Observation of Acousto-Optic Diffraction of Terahertz Radiation in Liquefied Sulfur Hexafluoride at Room Temperature

Pavel A. Nikitin, Boris A. Knyazev, Vitaly B. Voloshinov, and Michael A. Scheglov

Abstract—Diffraction of terahertz radiation on ultrasound in liquefied sulfur hexafluoride at room temperature was observed for the first time. The dependences of the diffraction efficiency on the sound frequency, angle of incidence, and amplitude of radio-frequency electrical signal applied to a piezo-transducer were studied. Deflection of diffracted radiation by an angle of 10° was observed. It was found that the diffraction efficiency at room temperature is significantly (4 times) higher than the one at 14°C known from the literature. The obtained and published data on the acousto-optic diffraction in liquefied sulfur hexafluoride contradict the well-known theory.

Index Terms—acousto-optics, diffraction, terahertz radiation, liquefied gas.

I. Introduction

The phenomenon of acousto-optic (AO) interaction is based on the effect of light diffraction by acoustic waves. This effect is widely used in the ultraviolet, visible, and middle infrared spectral ranges for electronical control of the radiation beams. However, AO methods are still not practiced in the terahertz (THz) range that is intensively mastered at present [1]. Acousto-optics can provide new opportunities to control parameters of THz radiation, as compared to the state of the art devices [2], [3]. Of special interest there are studies of AO diffraction of powerful THz radiation generated by such sources as gas and free-electron lasers (FEL) that are employed in wireless communication and matter analysis [4]. Only a few number of papers devoted to the problem of AO interaction in the THz range applying methanol lasers and FELs are known [5], [6]. In this paper, we investigated AO diffraction of the FEL high-power monochromatic radiation.

The ratio of intensities of the transmitted I_0 and diffracted I_1 radiation is defined as diffraction efficiency $\xi = I_1/I_0$. In the terahertz region, the diffraction efficiency is usually of the order $\xi = 0.01\%$ when single crystals are used as the medium of AO interaction. As known [6], the diffraction efficiency is equal to:

$$\xi = \frac{I_1}{I_0} = \frac{\pi^2}{2\lambda^2}\frac{M_2 P_\mathrm{a}}{d}L, \tag{1}$$

where λ is the radiation wavelength, $P_\mathrm{a} \propto U^2$ is the acoustic power, proportional to the square of the voltage U, applied to a piezoelectric transducer with width d and length L, and $M_2 \propto 1/V^3$ is the AO figure of merit. The piezoelectric transducer width d is usually chosen equal to the aperture D of a light beam ($D = d$).

The AO figure of merit $M_2 \approx 500 \cdot 10^{15}$ s^3/kg in liquids is a few times higher than that for crystals, because of lower sound velocity [7]. Therefore, liquids are more suitable for AO applications in the THz region. However, at ambient conditions (room temperature and 1 bar atmospheric pressure), the diffraction efficiency in liquids remains extremely small [7]. To overcome this drawback the author of paper [5] suggested using liquefied noble gases and sulfur hexafluoride (SF_6) as the medium of AO interaction, by virtue of their high transmittance in the THz range and low sound velocity. In this work it was found that among the examined media, sulfur hexafluoride was characterised by the highest value of M_2 of about $10^4 \cdot M_2^{\mathrm{SiO_2}}$, where $M_2^{\mathrm{SiO_2}} = 1.51 \cdot 10^{-15}$ s^3/kg is the AO figure of merit in fused silica. Note that the AO figure of merit in SF_6 is about 100 times higher than that in liquids. It was demonstrated that in the liquefied gas (SF_6), it was possible to achieve the diffraction efficiency of THz radiation close to 100%, but this required a significant voltage amplitude of the order of $U \approx 1$ kV [5].

To our knowledge, there are no works in which the AO interaction in a liquefied gas was studied. There is one exception related to the paper [5]. In this paper, the experiment was carried out only at two temperatures: $t = 13$°C and $t = 14$°C. The experimental results of this work disagree with the theory developed by the authors. The theory predicts a higher AO figure of merit M_2 (and hence a greater diffraction efficiency $\xi \propto M_2$) at higher temperature t of sulfur hexafluoride. However, decreasing the temperature by 1°C from 14°C to 13°C led in [5] to 3 times higher diffraction efficiency ξ at the same voltage U. Therefore, the aim of our research is to verify the results of

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the paper [5] and realize AO diffraction of THz radiation at room temperature to determine optimal conditions.

