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Spectrometr for investigation of Mandel'shtam-Brillouin light scattering*

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Construction and operation of a Fabry-Pérot interferometer built for investigation of Mandel'shtam-Brillouin spectroscopy in liquids have been described.

Introduction

High resolution Mandel'shtam-Brillouin spectroscopy, after introduction of laser excitation [1-3], became quickly a valuable technique in studies of thermal and transport properties of matter. Its important applications are measurements of velocity and attenuation of longitudinal thermal waves. The vastly increased frequency region, now available for these studies (up to several tens of GHz), made it possible the search of dispersion of hypersonic waves and investigation of several types of relaxation processes which may occur in liquids. This kind of spectroscopy is still a very powerful method for investigations of various dynamical processes in liquids ([4-8], and many others papers).

Polarized collimated monochromatic light scattered from a transparent material typically shows a frequency spectrum consisting of a Rayleigh component centred at the original frequency, and two symmetrically displaced Mandel'stham-Brillouin components. The latter are due to Bragg scattering of light by accoustic waves caused by density fluctuations in the medium. If the acoustic mode has a wave-vector $\vec{q}$, the Bragg condition is given by

$$\vec{q} = \vec{k}_s - \vec{k}_0,$$

where $\vec{k}_0$ and $\vec{k}_s$ are incident and scattered wave-vectors, respectively Since $k_0 \approx k_s$, we have

$$\nu_{M\text{-}B} = \frac{2vn}{c}\,\nu_0 \sin(\theta/2), \tag{2}$$

where $\nu_{M\text{-}B}$ is the frequency shift of the Mandel'shtam-Brillouin lines, v — velocity of the hypersound, ν_0 — frequency of the incident light, θ — scattering angle, n — refractive index of the scattering material, c — velocity of light.

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In general, as classical theory predicts [9], frequency distribution of the scattered light is in the form

$$I(\nu) \propto \frac{B}{1+\left(\dfrac{\nu-\nu_0 \pm \nu_{M\text{-}B}}{\Delta\nu_{M\text{-}B}/2}\right)^2} + \frac{R}{1+\left(\dfrac{\nu-\nu_0}{\Delta\nu_R/2}\right)^2}, \qquad (3)$$

where $\Delta\nu_{M\text{-}B}$ is the half width at half intensity points of the Mandel'shtam-Brillouin lines, $\Delta\nu_R$ is the width of the Rayleigh component, B and R are constants.

The spectrometer built in out laboratory allows to determine the spectrum of the scattered light. The spectrometer features were carefully analysed from the viewpoint of the instrumental errors influence on the investigated spectra. The contributions to the instrumental line width coming from: the pinhole diameter, the focal length of lens gathering light from the interferometer, solid state angle of the scattered light, the quality of Fabry-Pérot plates, and the aperture of the interferometer were taken into account. Some remarks on counting these contributions were given. The spectrometer enables a computer development, accommodation and averaging of the spectra without a multichannel analyser commonly used to this purpose. The original computer program for determination of the parameters of real scattered spectra was prepared. The advantages of the spectrometer are: high sensitivity and reliability in comparison with classical recording methods (sensitivity of d.c. detection method is about one order lower), possibility of computer development of spectra with the accuracy comparable with the set-up comprising the expensive multichannel analyser and relatively simple construction.

Apparatus

The experimental set-up is shown in fig. 1. A He-Ne laser operating at $\lambda = 632.8$ nm with maximum output power of about 170 mW was used as a light source. Typical power level employed in light scattering measurements was about 100 mW. Spectral linewidth of the laser light was 1.15 GHz. Laser power was continuously monitored; its fluctuations were less than 1%. In order to direct laser radiation to the scattering cell a prism was employed. Glan prism P was used for additional polarization of light in the plane perpendicular to the scattering plane. Thereupon the light passed through two diaphragms D_1 and D_2. The lens L_1, placed between this apertures, focussed the light beam on the scattering cell. The scattering cell was mounted in a brass chamber with electric resistance heater. The temperature of chamber was electronically stabilized with an accuracy better than 0.05° C. The cell temperature was controlled by Cu–Constantan termocouple. Scattered light limited by the aperture D_3 (diameter Φ_3

