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Theoretical aspects of Photoacoustic Signal Generation with Solid Crystals

Today, non-destructive analysis techniques play an important role in industrial applications and scientific, as well as technological research. Photoacoustic method is one of such non-destructive methods in which generation of acoustic waves takes place due to the absorption of the modulated incident radiation. Photoacoustic signal is a base for photoacoustic research. The generation of the photoacoustic signal is related to the nature of the cell used for investigation. Though a variety of explanations about signal generation in photoacoustic interaction are reported by many researchers, many aspects are yet to be studied in detail. While investigating a solid crystal in photoacoustics, factors as mode of operation, scheme of excitation, the shape of the cell, and pressure fluctuations in the cell are considered for analysing photoacoustic signal generation. Also, design and performance optimization of the photoacoustic cell play paramount role in determining efficiency of related signal generation. In this paper, theoretical aspects of photoacoustic signal generation for solid crystals are presented. While obtaining the expression for pressure fluctuations with solid crystal, cylindrical configuration of photoacoustic cell is preferred to get a better signal-to-noise ratio. Along with the analysis of other factors, pressure fluctuations generated by the enclosed gas in photoacoustic cell is mathematically determined.

Keywords: photoacoustic signal, solid crystal, energy transfer, light-matter interaction, non-radiative de-excitation, photoacoustic cell, photoacoustic transducers.

Introduction

A.G. Bell discovered photoacoustic effect in 1880 by using solar radiation, a chopper and an earphone as an acoustic sensor [1]. The generation of acoustic waves due to light absorption in the target material is called as photoacoustic effect [2, 3]. After the invention of laser, photoacoustic research changed drastically. When a laser beam is passed through a solid crystal placed in a closed cell, absorption of laser energy in the molecules of the crystal leads to the generation of photoacoustic signal [4, 5].

Several researchers studied various aspects of photoacoustic signal in the last three decades. Across two media interfaces, studies on heat transfer and effect of solid – gas thermal contact resistance on photoacoustic signal generation were amply discussed [6]. The pulse shape of the photoacoustic signal in the form of cylindrical ultrasonic pressure waves generated by the irradiation of laser in a weakly absorbing liquid was described [7]. In pulsed photoacoustic spectroscopy, signals have been studied to describe origin of the complex transients [8]. Attenuation and deformation of the photoacoustic signal due to thermoelastic and viscous losses were also studied [9]. The study of nano-scale temperature rises on photoacoustic signal generation based on thermal nonlinearity of optical absorbers gave a new dimension to this field [10]. Photoacoustic signal amplitude and its relationship with concentration of the gas enclosed in the cell were experimentally studied as well. The researchers also studied the stability of photoacoustic signals generated while studying trace gas components [11]. Recently theoretical aspects of transient temperature on cubic crystal surface during photoacoustic signal generation has been described [12]. Along with the other research, mathematical aspects of displacement fields of a cuboid crystal in photoacoustic signal generation in a cell are discussed [13].

Process in Photoacoustic Signal Generation

The creation of thermal energy within the sample is caused by non-radiative de-excitation processes that ordinarily occur in the photoacoustic cell [14]. After modulating the incident radiation, the creation of thermal energy within the sample will be periodic and will result in a thermal or pressure wave having the same frequency as that of modulation. The thermal wave or pressure wave transfers energy to the sample boundary, which results in a periodic temperature shift [15]. The creation of an acoustic wave in the adjacent surrounding gas to the crystal is caused by a periodic variation in the temperature at the sample's surface and this wave propagates through the volume of the gas to the detector where a signal is produced [16]. When

this detector or microphone signal is plotted as a function of wavelength, it represents a spectrum that is proportional to the crystal's absorption spectrum.

Factors involved in photoacoustic signal generation

The important factors involved in photoacoustic signal generation with a solid crystal are

- 1) Operation mode;
- 2) Method of Excitation;
- 3) Structure of the cell;
- 4) Pressure fluctuations in the cell.

Operation Mode

The confined air around the crystal in a closed photoacoustic cell will vibrate according to the modulation frequency of the source of the incident radiation. When the mode of operation is resonant, one of the resonant frequencies of the signal in the cavity will be the modulation frequency of the incident source. The pressure fluctuations in the cell will generate an acoustic wave whose amplitude will be amplified at the frequency of modulation of the incident radiation. The signal's amplification will be proportional to the quality factor Q . The relationship between the signal's resonant frequency and its bandwidth is represented by the quality factor. To amplify just the modulating frequency of the produced signal inside the cell, a significant gap between neighboring resonance frequencies, as well as a high-quality factor should be maintained [17]. The amplification of all remaining resonant frequencies will be inversely proportional to the difference between the square of modulating frequency and the square of produced signal's resonant frequency. If the photoacoustic cell is operating in resonant mode, then implemented modulation frequencies are relatively high – about 3 kHz. The goal of this selection is to reduce noise that is correlated with the reciprocal of the frequency.

External acoustic noise, noise due to amplification, and intrinsic noise of the detector are all associated with each other. For higher frequencies, the cavity length is shorter. Therefore, an intermediate value between a longer absorption length and a shorter cavity length should be preferred while choosing the resonant frequency. Mostly, smaller cavity lengths are recommended in order to have a compact cell with a faster response time [18].

