A review of non-polar liquids as materials for bulk acousto-optic devices operating with terahertz radiation

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Abstract. In this paper, the acoustic and optical properties of a number of liquids transparent in the terahertz range are systematized. The acousto-optic quality is calculated for the quasi-orthogonal and collinear geometry of the acousto-optic interaction. The parameters of acousto-optic deflectors and filters of terahertz radiation are determined. The diagrams linking the resolution for the filter and the maximum number of resolvable elements for the deflector with the diffraction efficiency are given.

1. Introduction

Acousto-optic (AO) devices are widely used to control electromagnetic radiation in the visible, infrared and ultraviolet ranges [1-3]. The most effective in terms of electrical power consumption are devices operating with shortwave radiation. This is due to the fact that the ratio of the intensity $I_1$ of diffracted radiation to the intensity $I_0$ of radiation incident on an AO cell is inversely proportional to the square of the radiation wavelength $\lambda$:

$$\frac{I_1}{I_0} \propto \frac{M^2 P_a}{\lambda^2},$$

where $P_a$ is the sound wave power, $M^2$ is the AO figure of merit, depending on the acoustic, optical and photo-elastic properties of the medium.

To control laser THz radiation, metallic and silicon diffraction elements are used. Thus, the AO effect can be used for real-time processing of information in the THz range in areas of science and technology that do not require large values of the intensity of diffracted radiation. The well-known AO crystals, such as paratellurite (TeO$_2$) and lithium niobate (LiNbO$_3$), are opaque in the THz range [4, 5] and the transparent crystals, such as germanium (Ge) and silicon (Si) have relatively small values of $M^2$. Therefore liquids become the most suitable media for AO interaction. To date, there are several review papers about the choice of the AO interaction medium in different spectral ranges of electromagnetic waves. Liquids were used as the AO interaction medium only in the visible range and an overview of their AO properties can be found in the works of Uchida, Watanabe and Hoff [6-8]. The purpose of this work is to select liquids that are most suitable for creating AO devices operating with THz radiation.
2. Acoustical, optical and photo-elastic properties of liquids

To obtain acceptable values of the diffraction efficiency, the length of the AO interaction region have to be of the order of 1 cm. Therefore, only relatively transparent liquids with a radiation absorption coefficient of no more than $\alpha < 5 \text{ cm}^{-1}$ were selected. It was assumed that the radiation intensity $I$ decreases exponentially with the distance $l$

$$I \propto \exp(-\alpha l).$$

As noted above, the suitability of the material from the point of view of acousto-optics is determined by its AO figure of merit $M_2$, which for liquids is determined by the following relation [6]

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

where $n$ – refractive index, $V$ – sound velocity, $\rho$ – density.

Table 1 shows the values of $n$ and $\alpha$ at $\lambda = 140 \mu\text{m}$ for a number of non-polar liquids, as well as for such polar liquids as alcohols. Since the values of the elasto-optical constant $\rho(\partial n/\partial \rho)$ in the THz range are unknown, table 1 was supplemented with the values of $M_2$ calculated by using the equation (2). Acoustic properties of the liquids are also shown in the table 1. Attenuation the sound was assumed exponential and its power $P_s$ is related to distance $l$ in the following way

$$P_s \propto \exp(-\alpha l).$$

| liquid | $\rho$ (g/cm$^3$) | $V$ (km/s) | $\alpha_s / (2F^2)$ ($10^{-17} \text{s/cm}$) | $n$ | $\alpha$ (cm$^{-1}$) | $M_2$ ($10^{15} \text{s}^3/\text{kg}$) |
|--------|-----------------|-----------|---------------------------------|-----|-----------------|--------------------------|
| $\text{C}_6\text{H}_{12}$ | 0.774 [9] | 1.255 [9] | 190 [10] | 1.421 [11] | 0.37 [11] | 604 |
| $\text{C}_6\text{H}_{14}$ | 0.655 [12] | 1.077 [12] | 60 [6,10] | 1.372 [13] | 0.69 [13] | 847 |
| $\text{C}_6\text{H}_{16}$ | 0.770 [12] | 1.338 [12] | 100 [12] | 1.428 [13] | 0.69 [13] | 520 |
| $\text{C}_6\text{H}_6$ | 0.881 [14] | 1.298 [14] | 850 [10] | 1.496 [11] | 5.5 [11] | 709 |
| $\text{C}_6\text{H}_{10}$ | 0.880 [10] | 1.360 [10] | 63 [10] | 1.492 [15] | 5.1 [15] | 605 |
| $\text{C}_6\text{H}_2\text{OH}$ | 0.789 [10] | 1.155 [10] | 54 [10] | 1.498 [16] | 130 [16] | 1135 |
| $\text{C}_6\text{H}_2\text{OH}$ | 0.814 [10] | 1.294 [10] | 97 [10] | 1.477 [16] | 45 [16] | 705 |
| $\text{CCl}_4$ | 1.584 [6] | 0.922 [6] | 550 [6] | 1.487 [11] | 1.4 [11] | 1053 |
| $\text{CHBr}_3$ | 2.877 [6] | 0.920 [6] | 290 [6] | 1.595 [17] | 6.0 [17] | 960 |
| $\text{CS}_2$ | 1.256 [6] | 1.141 [6] | 6100 [6] | 1.61 [18] | 3.5 [18] | 1228 |