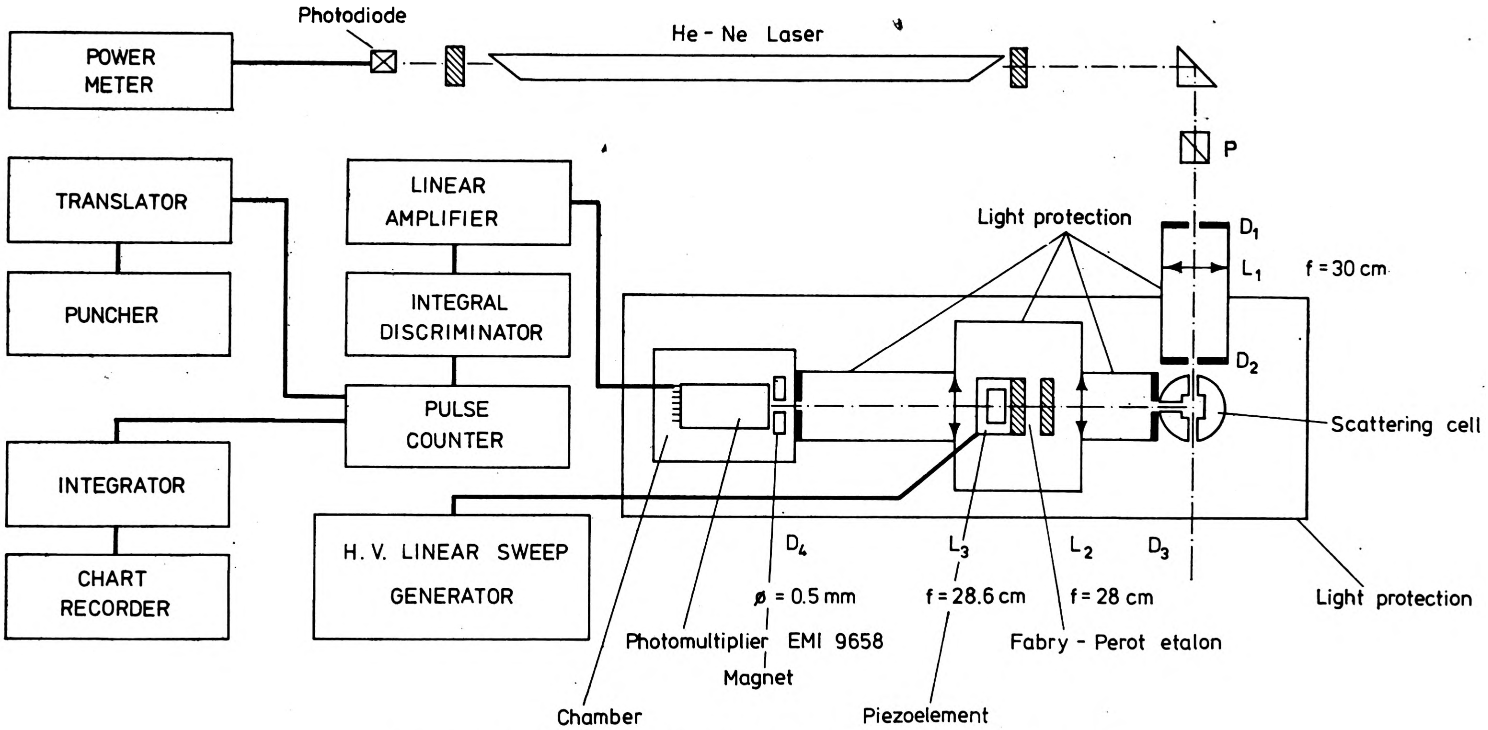

Fig. 1. Block diagram of the Mandel'shtam-Brillouin spectrometer

$= 3$ mm) was gathered by the lens L_2 (focal length $f_2 = 28$ cm). Spectrum of this light was then analized by a piezoelectrically scanned Fabry-Pérot interferometer. Its interference pattern was focussed by using lens L_3 on the pinhole D_4 placed at the entrance window of the photomultiplier. The diameter of the pinhole D_4 was equal to 0.5 mm.