II. Theory of AO interaction in liquefied gases

The AO figure of merit M_2, as well as the refractive index n, of liquids may be calculated using the Lorentz-Lorenz equation [5], [8]:

$$n = \sqrt{1 + \frac{2A\rho}{1 - 2A\rho/3}}, \qquad A = 2\pi a \frac{N_\mathrm{A}}{M_\mathrm{m}}, \tag{2}$$

$$M_2 = \left[\frac{(n^2-1)(n^2+2)}{6n}\right]^2 \frac{4}{\rho V^3}, \tag{3}$$

where a is the mean polarizability of a molecule, N_A the Avogadro number, ρ the density, n the refractive index, and M_m the molecular weight. A is a factor that can be assumed to be independent of the pressure p and density ρ; its value can be derived from equation (2) at a certain pressure p_0 and a temperature t_0:

$$A \approx \frac{3}{2\rho} \frac{n^2-1}{n^2+2}\bigg|_{(t_0,p_0)}. \tag{4}$$

The dependence of the sound velocity V in liquefied SF_6 on the temperature and pressure was measured in paper [5]. Data of the similar dependence of the density ρ of liquefied SF_6 is given in [9]. The values of p_0 and t_0 were set equal to $p_0 = 26$ bar and $t_0 = 20°\mathrm{C}$, since the refractive index value $n = 1.241 \pm 0.002$ was measured in [5] only at these conditions. The value of A factor in (4), as well as the results of the linear approximation of $V(t)$ and ρ in the temperature range $t = 10 - 30°\mathrm{C}$ at a constant pressure p_0 by the least-square method, are as follows:

$$A = 1.64 \cdot 10^{-4}\ \mathrm{m^3/kg}, \tag{5}$$

$$V = (344 \pm 10) - (6.9 \pm 0.5)t, \tag{6}$$

$$\rho = (1600 \pm 50) - (11 \pm 3)t, \tag{7}$$

where V is in (m/s), ρ is in ($\mathrm{kg/m^3}$), and t is in (°C).

The dependence of the AO figure of merit M_2 on the temperature predicted in [5] using approximate relations for the refractive index and photoelastic constant and the same dependence calculated using directly the equation (3) and data from [5], [9] are almost the same with a difference of about 10% at the edges of the temperature range $t = 10 - 30°\mathrm{C}$. This dependence shows non-linear growth of M_2 with temperature, and one can see from equation (1) that the higher the temperature, the stronger the diffraction efficiency.

However, due to the large length of the AO interaction of about $L \approx (10 - 20)$ cm, the spatial deviation of the diffracted beam from the transmitted beam of THz radiation becomes significant (about a few centimeters). In addition, it is necessary to shift the AO cell so that the incident THz beam travels at some distance l from the piezoelectric transducer. This requires consideration of acoustic attenuation $P_\mathrm{a} = P_\mathrm{u} U^2 \exp(-\alpha_\mathrm{s} l)$:

$$\xi = \frac{\pi^2}{2\lambda^2} \frac{M_2 P_\mathrm{u} U^2}{d} L \exp(-\alpha_\mathrm{s} l), \tag{8}$$

where α_s is the sound attenuation coefficient, which is temperature dependent, and P_u is the coefficient of proportionality of the voltage U applied to the piezoelectric transducer and the acoustic power P_a.

The data on the temperature dependence of sound attenuation at different sound frequencies F are reported in [5]. Since α_s arises quadratically with F and is assumed to be independent of the pressure p, the data for $\alpha_\mathrm{s}(t)$ was taken for $F_0 = 300$ kHz and was approximated by a smooth function:

$$\alpha_\mathrm{s} = \left(\frac{F}{F_0}\right)^2 \cdot [(2466 \pm 22) + (4.81 \pm 0.06)t^{1.875}] \cdot 10^{-4}, \tag{9}$$

where the value 1.875 corresponds to the smallest error in the coefficients, F is in (kHz), α_s is in ($\mathrm{cm^{-1}}$), and t is in (°C).

