МИФИ

МОСКОВСКИЙ ГОСУДАРСТВЕННЫЙ
ИНЖЕНЕРНО-ФИЗИЧЕСКИЙ ИНСТИТУТ
(ТЕХНИЧЕСКИЙ УНИВЕРСИТЕТ)

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AT AN ANGLE OF THE FIRST-ORDER RAINBOW CONTROL OF THE SIZE OF SUSPENDED SINGLE PARTICLES BY LIGHT SCATTERING

Moscow 1997

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Methods of particle size control are based on the measurements of the intensity of light scattered by an immobile particle at an angle of the first-order rainbow. Single drops of benzene suspended in water by ultrasound were the object of the study. Modified methods based on the use of the frequency of proper high-quality spheroidal oscillations of the drop as function of its size are considered. Design of the experimental unit is described.

Редактор Н.Н. Антонова
Ответственный за выпуск А.А. Болдырев

Подписано в печать 15.07.97	Уч.-изд. л. 0.75	Формат 60*84 1/16
Печ. л. 0.75 Изд. № 019-97	Заказ 606	Тираж 100 экз.

ISBN 5-7262-0136-1

Study of optical and physical properties of particles suspended in a certain medium is of interest for different fields of science [4]. Development of an efficient method of size control of studied particles in situ is a first-priority task. Size control of suspended particles using light scattering is one of the widely used techniques.

The present paper considers methods of size control of suspended single particles by light scattering at an angle of the first-order rainbow. Rainbow phenomenon has been studied well enough [1-3]. All kinds of beams - reflected ones, exiting at once or after single or multiple reflections - may contribute to the light scattered into a singular solid angle near a certain given direction. As a result of reflection and refraction, initially homogenous spatial distribution of beams falling on the drop may concentrate around certain directions, like beams concentrated behind the lens. Scattering angles at which scattered light intensity sharply increases under the laws of geometrical optics have been named rainbow angles [2-3].

It follows from Eiry's approximation that light intensity *Is* at range *r from the centre of the drop is function of scattering angle* b and is expressed through Eiry's function A(b) squared [1-3] :

$$Is = Io\left(\frac{R}{r}\right)^2 \left[A(b)\right]^2, \qquad (1)$$

where I_0 is intensity of radiation falling onto the drop, *R* is radius of the drop (Eiry's theory is valid for particles with *R* i 500 m at wavelength of the light l = 0.63 m [1]),
b is parameter of Eiry's function. First-order rainbow is related to interference of reflected beams and those refracted beams which had one internal reflection. For this rainbow at a maximum b » 1.08, and "rainbow integral" $A(b)^2$ » 1.005 [1]. Therefore, *R* can be determined from measured values of I_0 *and* I_s *at fixed* r *using expression (1). Rigorous theory Mi should be used for particles of smaller R.*

Angle value for a *k-order rainbow* $b^{(k)}$ in the scatter indicatrix is determined by the following expression [2]:

$$\beta^{(\kappa)} = (\kappa - 2)\pi + 2\left[\varphi - (\kappa - 1\psi)\right] \qquad (2)$$

where

$$\cos\varphi = \frac{n^2-1}{\kappa^2-2\kappa}, \quad n = \frac{\sin\varphi}{\sin\psi}$$

$n = n_i/n_0$ - relative index of refraction; n_i, n_o - refraction indices for the drop and medium, respectively; j - incidence angle for light beam in the outside medium; Y - angle of refraction of incident light beam; positive integer *k characterizes beams, exiting the drop and named as derivatives of the beam which was the first to fall onto the drop. For a first-order rainbow k = 3.*

Stable suspension of a drop in a fixed point of space of an acoustic resonator, when the gravity is compensated by a radioactive pressure from a stationary ultrasonic wave [5], allows to use oscillation of the drop shape [6,7] to raise sensitivity and widen possibilities of the above method of particle size control. In fact, since initiation of drop's shape oscillation is of resonant nature, the amplitude of forced oscillations and therefore the amplitude of light scattered by the drop will increase proportionally to Q-factor at a resonance frequency of a corresponding mode [8-11].