The liquids given in table 1 have approximately the same refractive index $n \approx 1.4$ in the THz range. Among non-polar liquids, cyclohexane ($\text{C}_6\text{H}_{12}$) is the most transparent. Non-polar liquids, such as alcohols, absorb THz radiation much more strongly. This is due to the resonant stretching of hydrogen bonds. The acoustic properties of the listed liquids are very similar: the density is about $\rho \approx 0.8$ g/cm$^3$, and the speed of sound is $V \approx 1.2$ km/s. It should be noted that at relatively high sound frequencies $F$ value of the attenuation coefficient $\alpha_s$ of sound becomes comparable or even higher than radiation absorption coefficient $\alpha$. Therefore value of some combination of $\alpha_s$ and $\alpha$ will be a criterion for using a non-polar liquid as an AO interaction medium.

3. Quasi-orthogonal geometry of acousto-optic interaction

With the quasi-orthogonal geometry of the AO interaction, the diffracted radiation propagates approximately in the same direction as the transmitted. This configuration is shown in figure 1 and can
be used to create THz radiation modulators and deflectors. The angle of deviation of the diffracted radiation is equals to twice the Bragg angle $\theta_B \ll 1$ and depends linearly on the frequency of the sound $F$ [19]

$$\sin \theta_B = \frac{\lambda F}{2V}. \quad (5)$$

**Figure 1.** Scheme of the quasi-orthogonal diffraction of radiation on the phase grating, induced by sound wave.

Since the angle of deflection of the diffracted radiation is much less than unity, then during calculations we set Bragg angle equal to $\theta_B = 0.1$ (or approximately $5.7^\circ$). The AO interaction will be observed at the ultrasound frequency $F$

$$\sin \theta_B \approx \theta_B = 0.1; \quad F = 2 \sin \theta_B \frac{V}{\lambda} = 0.2 \frac{V}{\lambda}. \quad (6)$$

As established in the paper [20], in an absorbing medium at low diffraction efficiency the ratio of the intensity $I_1(L)$ of the diffracted radiation at the output of an AO cell to the intensity $I_0(0)$ of the radiation incident on an AO cell is determined by the following expression

$$\frac{I_1(L)}{I_0(0)} = \frac{\pi^2 M_s P}{2 \lambda^2} \frac{L}{d} \exp(-\alpha L), \quad (7)$$

where $L$ is the length of the AO interaction region, approximately equaled to the width of the sound beam, and $d$ is the width of the sound beam in the plane orthogonal to the AO interaction plane (see Figure 1).

From the expression (7) it follows that with increasing $L$, the diffraction efficiency linearly increases due to the interaction of radiation with a large number of periods of the sound wave on the one hand and decreases exponentially due to the absorption of radiation in the medium on the other hand. Thus, there is an optimal length $L_{\text{opt}}$ of AO interaction, at which the maximum diffraction efficiency is achieved [20]

$$L_{\text{opt}} = \frac{1}{\alpha}; \quad \frac{I_1(L_{\text{opt}})}{I_0(0)} = \frac{\pi^2 M_s P}{2 \lambda^2} \frac{1}{d \alpha e} \quad (8)$$

It was assumed that the diffraction efficiency at the wavelength of $\lambda \pm \Delta \lambda / 2$ is 2 times less than the diffraction efficiency at the wavelength $\lambda$. Under the quasi-orthogonal geometry of the AO
interaction, the following proportion between the bandwidth of the radiation wavelength and the bandwidth of the sound frequency is valid: $\lambda / \Delta \lambda = F / \Delta F$ [21].

As is known from the literature, the ultrasound frequency bandwidth $\Delta F$, in which an effective AO interaction is observed (at the edges of this region, the diffraction efficiency decreases by 2 times) is calculated as [22]

$$\Delta F = \frac{1.8nV^2}{\lambda FL}.$$  \hspace{1cm} (9)

The number $N = \Delta \theta / \delta \phi$ of resolvable elements is determined by the ratio of the angle bandwidth $\Delta \theta$, in which the effective scanning of the diffracted beam is realized, to the diffraction divergence $\delta \phi = \lambda / (nd_i)$ of the radiation in the AO interaction plane. Here $d_i$ is the width of the diffracted radiation beam in the AO interaction plane, which is assumed to be equal to the width $d_i$ of the beam incident on the AO cell (see figure 1).