Using formula (8), a series of temperature dependences of the diffraction efficiency ξ at $l = 5$ cm and $p_0 = 26$ bar for various frequencies F of ultrasound was calculated. The dependencies are shown in Fig. 1. For convenience of the analysis, ξ was normalized to its value ξ_0 at $t_0 = 20°\mathrm{C}$, $p_0 = 26$ bar and $F_0 = 300$ kHz:

$$\frac{\xi}{\xi_0} = \frac{M_2(t)}{M_2(t_0)} \exp\{-[\alpha_s(t,F) - \alpha_s(t_0,F_0)]l\}, \tag{10}$$

where ξ/ξ_0 is independent on the radiation wavelength λ, the sizes of the piezoelectric transducer L and d, and the driving voltage U.

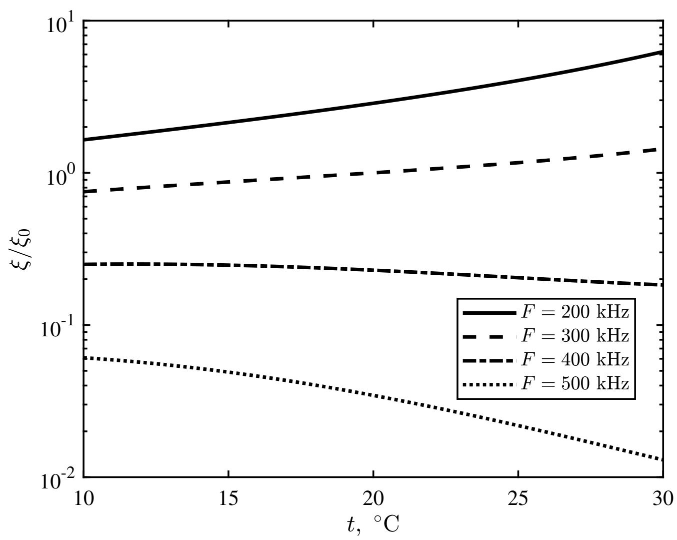


Figure 1: Dependences of the normalized diffraction efficiency in liquefied sulfur hexafluoride under constant pressure 26 bar on the temperature at different sound frequencies.

It follows from Fig. 1, that at higher sound frequencies F, the diffraction efficiency ξ decreases with temperature t. However, at low frequencies F, the efficiency is growing. This trend may be explained in the following way. The AO figure of merit M_2 non-linearly increases with t and it does not depend on F. It should be mentioned that the attenuation of sound α_s must also be considered here. The attenuation α_{s} increases at higher t and F. However, at $F = 200$ kHz and $F = 300$ kHz, the sound attenuation remains small, therefore the influence of AO figure of merit is dominating. As a result, at these moderate sound frequencies, the diffraction efficiency increases with temperature. On the other hand, at $F = 400$ kHz and $F = 500$ kHz, the sound attenuation has a stronger impact on the diffraction efficiency than the AO figure of merit. That is why, the diffraction efficiency at high sound frequencies and hight temperature decreases.

Nonetheless, the experimental results obtained in the paper [5] disagree with the developed theory; namely, the diffraction efficiency at $t = 13°$C turned out to be about 3 times greater than that at $t = 14°$C. This indicates the imperfection of the theory.

III. Experimental results and discussion

A. Experimental setup

AO devices, on the one hand, are characterised by high spectral resolution [10]. On the other hand, the diffraction efficiency of AO interaction is weak in the THz range due to relatively high wavelength ($\xi \propto 1/\lambda^2$.,Therefore the experiments were performed using the high-power monochromatic radiation of the Novosibirsk FEL [11]. The radiation wavelength was chosen equal to $\lambda = 130$ μm, corresponding to the minimal absorption of the atmosphere in the transparency window 128–132 μm [12]. The scheme of the experimental setup is shown in Fig. 2 and the cross-section of the AO cell is shown in Fig. 3. All dimensions are in millimeters.

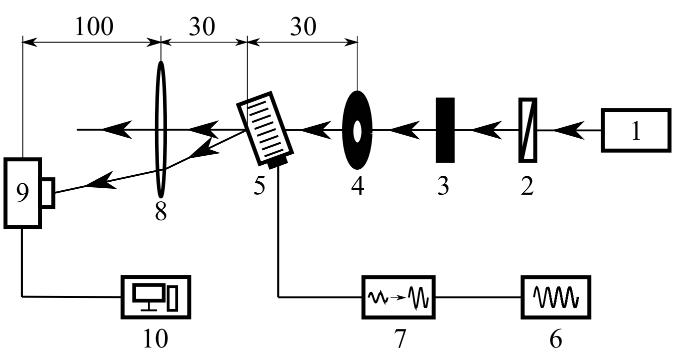


Figure 2: Schematic drawing of the experimental setup: 1 – free-electron laser; 2 – polarizer; 3 – attenuator; 4 – diaphragm; 5 – AO cell; 6 – radio-frequency generator; 7 – radio-frequency amplifier; 8 – lens; 9 – microbolometer array; 10 – computer.