Analysis of forced oscillations of a spherical particle suspended in the field of a flat acoustic wave indicates that excitation of the following major modes of drop oscillations is possible in the first approximation: radial (*l = 0), reciprocating (l = 1) and spheroidal (l = 2); l - spherical function index* [6-8, 12]. With $l = 2$ an oscillating drop has a form of a spheroid once oblate and then extended along the axis. The first approximation is valid if the radius of a spherical particle is small compared to the length of an acoustic wave and the amplitude of spheroidal oscillations is small compared to the radius of the particle. In this case the expression for angular natural frequency of spheroidal oscillations is[13]:

$$\Omega_2^2 = \frac{24\sigma}{(2\rho_e + 3\rho_i)R^3} \qquad (3)$$

where $W_2 = 2p.f_2$; s - surface tension of the drop substance on the border with the surrounding medium; r_e, r_i - density of the medium substance and the drop, respectively. With $r_i >> r_e$ expression (3) turns into a wel-known Rayleigh formula [8], describing a spectrum of characteristic frequencies of spherical particle oscillations in the air. Expression (3) shows that the resonance frequency grows with the decrease of the bubble radius. Calculations show that spheroidal oscillations have the highest Q-factor [13], therefore the mode with $l = 2$ *was given preference in the experiments. High Q-factor of spheroidal oscillations makes it possible to increase accuracy of R* measurements using expression (1) and to widen diagnostic possibilities of the methods.

Fig.1 shows a diagram of the experimental unit. Optical system 1 is intended for producing a parallel linear-polarization beam of laser radiation of sufficient cross section. Radiation source is a He-Ne laser with radiation wavelength l = 0.63 m and output P_o = 1 MW. Central part of the unit is a system for particle suspension and excitation of oscillation of particle shape. It includes acoustic resonator 4, particle suspension system 8 and system 9 for oscillation excitation. The acoustic resonator is made of transparent optical glass and is in the form of a cylinder. The resonator is filled up to a calculated height with fresh double-distilled water, put through a system of microparticle filtering. The resonator is closed from above with a cap, which makes an acoustic reflector. An opening in the cap is for the input of examined particles.

Suspension system is to excite in the resonator an acoustic field with a configuration and power sufficient to keep an examined particle in a given point in the longitudinal axis of the resonator in a stable way. The suspension system includes a master oscillator of low-frequency signals GZ-51/1, a power amplifier, a matching reactive two-port and a ring piezoelectric radiator. The radiator efficiency depends upon its parameters and a proper selection of the matching circuit with the master oscillator in the required frequency band. So the efficiency is determined by a possibility to make minimum losses of power and signal distortion. In the search for optimal solution, 20 piezoelectric elements of different shape, made of ceramics CTS-19, CTSNV-1, CTS-22, CTS-23, CTS-24 and CTBS-3 were studied. Langeven's radiator was designed, manufactured and successfully used. It consists of two thin piezoelectric discs CTS-22 fixed in between two metal plates made of an aluminium alloy.

The principle of operation of the suspension system is the following. A change of frequency of the master oscillator excites one of the working modes of the piezoelectric radiator and as a result a definite type of acoustic oscillations with a specified field distribution in the whole space is set up in the resonator. For a glass resonator with an inside diameter $d = 27$ *mm and a water column* $h = 190$ *mm at sound velocity*

$v = 1500$ *m/s the working frequency of mode excitation* $(r, j, z) = (1, 0, 16)$ equals

$f_{1,0,16} = 92.63$ kHz. Length of the standing wave along *z coordinate (resonator axis) is in this case* l = 23.8 mm. Experimental study of distribution of the field of the standing wave of mode (1, 0, 16) in radial direction and in the longitudinal axis proved the accuracy of theoretical calculations.

Thus, the resonator produces an axis-symmetrical mode with a given distribution of the field in longitudinal and cross sections, securing a studied drop in a definite point along the axis of the resonator.

The system of excitation of particle shape oscillations is designed to form an acoustic field with a configuration, frequency and amplitude enabling to obtain forced resonance oscillations of the shape of a suspended drop at a frequency of natural oscillations of one of the modes with l = 1, 2, 3,.... The excitation system includes; high-frequency ultrasonic generator UZT-1.01F, matching reactive two-port, low-frequency signal generator GZ-18 and a piezoceramic radiator. Modulating voltage frequency f_2 is set up based on the calculations from expression (3) for a drop of a known radius, density and coefficient of surface tension.

After reaching a stable levitation of the studied drop, amplitude of the modulating signal must be raised to a value, which provides optimum oscillation of the drop shape. Then maximum Q-factor of spheroidal oscillations of the drop must be reached by adjusting frequency f_2.