Method of Excitation

The photoacoustic effect is based on sample heating caused by optical absorption, as previously stated. Periodic heating and cooling of the sample are required to generate pressure fluctuations in order to create acoustic waves that can be monitored by pressure-sensitive transducers.

Modulated Excitation

Radiation sources whose intensity changes periodically in the shape of a square or sine wave, resulting in a 50 percent duty cycle, are used in modulated excitation systems. This can be achieved by, for example, mechanically chopping light source. To overcome the barrier of the 50 percent duty cycle, option to vary the phase of the emitted radiation rather than the amplitude is preferred. The determination of UV or IR absorption spectra of opaque solid crystals is done using chopped or modulated lamps or IR sources from commercial spectrometers [19].

The most popular sources for photoacoustic analysis are modulated continuous-wave lasers. The photoacoustic cell is a crucial part of the photoacoustic effect. This fact can be used to improve signal quality via acoustic resonance. Therefore, acoustic resonance curves must be taken into account while designing photoacoustic cells.

Pulsed Excitation

Laser pulses with durations in the nanosecond range are commonly used for excitation in pulsed photoacoustic spectroscopy. Since the repetition rates are only a few Hz, the outcome is a short period of illumination followed by a considerably longer period of darkness, resulting in a low duty cycle. This causes the sample medium to expand rapidly and adiabatically, resulting in a short shock pulse. In such cases, data analysis is done in the time domain. Hence, oscilloscopes, boxcar systems, and fast A to D converters are used to record the signal. The frequency – domain transformation of the signal pulse produces a wide spectrum of acoustic frequencies up to the ultrasonic range [20]. In this way, laser beams modulated in the form

of short laser pulses generate broadband acoustic sources and a sine wave of a laser excites a single acoustic frequency.

Structure of the cell

The internal cavity of a resonant photoacoustic cell is fabricated to the appropriate size to match the acoustic wavelength of the photoacoustic signal [21]. Figure 1 shows a schematic representation of a cylindrical photoacoustic cell for a solid crystal. By attaching additional buffers to the central section of the cell, having different cross-sections, its structure can be modified. The addition of buffers helps to avoid noise caused by the cell's coupling with other measuring equipment.

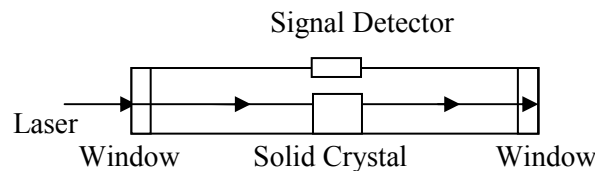


Figure 1. A schematic diagram of cylindrical photoacoustic cell for a solid crystal

A one-dimensional photoacoustic cell is a resonator with short dimensions that is oriented perpendicularly to the propagating acoustic signal. Standing acoustic waves produced by exciting sound signals are amplified when propagating sound waves are reflected back into the cell if the phase difference between the waves is 2π or, in its multiple. The phase difference is dependent upon the reflections of waves at the ends of the cell, as well as the acoustic path length. A pressure antinode will be formed at the closed end of the cell. The cell material has a higher acoustic impedance than that of air is the reason for this phenomenon [22].

Because of the matching nature with the symmetry of the laser beam, the cylindrical form of the photoacoustic cell is the most common for a small size acoustic sensor or a microphone, the measured signal is proportional to the amplitude of pressure fluctuations at its position. By placing the sensor at the node of the generated amplified wave, unwanted sound signals, generated in resonance from the external sources, can be reduced. For a cylindrical photoacoustic cell, desired value of the Q factor value can be set up to 900.

Pressure fluctuations in the cell

Let p be the pressure fluctuation in the gas, T be its temperature, and V be its volume surrounding the crystal in a closed photoacoustic cell. The change in pressure in the surrounding gas is given by

$$dp = \left(\frac{\partial p}{\partial T}\right) V dT + \left(\frac{\partial p}{\partial V}\right) T dV. \quad (1)$$

It has been assumed that (a) conduction of heat from the enclosed gas in the cell to the gas outside the cell is negligibly small, (b) the sample dilation has negligible impact on the model of mechanical piston, (c) heat energy distribution in the solid sample has uniform nature along its plane, (d) during laser absorption, only thermoelastic bending is considered. This situation is schematically represented in Figure 2.