The polarization of the THz radiation from FEL 1 was set by the polarizer 2, while its intensity was controlled by the set of calibrated attenuators 3. The THz beam aperture $D = 6$ mm was determined by the diaphragm 4. The AO cell 5 had nearly the same cross-section as in paper [5]. Our cell was also built of heavy duty steel. It was fabricated in form of a hollow round tube with the length of 10 cm, diameter of 15 cm, and two flanges at the ends. The flanges were 3 cm thick. Our

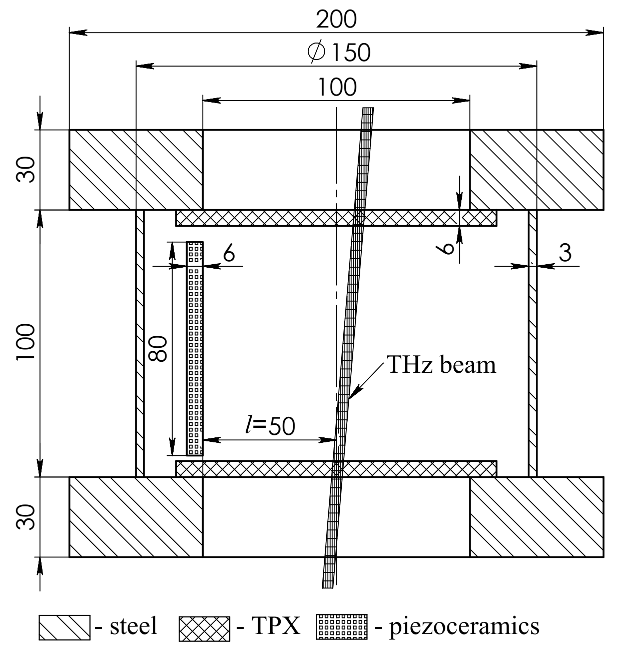
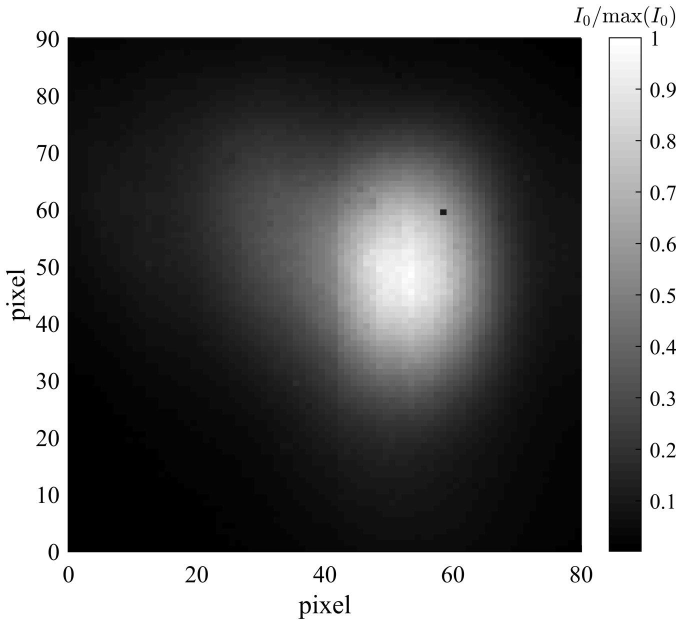


Figure 3: Cross-section of the acousto-optic cell for a liquefied gas.