Optical TV system 10 for particle imaging is designed for real-time observation of oscillations of the drop shape and for estimation of sizes of studied particles. The system is based on a small-size TV unit MTU-1 which comprises a TV-camera and video-display. The optical section of the system includes an illuminant, reading microscope with adjustable magnification from 19x to 33x (measuring range 0.015 - 6 mm) and vidicon of the TV camera. Images of the drop on the photocathode of the vidicon of the camera are produced by a direct-shadow method. Extension rings for the camera lens enable to have images of particles down to 100 m. A stroboscope method is used to watch different phases of drop oscillation. Stroboscope SSh -2 (flash repetition frequency from ~ 10 to 150 Hz) allows to observe other dynamic processes: fluctuation of drop centre position, small drop coagulation, drop "boiling", development of gas bubbles inside a liquid drop, etc.

Photoelectric system 11 is designed for recording the light scattered by particle. Optical part consists of a lens, slit diaphragm, interference filter for l = 0.63 m (halfwidth of the transmission band D l = 50 A) and photocathode FEU-106 which is used as a converter of optical signals to electrical.

Modulation is applied to laser radiation amplitude using a mechanical chopper when the suspended particle sizes are controled without excitation of shape oscillation. If there is excitation of shape oscillation the signal is amplitude modulated with frequency f_2. In both cases the signal parameters are determined with an electronic millivoltmeter V3-43 and oscillograph C1-73. A variable signal after convertion into d.c. voltage may be registered by recorder LKSZ-003.

System of diagnostics 12 of acoustic field distribution in resonator 4 consists of a small-size probe with a piezoceramic microtransducer as a sensing element. To provide transducer movement along the axis of the resonator and on its radius the probe is fixed to a substage ST-12 which guarantees movement along both coordinates with an accuracy of 0.1 mm.

Benzene drops with ni = 1.120 were studied using the described experimental unit. Theoretical value $b^{(3)}$ for benzene is 91.50. Calculated value of the first-order rainbow angle is a rough value since it depends on the drop radius [11, 13].

Fig.2 shows possible control of sizes of suspended single drops of benzene from relation (1). Fig.2 shows possible control of sizes of suspended single drops of benzene from relation (1). Experimental values are marked by dots. The straight line is drawn based on calculations from (1). An experimental plot of normalized light intensity as function of the square of the radius of benzene drops was constructed for *R from* ~ 80 m (estimated value) to 1200 m. Normalization was done to the intensity of light scattered by a drop with *R = 630* m, corresponding to the middle of the size range of studied drops. The recording system was calibrated with the optical TV system. Good agreement of experimental and theoretical relations indicates that measurements of the intensity of light scattered by a suspended drop at an angle of the first-order rainbow allow to determine radius of single particles in the studied range with an error not more than 1-3 %.

Results of an experimental study of the resonance frequency of spheroidal oscillations of a benzene drop as function of its size are shown in Fig.3. Experimental values are marked by dots. The straight line is constructed based on calculations from expression (3). The intensity of light scattered by an oscillating benzene drop was measured at an angle of the first-order rainbow $b^{(3)}$. Resonant excitation of spheroidal oscillations of a suspended benzene drop was achieved by adjusting frequency $\Omega 2$ of a modulating signal. The onset of resonance was controlled in two ways: visually - by a complete wash-out of interference bands on the display; and by the readings of millivoltmeter and oscillograph i.e. by maximum agreement between an alternating and direct components of the signal. In this case a condition of forced spheroidal oscillations of a benzene drop with maximum Q2-factor close to a theoretical Q-factor was realised. Fig.4 shows an experimental resonance curve for spheroidal oscillations of a benzene drop with R = 1752 m. The following calculated data were obtained for this drop: f_2 @ 29 Hz, Q_2 @ 15. An experimental value of Q-factor of spheroidal oscillations of a drop determined from the width of a resonance curve of level 0.7 is $Q_2 = 13$.

Thus, the experiments show that measurements of the intensity of the light scattered by a suspended drop at an angle of the first-order rainbow allow for a reliable control of particle sizes in situ.

The authors express their gratitude to professor Rinkevichus B.S. for his critical and useful remarks contributing to completion of the present work.