The Bragg interaction is resonant in nature and requires the fulfillment of the condition of Bragg synchronism. When light is diffracted into $+1$ diffraction order, this condition has the form $k_1 = k_s + K + \Delta K / 2 + \eta$, where $\Delta K \equiv |\Delta K| \ll |K|$ and $|\eta| \ll |K|$. Here $K$ is the wave vector of sound corresponding to the Bragg matching condition; the value of $\Delta K$ corresponds to the bandwidth of sound frequency from the relation (9); $k_s$ and $k_1$ are the wave vectors of the incident and diffracted radiation and their moduli are the same $|k_s| = |k_1| = k$. In addition, the notation $\eta$ is used for the mismatch vector, which is directed orthogonally to the sound beam [23].

The relation between the wave vectors of the interacting waves can be shown on the wave vector diagram. Since the wave surface has the shape of a circle and the Bragg angle is small $\theta_B \ll 1$, then $|\eta| \ll \Delta K$. Therefore, the bandwidth of angles $\Delta \theta$ in the medium can be calculated as the ratio of the bandwidth of sound vectors $\Delta K$, in which the AO interaction is observed, to the module of the wave vector of light $k$. Thus, the number $N$ of resolvable elements is proportional to the width $d_i$ of the light beam and inversely proportional to the length $L$

$$N = \frac{\Delta \theta}{\delta \phi} = \frac{\Delta K / k}{\lambda / (nd_i) / (2\pi n / \lambda)} = \frac{1.8nVd_i}{\lambda FL} = \frac{0.9}{\sin \theta_B} \frac{d_i}{nL}. \hspace{1cm} (10)$$

4. Collinear geometry of acousto-optic interaction

A feature of the collinear AO interaction in liquids is that the diffracted radiation propagates towards the incident one. In the literature, the term “backward” collinear diffraction is used to describe this type of AO interaction. From the vector diagram of phase matching, it follows that collinear diffraction is observed when the wave vector of the sound is 2 times larger than the wave vector of light. The required ultrasound frequency is determined by the following relation [19]

$$F = \frac{2nV}{\lambda}. \hspace{1cm} (11)$$

Since the refractive index of liquids $n \approx 1.5$ is small, the maximal of angle of incidence of sound on the input optical window is limited by approximately $40^\circ$. Therefore, it becomes possible to construct an AO cell so that the path travelled by the sound to the input optical window is much less than the length of the AO interaction, and the attenuation of sound in this region can be neglected. In this case, the ratio of the intensity of diffracted radiation to the intensity of the incident radiation, as well as the optimal length of the AO interaction are calculated from the following equations [24]

$$L_{opt} = \infty, \hspace{1cm} \frac{L_{11}(0)}{I_0(0)} = \frac{\pi^2 M_s P_0}{2\lambda^2 S} \left( \alpha + \frac{\alpha_s}{2} \right)^2, \hspace{1cm} (12)$$
where $S$ is the cross-sectional area of the sound beam, equal to the area of the piezoelectric transducer.

The value of the two-sided band $\Delta k$ by the wave vectors of light is determined from the expression for $\Delta K$ taking into account the fact that $\Delta K = 2\Delta k$

$$\Delta K = 2\Delta k = 2\left(\alpha + \frac{\alpha_s}{2}\right). \quad (13)$$

Thus, the spectral resolution $R$ of the AO filter depends mainly on the absorption coefficient of the radiation $\alpha$ and the attenuation coefficient of the ultrasound $\alpha_s$

$$R = \frac{k}{\Delta k} = \frac{2\pi n \lambda}{\alpha + \alpha_s / 2}. \quad (14)$$

5. Results and discussion

It was assumed that $d_i = 10$ mm and $d = 5$ mm. When the width of the sound beam in the AO interaction plane is equal to the optimal length $L_{opt} = 1/\alpha$, and the ultrasonic power is $P_u = 1$ W, the values of the diffraction efficiency $I_i/I_o$ and the number $N$ of solvable elements of AO deflectors are calculated and given in table 2.