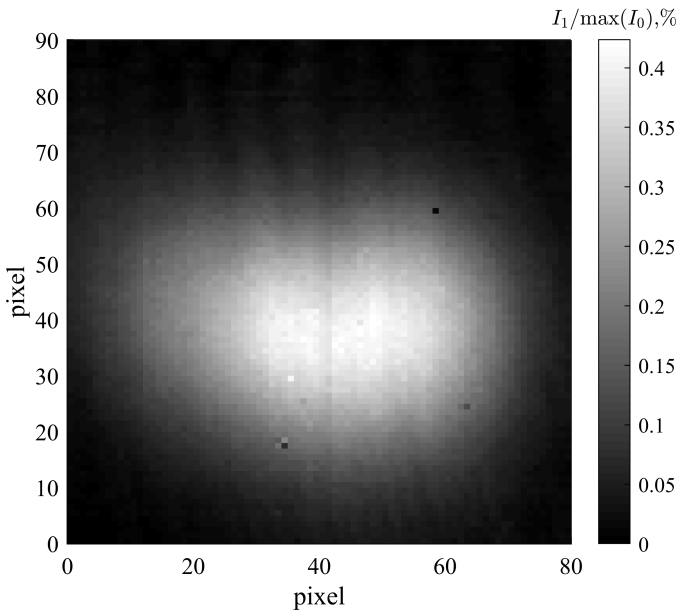
design is simpler and enables one to apply pressure up to 100 bar. The optical windows with the thickness of 6 mm were made of TPX, known as a transparent and strong material [13]. The openings in the flanges were $100{\times}10$ mm. An electrical signal from the generator 6 and the radio-frequency amplifier 7 was applied directly to the electrodes of the ceramic piezoelectric transducer having the dimensions $80 \times 8 \times 6$ mm ($L = 80$ mm). The electrodes were mounted on the 80×8 mm faces. The transducer was aligned parallel to the AO cell axis of symmetry, whereas the diffracted radiation was deflected in the horizontal plane. Because of the strong divergence of the THz radiation, the diffracted and transmitted beams were focused by the kinoform lens 8 having the focal length 8 cm [14]. The distributions of intensity over the cross-section of the beams were detected by the uncooled microbolometer array 9 with resolution 320×240. The array was placed at a distance of 10 cm from the lens to eliminate interference artefacts [15]. Data from the microbolometer array was processed by personal computer 10.

B. Diffraction efficiency

Figure 4 shows the spatial profiles of the diffracted and transmitted radiation intensities that were obtained for the first time. The oval shape of the diffracted beam was associated with its inclined incidence on the microbolometer array. The angle of incidence approximately equaled to the doubled Bragg angle $2\theta_{\text{B}} = \lambda F/V \approx 10°$. Note, that the microbolometer array could detect only the intensity distribution. Therefore distribution of the phase in the beams was not measured. In accordance with the theoretical results given in [16], we expected uniform profile of the phase across the beams cross-sections. This approach is valid in the case of a plane acoustic wavefront and also in absence of the THz beams spreading.



(a)



(b)

Figure 4: Normalized intensity obtained from the microbolometer array for: a) transmitted beam; b) diffracted beam.

These results were obtained under the following conditions inside the AO cell: temperature $t = (24 \pm 1)°\mathrm{C}$, pressure $p = 24$ bar. The electrical signal had the amplitude $U = 75$ V and the frequency $F = 328$ kHz, corresponding to the fundamental frequency of the piezoelectric transducer. All measurements were made using a continuous mode of ultrasound. The AO cell was adjusted in such a way that the THz beam was incident at the Bragg angle. For correct comparison of the obtained results, the intensities were normalized relatively to the maximum value of I_0. The images from the microbolometer array were cropped to the same size of 80×90 pixels. The sound wave velocity $V = (240 \pm 10)$ m/s in liquefied sulfur hexafluoride was determined based on known distance (12.5 mm) between the centers of the diffracted and transmitted THz beams in the plane of the microbolometer array. Using the calculated value of the velocity, we estimated the time response of the intensity modulation as $\tau = D/V = 25$ μs [17].

The value of the diffraction efficiency $\xi = (0.80 \pm 0.09)\%$ in liquefied SF_6 was about 100 times higher than that in the best non-polar liquid hexane (C_6H_{14}) [7]. This was mainly due to the extremely high AO figure of merit $M_2 \approx 10^4 \cdot M_2^{\mathrm{SiO_2}}$ of SF_6. To compare our results with the data in the paper [5], it is necessary to take into account the temperature dependence of the AO figure of merit M_2, as well as of the sound attenuation coefficient α_s given in paper [5]. The experimental conditions were as follows:

- in our work: $t = 24°\mathrm{C}$, $p = 24$ bar, $\lambda = 130$ μm, $M_2 = 2.05 \cdot 10^4 \cdot M_2^{\mathrm{SiO_2}}$, $\alpha_\mathrm{s} = 0.44$ cm^{-1}, $l = 5$ cm, $L = 80$ mm, $d = 8$ mm, $U = 75$ V;
- in paper [5]: $t = 14°\mathrm{C}$, $p = 28$ bar, $\lambda = 119$ μm, $M_2 = 0.85 \cdot 10^4 \cdot M_2^{\mathrm{SiO_2}}$, $\alpha_\mathrm{s} = 0.32$ cm^{-1}, $l = 2$ cm, $L = 180$ mm, $d = 10$ mm, $U = 150$ V, $\xi = 2.7\%$;

The proportionality coefficient $P_\mathrm{u} = 28 \cdot 10^{-6}$ W/V^2 in equation (8) for the diffraction efficiency ξ and voltage U applied to the piezoelectric transducer was calculated by substitution of the results of the paper [5] in relation (8). The value of ξ expected under the conditions of our experiment was $\xi = 0.16\%$. However, from Fig. 5 one can see that the diffraction efficiency in liquefied SF_6 at the temperature $t = 24°\mathrm{C}$ was 4 times greater than the expected one. There were two reasons for this difference. On the one hand, the piezoelectric transducer used in our experiments was made of a single piece of PZT piezo-ceramics, whereas in the paper [5], it was built of small length sections cut out of PZT. It is obvious that the acoustic field depends on the piezoelectric transducer. However, there are no data on the dependence of the AO diffraction efficiency on the number and size of PZT elements. On the other hand, according to the results in Fig. 1, the diffraction efficiency will increase with temperature at the sound frequency $F = 300$ kHz. Therefore, if the proportionality coefficient P_u can be assumed to be independent of the type of piezoelectric transducer, the AO figure of merit for SF_6 has a stronger dependence on the temperature than that predicted in [5].

We believe that in order to describe the acousto-optic interaction in a liquefied gas correctly, it is necessary to use a more complex model for the refractive index. As such a model, one can use, for example, equation (2) from [18], in which the mean polarizability of the molecule is presented as a power series in density, whereas in [5] the polarizability of the molecule assumed to be density independent.

To estimate the voltage U_{opt} corresponding to the maximal diffraction efficiency $\xi = 100\%$, the well-known model of strong AO interaction [5] complemented by the consideration of the sound attenuation can be used:

$$\xi = \sin^2\left(\frac{\pi}{2}\sqrt{\frac{2}{\lambda^2}\frac{M_2 P_{\mathrm{u}} U^2}{d} L \exp(-\alpha_{\mathrm{s}} l)}\right). \quad (11)$$

Setting in (11) $\xi = 1$, we get:

$$U_{\mathrm{opt}} = \lambda\sqrt{\frac{d}{2M_2 P_{\mathrm{u}} L \exp(-\alpha_{\mathrm{s}} l)}}. \quad (12)$$

As the diffraction efficiency in our experiment was $\xi = 0.8\%$, the product of the normalized AO figure of merit and the coefficient P_{u} was $M_2 P_{\mathrm{u}}/M_2^{\mathrm{SiO_2}} = 2.9\ \mathrm{W/V^2}$. Therefore, in accordance with (12), the optimal voltage should be equal to $U_{\mathrm{opt}} = 1.3$ kV. Note, that the used model did not take into account the effect of cavitation leading to lower acoustic power at high driving voltage.

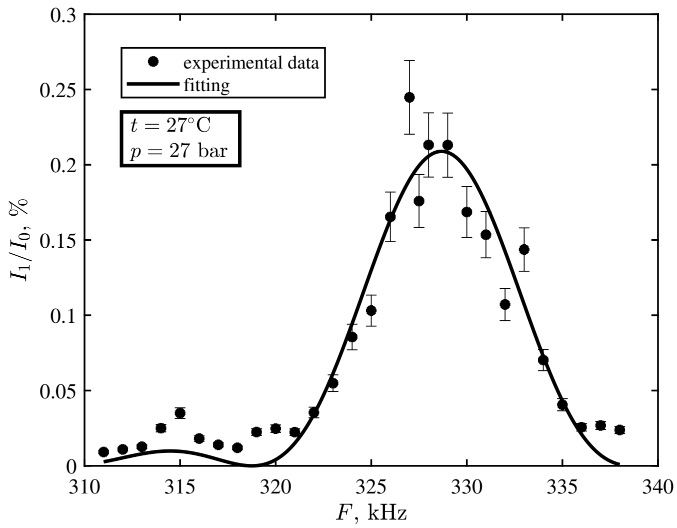


Figure 6: Diffraction efficiency as a function of the acoustic frequency.

