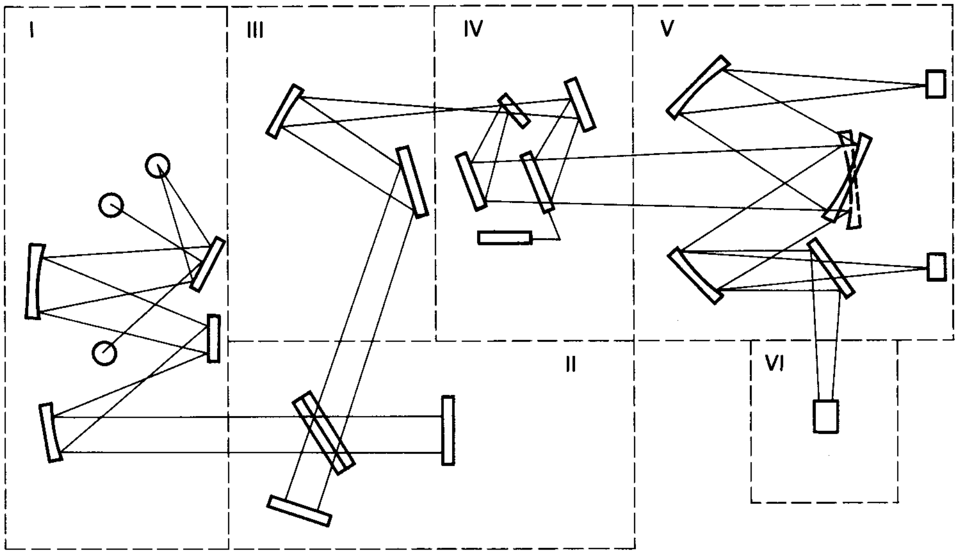


Fig. 12. Optical system of the AFS-01 automatic photoelectric spectrometer.

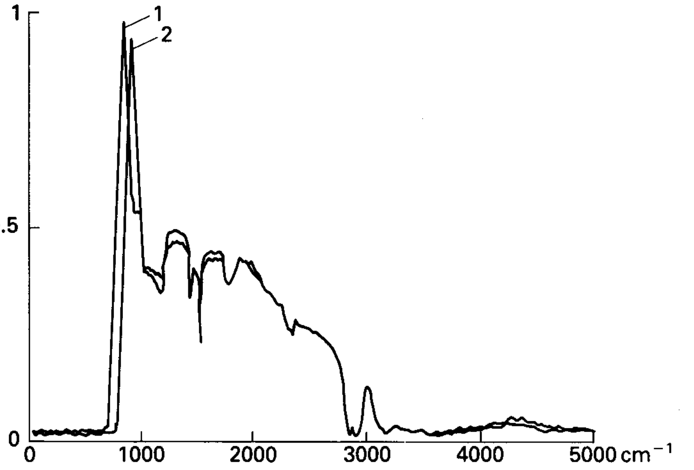


Fig. 13. Photoconductance spectrum of $Hg_{0.8}Cd_{0.2}Te$.

in the photoconducting mode. The position of the absorption cutoff was studied as a function of temperature (Fig. 13). The temperature coefficient of the forbidden zone width was established to be

$$\frac{\partial E_g}{\partial T} = 0.465 \text{ meV K}^{-1}.$$

The last but one spectrometer in our list—the LSFS-01 unit—has a polarization interferometer constructed to the Martin–Paplette layout which is more efficient for its range than other systems. The optical system of this interferometer (Fig. 14) employs a wire grid which effectively splits the polarized light beam for all wavelengths exceeding the period of the grating, thus covering the entire spectrum in the range 3–300 cm^{-1}. This optical scheme allows additional investigation of specimens in polarized light with two mutually orthogonal polarizations.

Figure 15 represents the emission spectrum of a backward wave tube recorded with a resolution of 0.05 cm^{-1}.

The optical scheme of the spectral radiometer BFS-01 (Fig. 16) is essentially a combination of two polarization interferometers of the Martin–Paplette type with a single simultaneous scanning facility. The O-mode energy of the studied radiation is directed to one interferometer, and the E-mode energy to the other. The movable and stationary reflectors are made as dihedral mirrors.

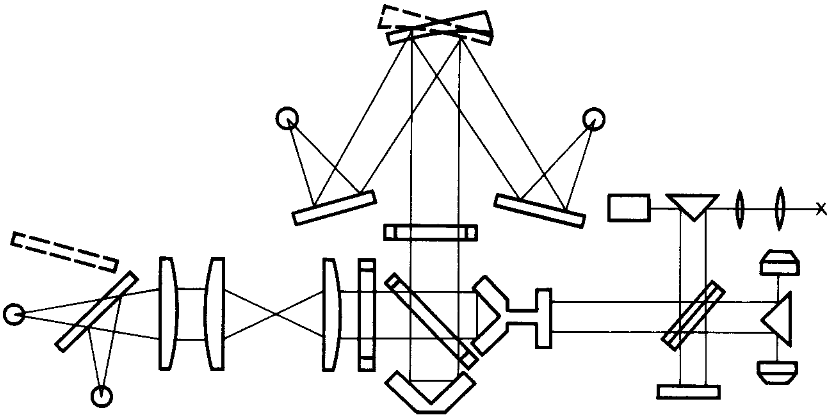


Fig. 14. Optical system of the LSFS-01 laboratory Fourier spectrometer for submillimetre range.