Let us now consider some geometric features of this spectrometer. Scattering angle being $90° + 0.2°$ gave an error up to 0.5 % in determination of the position and width of Mandel'shtam-Brillouin lines. Diaphragm D_3 with a diameter of $\Phi = 3$ mm enables observation of the scattered light within a solid angle of $\Delta\theta \sim 10^{-3}$ sr. This leads to broadening of line by about 30 MHz. This is negligible if compared with the instrumental line width. The width of the instrumental line together with the laser line was about 1.15 GHz. Focal length of the lens L_3 and diameter of the pinhole D_4 were carefully chosen to get the proper value of the so-called scanning aperture [10] defined in angular terms by $\vartheta_a = a/f$ (a — diameter of pinhole, f — focal length of a lens gathering light from interferometer). This scanning aperture determines a rectangular band pass of width ν_a equal to

$$\nu^a = \left(\frac{\vartheta_a}{\vartheta_1}\right)^2 \cdot \nu_{FSR},$$

where $\nu_{FSR} = c/(2nd)$ is the interferometer free spectral range, d is the Fabry-Pérot plates distance, ϑ_1 is the angular diameter of the first bright fringe from a brigth centre, and given by $2\sqrt{\lambda/nd}$. In general, ν_a ought to be less than one fifth of the spectral width of the line studied.

The Fabry-Pérot plates had a flatness $\lambda/60$, reflectivity — 98 %, and effective diameter $\Phi = 45$ mm. All these geometrical and material features gave the theoretical instrumental finesse equal to 20. In practice we have got a little less finesse because of imperfection in adjusting the parallelism of the plates. The free spectral range of the interferometer varying from 100 GHz to 1 GHz in any given investigated case was fixed of about three times larger than the observed displacement of the Mandel'stham-Brillouin lines. This allowed to avoid the overlapping of the lines under investigations. The distance between Fabry-Pérot plates was slowly scanned using a piezoelement driven by a linear sweep generator. The main part of this generator was a Beckman spiral potentiometer, it covered 2.5 M Ω in 40 revolutions with a very high degree of linearity. To obtain a fine adjustment of the potentiometer it was connected in series with 3 parallel 5 M Ω potentiometers which supplied three piezoelectric elements with a voltage ranging from 0 to 600 V. The Beckman potentiometer driven by an electric motor supplied all piezoelements simultaneously with slow sweep-voltage from 0 fo 900 V. The interferometer was scanned through four and half interference orders at full sweeping cycle. The scattered light was detected by photomultiplier EMI 9658. This photomultiplier was cooled down to about 253 K. Further dark current suppression was due to special defocussing magnet applied directly to the entrance window of the photomultiplier. The photomultiplier chamber was placed on a mount having orthogonal motions along the three axes (x, y, z), due to precise adjustment of the input pinhole of the photomultiplier (D_4) on the centre of interference fringes. Light signals were detecded in an one-channel photon counting régime. Output from the ·pulse counter was registered on a punched tape, and developed with computer. A whole procedure for numerical analysis of scattering data was elaborated. Photon pulses from the pulse counter were also directed to an integrator giving analogue voltage proportional to the counts rate. Finally they were recorded on the chart recorder. This enables visual observation on the quality of the spectrum, and allows a proper adjustment of the Fabry-Pérot interferometer.

Interferometer testing data

Preliminary scattering measurements were carried out using benzene (fig. 2) and carbon tetrachloride. The samples were reagent grade.