**Table 2. Properties of acousto-optic deflectors based on liquids.**

| liquid      | $L_{opt}$, cm | $F$, MHz | $I_i/I_o$, $10^4$ | $N$  |
|-------------|---------------|----------|-------------------|------|
| $C_6H_{12}$ | 2.7           | 2.5      | 3                 | 5    |
| $C_6H_{14}$ | 1.4           | 2.1      | 2.3               | 9    |
| $C_6H_{16}$ | 1.4           | 2.9      | 2.7               | 9    |
| $C_6H_{18}$ | 0.18          | 3.0      | 0.5               | 70   |
| $C_6H_{20}$ | 0.20          | 3.1      | 0.4               | 70   |
| $C_6H_{22}$ | 0.008         | 2.7      | 0.03              | 1800 |
| $C_6H_{24}$ | 0.022         | 2.9      | 0.06              | 600  |
| $C_6H_{26}$ | 0.17          | 2.3      | 0.6               | 90   |
| $C_6H_{28}$ | 0.29          | 2.8      | 1.3               | 50   |

When calculating the parameters of AO filters, it was assumed that the piezoelectric transducer area is $S = 5 \times 5$ mm$^2$, and the sound power is $P_s = 1$ W. The results of the calculations are given in table 3.

**Table 3. Properties of acousto-optic filters based on liquids.**

| liquid      | $F$ (MHz) | $\alpha_i$ (cm$^{-1}$) | $I_{i_i}/I_o$, $10^4$ | $R$ ($10^5$) |
|-------------|-----------|------------------------|-----------------------|--------------|
| $C_6H_{12}$ | 25.4      | 2.5                    | 2.4                   | 0.4          |
| $C_6H_{14}$ | 21.1      | 0.5                    | 9.3                   | 0.6          |
| $C_6H_{16}$ | 27.2      | 1.5                    | 1.5                   | 0.4          |
| $C_6H_{18}$ | 27.7      | 13                     | 0.05                  | 0.06         |
| $C_6H_{20}$ | 29.0      | 1.1                    | 0.19                  | 0.12         |
| $C_6H_{22}$ | 24.7      | 0.7                    | 0.001                 | 0.005        |
| $C_6H_{24}$ | 27.3      | 1.4                    | 0.003                 | 0.014        |
| $C_6H_{26}$ | 19.6      | 4.2                    | 0.9                   | 0.19         |
| $C_6H_{28}$ | 20.9      | 2.2                    | 0.19                  | 0.10         |
| $C_6H_{30}$ | 26.2      | 47                     | 0.017                 | 0.027        |
Based on the obtained results, a diagram was constructed for the AO deflectors (see figure 2), which interlinks the number $N$ of resolvable elements and the diffraction efficiency $I_t / I_0$ for the liquids considered at the radiation wavelength $\lambda = 140\mu m$.

![Diagram](image)

**Figure 2.** Dependence of the number of resolvable spots on the diffraction efficiency.

From figure 2, it follows that AO deflectors with a higher diffraction efficiency will be characterized by a smaller number of resolvable elements. That could be explained by the fact that the diffraction efficiency is proportional to the length of the AO interaction $L$, while the number of solvable elements is inversely proportional to $L$. Therefore, the isolines (corresponding to a certain liquid) in this diagram will be parallel to the diagonals of the squares, since the value of the product $N \cdot (I_t / I_0)$ remains constant as $L$ changes.

A similar diagram (see figure 3) was also constructed for AO filters. It interlinks the resolution $R$ of AO filters and the diffraction efficiency $I_t / I_0$ for the liquids considered. From the diagram a trend is visible: the higher resolution $R$ of the devices in which liquid is used as the interaction AO medium, the larger maximum achievable diffraction efficiency $I_t / I_0$. Moreover, as $I_t / I_0$ increases by 2 orders of magnitude, $R$ increases by an order of magnitude. It can be explained in the following way. As follows from the relations (12) and (14), the diffraction efficiency is inversely proportional to the square of the combination $(\alpha + \alpha_s / 2)$ of the sound attenuation coefficient and the radiation absorption coefficient, while the resolution is inversely proportional to the first power of the specified combination. This can be qualitatively explained as follows. The larger number of sound wave periods radiation interacts with, the higher diffraction efficiency and resolution. In this case, the length of the region of effective AO interaction decreases with increasing attenuation of the sound and absorption of the radiation.

6. **Summary**

The first time systematization of the acoustic and optical properties of liquids made it possible to estimate the value of their AO figure of merit in the THz range. It is shown that due to the low diffraction efficiency, the length of the piezoelectric transducer should be equal to the reciprocal of the absorption coefficient of the radiation. Correlation between resolution and diffraction efficiency for the AO filter as well as correlation between the number of resolvable elements and diffraction efficiency for the AO deflector based on the liquids were estimated. It is established that for the manufacture of these devices it is advisable to use saturated hydrocarbons as the medium of AO interaction.
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