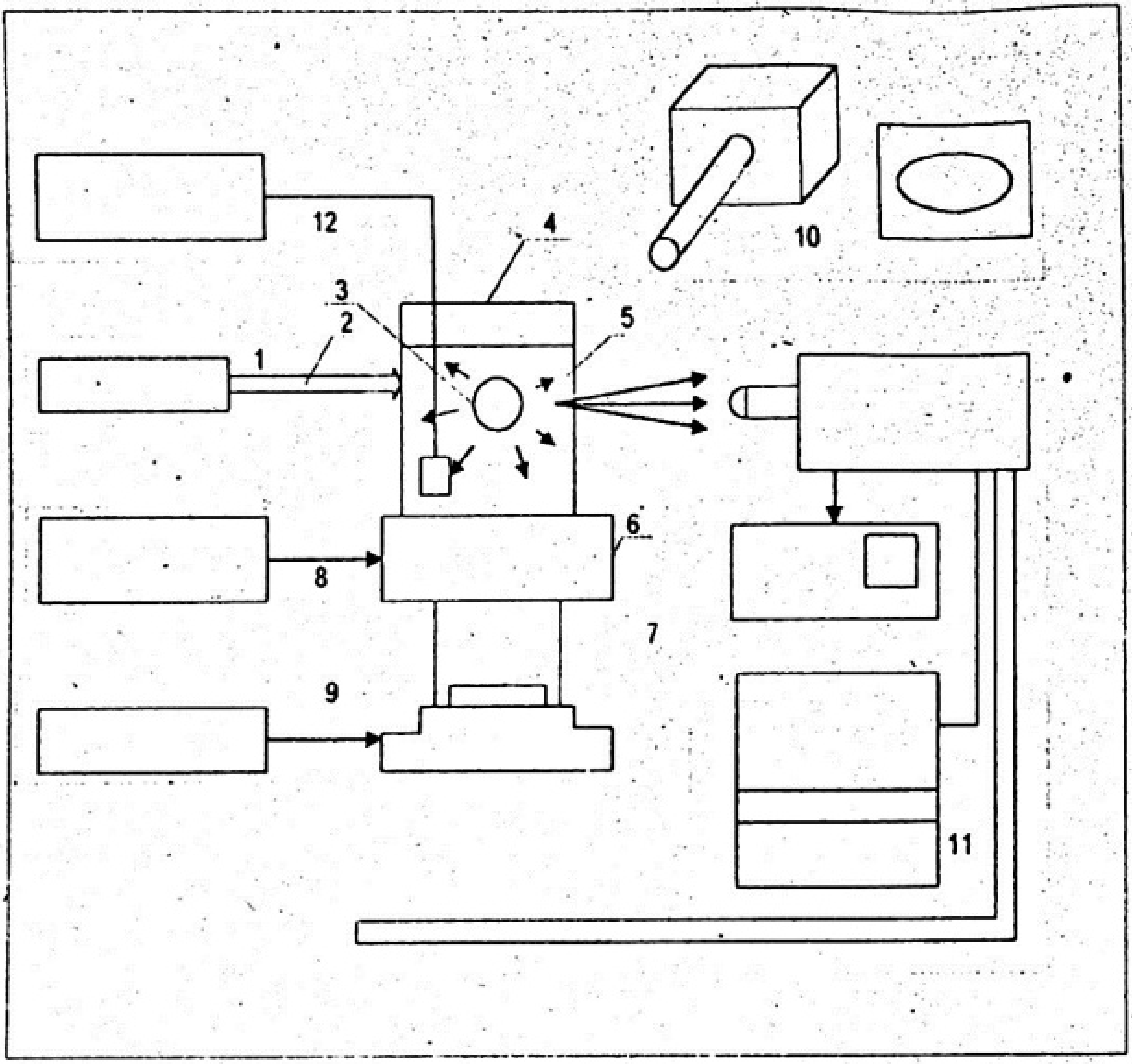

Fig.1 Block-diagram of experimental unit: 1 - optical system of light beam formation for particle sounding; 2 - sounding light beam; 3 - studied particle; 4 - acoustic resonator; 5 - liquid to fill up resonator; 6 - piezoelectric radiator of levitator; 7 - piezoelectric radiator exciting oscillations of particle shape; 8 - particle levitation system; 9 - system of excitation of oscillations of particle shape; 10 - optical TV system of particle imaging; 11 - photoelectrical system for recording particle scattered light; 12 - system of diagnostics of acoustic field distribution in a levitator's resonator