C. Angular and frequency dependences of the diffraction efficiency

The dependence of the diffraction efficiency I_1/I_0 on the deviation $(\theta - \theta_{\mathrm{B}})$ of the angle of incidence onto the input window from the Bragg angle was examined at the same conditions as before. The results, approximated with the squared sinc function, are shown in Fig. 5. It follows that the AO interaction in liquefied SF_6 is sensitive to the angle of incidence θ: the angle bandwidth was about 1.2°. The value of I_1/I_0 was calculated as the ratio of the integral intensities of the transmitted and diffracted THz beams.

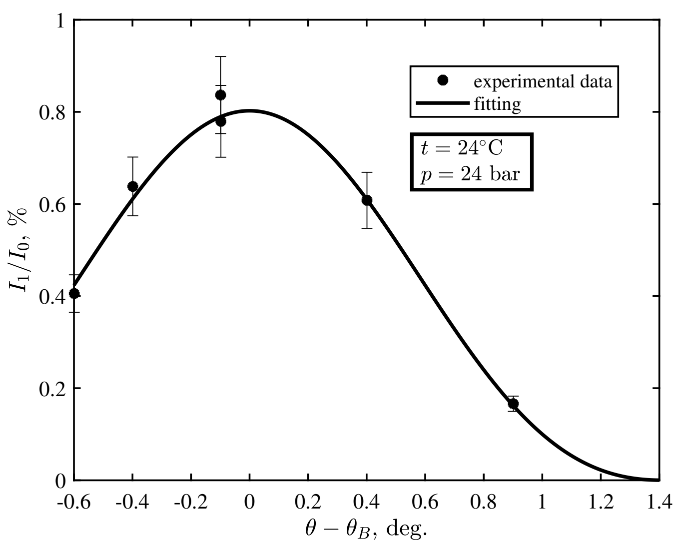


Figure 5: Diffraction efficiency as a function of the deviation of the angle of incidence from the Bragg angle.

In the subsequent experiments, the temperature and pressure of sulfur hexafluoride were higher due to the absorption of THz radiation and sound attenuation: $t = (27 \pm 1)°$C, $p = 27$ bar. The measured data of $\xi(F)$, as well as the curves calculated by the least squares method, are shown in Fig. 6.

From the dependences $I_1(\theta)$ and $I_1(F)$, one can derive the value of the effective length L_{eff} of the AO interaction region. Usually this length is slightly shorter than the length of the piezoelectric transducer $L_{\mathrm{eff}} \simeq L$ and can be calculated using the following relations [19], [20]:

$$\Delta\theta = \frac{0.9nV}{FL_{\mathrm{eff}}}, \qquad \Delta F = \frac{1.8nV^2}{\lambda F L_{\mathrm{eff}}}, \quad (13)$$

where $\Delta\theta$ and ΔF are the angular and frequency bandwidths at the conventional -3 dB criterion for the diffraction efficiency.

With consideration of the temperature dependence of the sound velocity (see equation (6)), the value of the effective length L_{eff} was estimated as $L_{\mathrm{eff}} = (2.8 \pm 0.4)$ cm at $t = 24°$C (determined from $I_1(\theta)$) and $L_{\mathrm{eff}} = (14 \pm 1)$ cm at $t = 27°$C (determined from $I_1(F)$). The two times difference between L_{eff} at $t = 27°$C and the real length of the PZT transducer $L = 8$ cm was explained by the fact that the frequency bandwidth ΔF was 2 times narrower than expected. This in turn was because of a direct electrical termination of the PZT transducer to the electrical generator without any electrical matching network. It leads to a resonant sound generation. Therefore, the frequency bandwidth of sound excitation was about 2 times narrower than the frequency bandwidth of the efficient AO interaction.

On the other hand, the effective length $L_{\mathrm{eff}} = 2.8$ cm at $t = 24°$C was about 3 times shorter than the length of the PZT transducer. It means that efficiency of the AO interaction is in a weaker dependence on the angle of incidence of THz radiation in the AO cell as compared to the model of plane wave interaction. We believe that it was related to the divergency of the THz beam $\delta\phi = \lambda/D = 1.2°$. Since the expected angular bandwidth of AO interaction of plane waves is about 0.4°, the angular bandwidth of the AO interaction was determined mainly by the diffraction divergency of the THz beam.