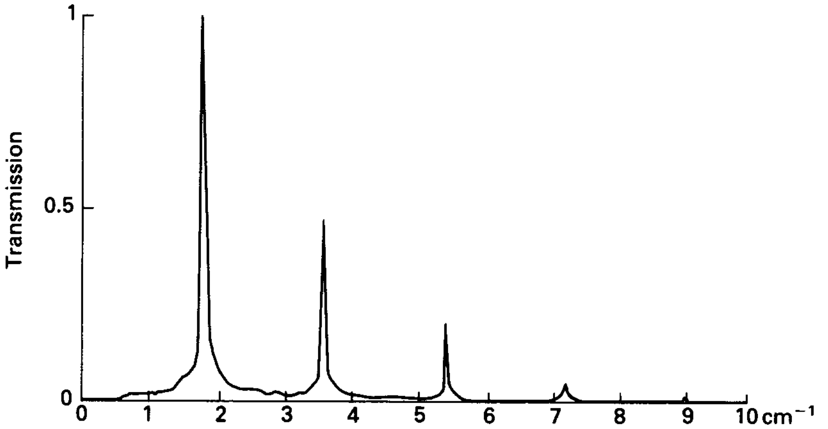


Fig. 15. Emission spectrum of backward wave tube.

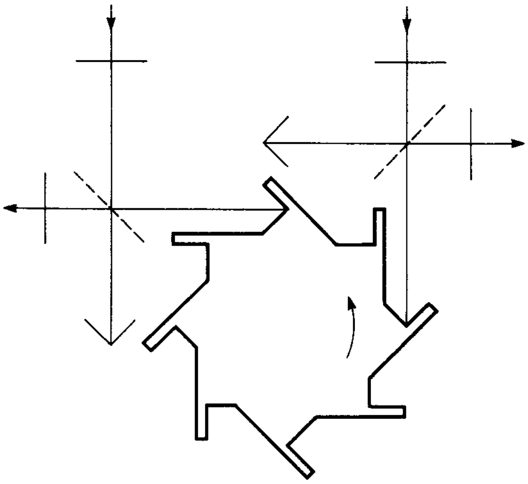


Fig. 16. Schematic diagram of BFS-01 optical system.

The polarizer, analyser and beam splitter are made as fine, one-dimensional, tungsten-wire gratings with 60-μm period.

The scanning facility [11] consists of eight dihedrals fixed on a single platform. When the platform is rotated, the dihedrals come in turn into the operating range of the scan, taking care of the optical path difference in the arms of the interferometers. Using dihedral mirrors as reflectors enables the

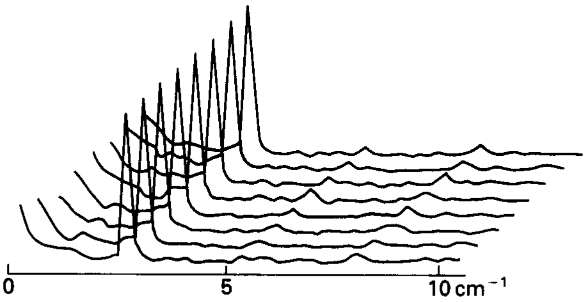


Fig. 17. Emission spectrum of a backward wave tube, $T = 0.005$ s.

incident and reflected beams to be parallel to the plane bisecting the angle between the planes of the dihedra.

The detectors are InSb probes cooled by liquid helium.

The BFS-01 spectral radiometer has no unit for direct measurement of optical path difference. For this purpose it uses a train of pulses generated by an internal generator while the mirror platform rotates at a constant speed. At a rotation rate of 800 Hz, the instability of platform rotation rate amounts to about 10^{-4}. By way of example, Fig. 17 shows the spectrum of emission of a backward wave tube recorded with a resolution of 0.1 cm^{-1} at successive intervals of 0.005 s.

The collection of Fourier spectrometers developed in the Central Bureau for Unique Instrumentation, Moscow (UFS-02, FS-01, AFS-01, LSFS-01, BFS-01) possesses all the advantages basic to Fourier spectrometry. The principles of continuous scanning (Fourier frequency modulation) and accumulation of interferograms very much reduce the experimental time required to achieve the desired signal-to-noise ratio. The computer integrated within the instrument not only performs Fourier transformations and controls the spectrometer but also carries out secondary processing of spectra and assists the experimenter in interpreting the results. These spectrometers have been used for solving numerous problems of physics, chemistry, biophysics and materials science, specifically in studies of the optical characteristics of high temperature superconductors [12].

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