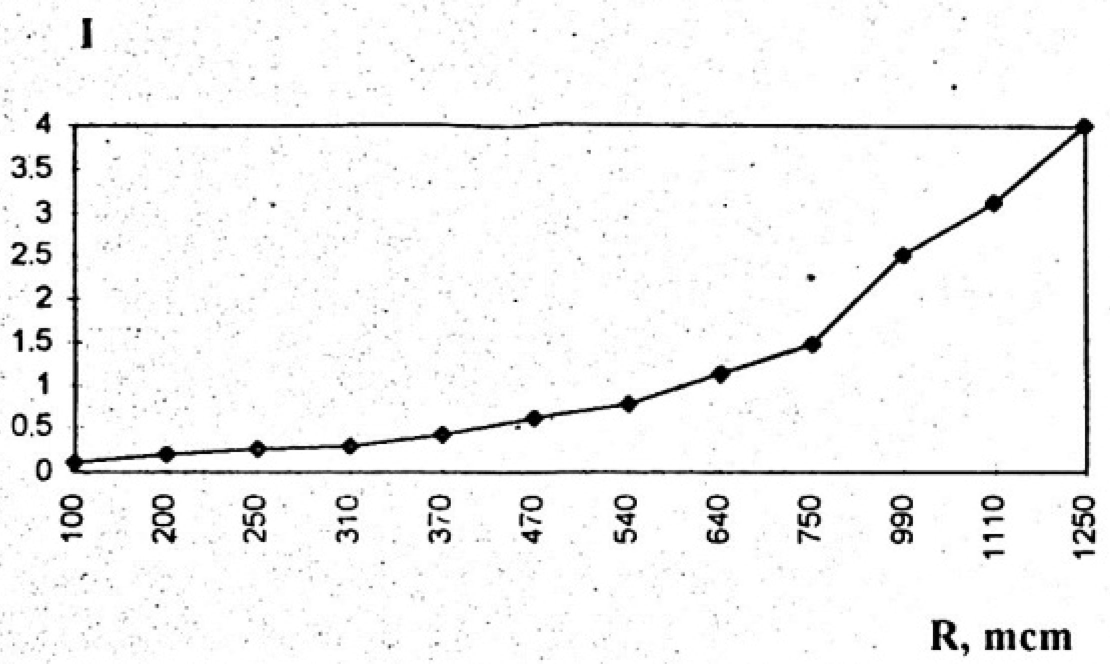

Fig.2. Normalized light intensity as function of the square of drop radius

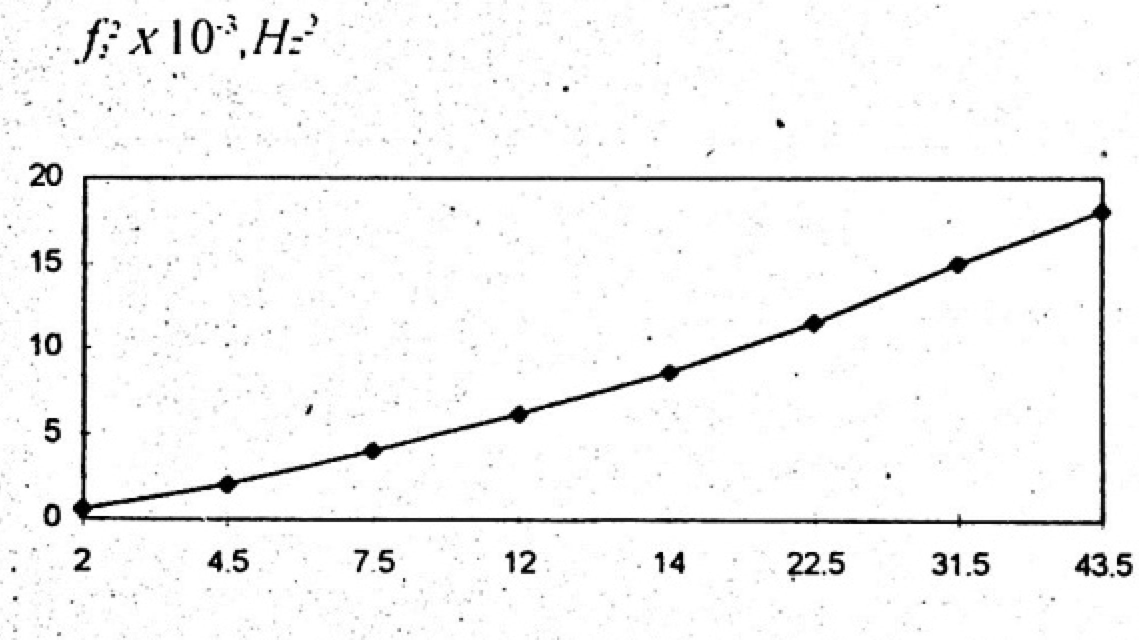

Fig.3. The square of proper frequency of spheroidal oscillations as function of R_0^{-3}