The used relation $\delta\phi = \lambda/D$ is based on the theory of diffraction of a plane electromagnetic wave on a circular aperture. This approximation is justified, since the radiation of the Novosibirsk free-electron laser is characterized by the divergence of 0.17° [21].

D. Influence of the acoustic power on the diffraction efficiency

The measured dependence of the diffraction efficiency on the amplitude of electrical signal applied to the piezoelectric transducer at $t = 27°$C is shown in Fig. 7. As one can see, at a higher temperature of the liquefied sulfur hexafluoride $t = 27°$C (in [5], $t = 14°$C), the dependence of the diffraction efficiency on the applied voltage U is not quadratic. Moreover, we registered something like a saturation effect at the voltages $U > 60$ V.

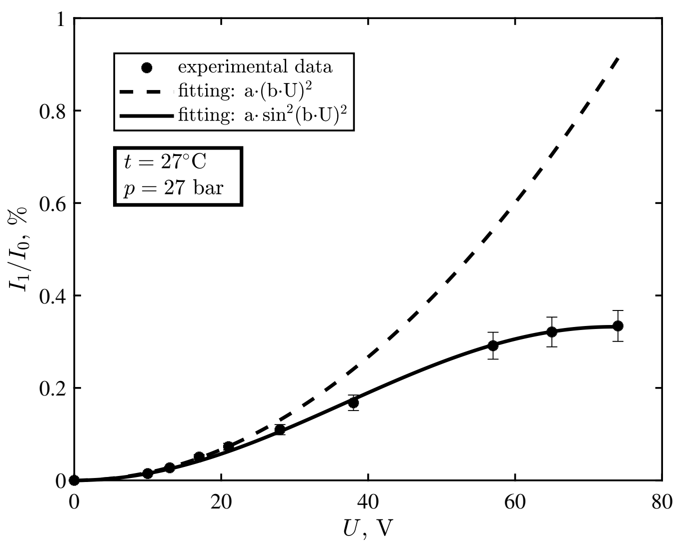


Figure 7: Diffraction efficiency as a function of the driving voltage.

We found that the $\sin^2(b \cdot U)$ model applicable at strong AO interaction approximated the experimental data best of all [22]. However, as follows from the data in the paper [5], the strong AO interaction is observed at the sufficiently higher voltage $U \approx 1$ kV. The saturation effect can be associated with non-ideal experimental conditions, i.e., far from the plane wave interaction (for example, complex structure of the acoustic field). The PZT transducer used in our experiment was of nearly the same width d as that used in paper [5]. Therefore it was considered that there was some other reason for the suturation effect. By direct observation we found that at $t = 27°$C the cavitation effect took place at $U > 20$ V: small bubbles flowed from the PZT transducer and the number of bubbles was increasing with the voltage amplitude. Moreover, the fitting of the first five experimental points with the quadratic function shown in Fig. 7 suggested absence of cavitation and proved that the diffraction efficiency value at $U = 75$ V could be the same as that at $t = 24°$C, i.e., $\xi = 0.8\%$. Thus, the effect of cavitation was the main reason for the reduction in the diffraction efficiency. This effect can be reduced, for example, by using chemically pure liquefied gas and its degassing.

IV. Conclusion

Our experiments showed that the AO interaction at room temperature (24–27)°C was stronger than that at the lower temperature 14°C known from literature. Disagreement of the experimental results for a number of temperatures with the well-known theory indicates its imperfection, which requires the use of a more complex model. The sound frequency bandwidth of the AO interaction was shortened by the resonant generation of sound, whereas the angular bandwidth broadened due to the divergence of the incident THz beam. It was found that the main factor of lower diffraction efficiency at the high level of applied voltages was associated with the cavitation process near the piezoelectric transducer. In order to increase the diffraction efficiency, we are going to carry out further experiments to damp the cavitation in the liquefied gas. We also plan to use the better design of the piezoelectric transducer.

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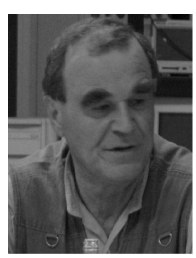

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