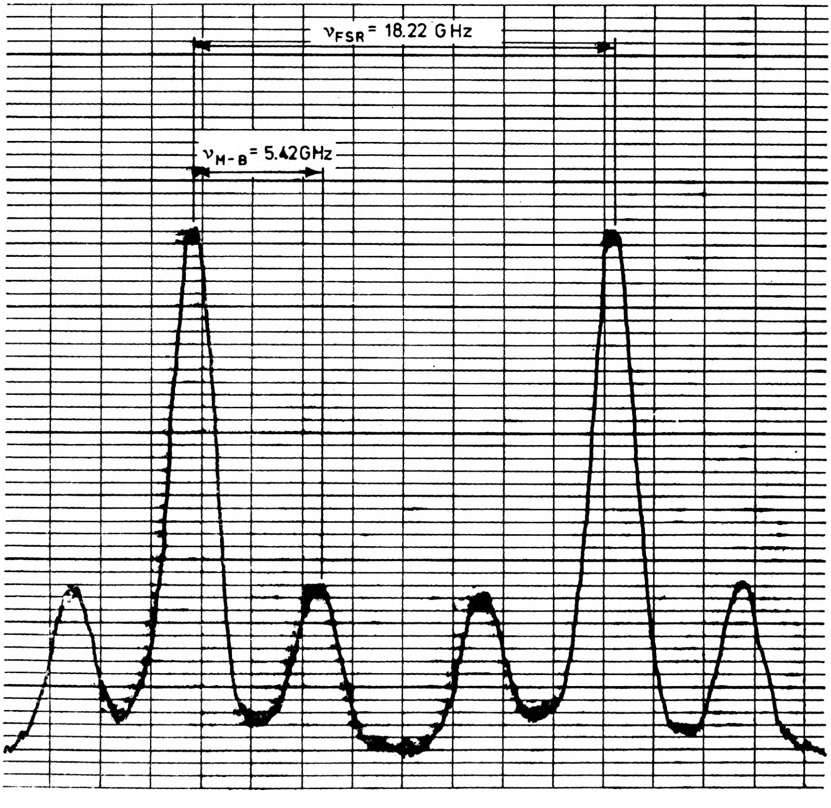

Fig. 2. Mandel'shtam-Brillouin spectrum of benzene at temperature 295.2 K. The free spectral range ν_{FSR} and frequency shift of Mandel'shtam-Brillouin $\nu_{M\text{-}B}$ component were indicated

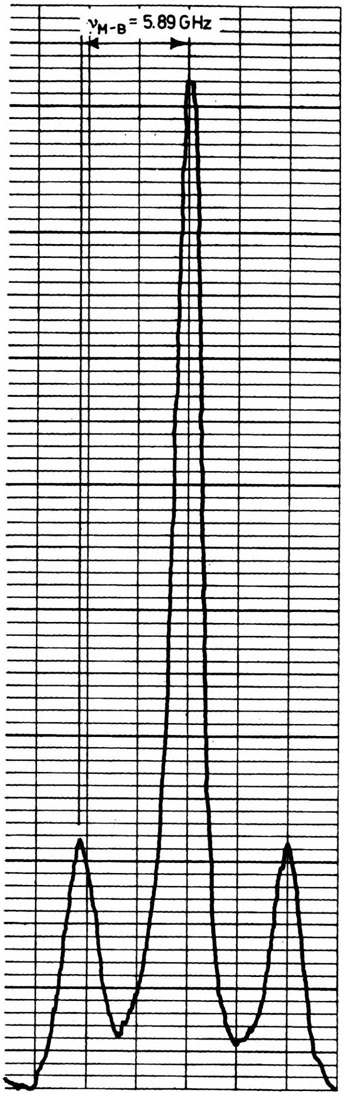

Fig. 3. Spectrum of light scattered in veratrol at temperature t = 300.3 K

To minimize the Tyndall scattering from dust particles, the liquid passed through a 0.15 μm millipore filters directly into the cell. Some preliminary results are shown in table. Values for benzene and carbon-tetrachloride

Table

Liquid	Temp. [K]	ν_{M-B} [GHz]	$\Delta\nu_{M-B}$ [MHz]
C_6H_6	295.2	5.42∓0.08	193
CCl_4	291.2	3.78∓0.05	350
$C_6H_4(OCH_3)_2$ veratrol	300.3	5.89∓0.07	497

are (with an experimental error) in good agreements with those given by others authors [5, 11-13]. Spectral widths of the Mandel'shtam-Brillouin components were also determined [14].

More detailed information concerning the measurements in veratrol (see fig. 3, and table) will be published elsewhere.

Relatively great error in these data results from some inaccuracies in parallelism of the adjustment of Fabry-Pérot plates and laser line width.

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Спектрометр для исследований спектра Мандельштама-Бриллюэна рассеянного света

В работе описан спектрометр для исследований спектра Мандельштама-Бриллюэна рассеянного в жидкостях света. В конструкции использовали: лазер He—Ne большой мощности, интерферометр Фабри-Перо, перестраиваемый пьезоэлектрически, а также электронную систему, работающую как счётчик фотонов. Подробно проанализированы условия работы и параметры спектрометра.

Проведенные исследования в бензоле и четырёххлористом углероде дали результаты, соответствующие данным в литературе.