24	25	26	27	27.5	28	28.3	29	29.7	30	30.5	31	31.2	31.8
0.05	0.06	0.08	0.18	0.4	0.61	0.93	1.1	0.98	0.88	0.66	0.3	0.25	0.16

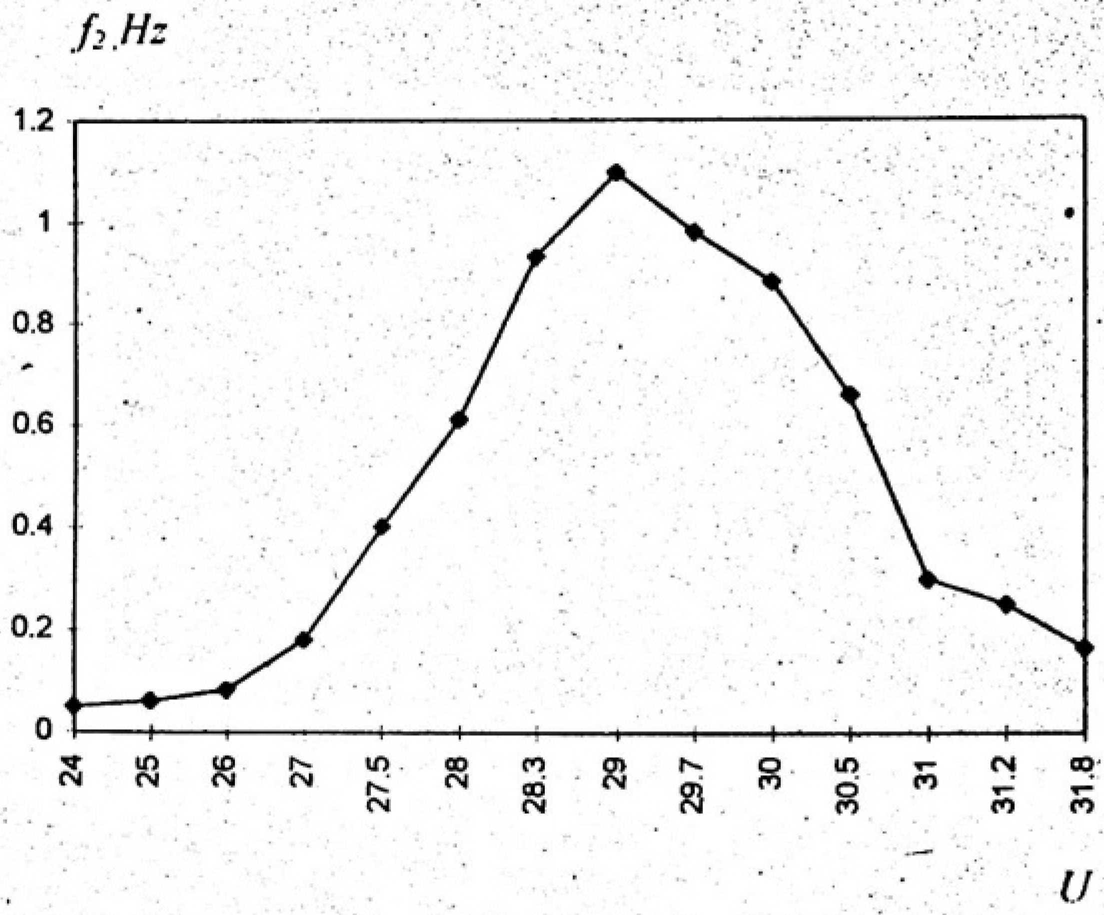

Fig.4. Experimental resonant curve of spheroidal oscillations of a benzene drop of radius

R = 1752 m suspended in water