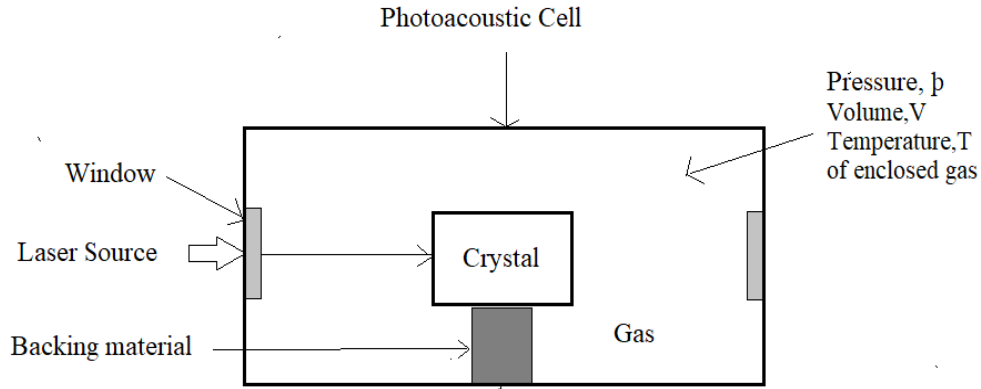


Figure 2 : A crystal kept in a Photoacoustic Cell

For ideal gas, at constant volume,

$$\left(\frac{\partial p}{\partial T}\right) = \frac{p}{T}, \tag{2}$$

At constant temperature,

$$\left(\frac{\partial p}{\partial V}\right) = -\frac{p}{V}. \tag{3}$$

Here, p is assumed to be uniform in the cell. According to the condition of structural design,

$$l_g < \frac{\Delta s}{2}. \tag{4}$$

So, the pressure fluctuation in the cell becomes

$$dp = \frac{p}{T} dT - \frac{p}{V} dV. \tag{5}$$

If σ is internal stress, ρ is density of the material of the crystal, u be the displacement field in the crystal and $\frac{\partial^2 u}{\partial t^2}$ be the second derivative of displacement field in the crystal, then

$$\left(\frac{\partial \sigma}{\partial x}\right) = \rho \frac{\partial^2 u}{\partial t^2} \tag{6}$$

According to the Landau and Lifshitz [23],

$$\sigma = -K\beta (\Delta T) \delta + 2\mu \left(\frac{\partial u}{\partial t} - \frac{\delta}{3} \frac{\partial^2 u}{\partial t^2}\right) + Ku \tag{7}$$

where K is isothermal compressibility having value

$$K = -\frac{1}{V} \frac{\partial V}{\partial p}, \tag{8}$$

at constant temperature and β is thermal expansion coefficient of the material of the crystal, whose value is

$$\beta = \frac{1}{V} \frac{\partial V}{\partial T}, \text{ at constant pressure } p. \tag{9}$$

Substituting this value in equation (6),

$$\mu \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2}\right) U + \left(K + \frac{\mu}{3}\right) \nabla \nabla u - \rho \frac{\partial^2 u}{\partial t^2} = K\beta (\Delta T). \tag{10}$$

If Δs is change in the surface area of the crystal surface due to the thermal expansion, and if $\Delta \theta$ is temperature fluctuations in the same surface, then the pressure fluctuations in the enclosed gas in the cell are given by

$$dp = \frac{p}{T} dT - \frac{p}{V} \Delta s + \frac{p}{T} \Delta \theta. \tag{11}$$

Conclusions

An exact equation for pressure fluctuations in the enclosed gas around the solid crystal kept in a cylindrical-shaped photoacoustic cell is exactly determined. This expression allows the calculation of various parameters of photoacoustic signal related to solid crystal in the designed photoacoustic cell. The signal dependence in terms of operation mode, method of excitation, and structure of the cell is theoretically presented. This theoretical approach constitutes an important step towards the determination of various

aspects of photoacoustic signal generation for solid crystals. This work will be useful in photoacoustic research to explore more scientific and industrial applications in various fields in the future.

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А.П. Сароде, О.Х. Махаджан

Қатты кристалды фотоакустикалық дабылдарды генерациялаудың теориялық аспектілері

Бүгінгі таңда беріктік бақылау әдістері өнеркәсіптік қолдануда, сонымен қатар, ғылыми жаңа технологиялық зерттеулерде маңызды рөл атқарады. Фотоакустикалық әдіс — бұл беріктік бақылау әдістерінің бірі, онда акустикалық толқындардың пайда болуы модуляцияланғаннан түскен сәулеленуді сіңіру арқылы жүреді. Фотоакустикалық дабыл фотоакустикалық зерттеулердің негізі болып табылады. Фотоакустикалық дабылдың пайда болуын зерттеу қолданылатын ұяшықтың

табиғатымен байланысты. Көптеген зерттеушілер фотоакустикалық өзара әрекеттесу кезінде дабыл генерациясының әртүрлі түсіндірмелері туралы есеп берсе де, бірқатар аспектілері әлі егжей-тегжейлі зерттелмеген. Фотоакустикадағы қатты кристалды зерттеу кезінде фотоакустикалық дабылдың пайда болуын талдау үшін жұмыс режимі, козу схемасы, ұяшық пішіні және ұяшықтағы қысымның ауытқуы сияқты факторлар ескеріледі. Сонымен қатар, фотоакустикалық ұяшықтың құрылымы мен өнімділігін онтайландыру тиісті дабылдың тиімділігін анықтауда маңызды рөл атқарады. Мақала авторлары қатты кристалдарға арналған фотоакустикалық дабылдың пайда болуының теориялық аспектілерін ұсынған. Қатты кристалды қысым флуктуациялары үшін өрнекті алған кезде, дабыл/шу арақатынасын жақсарту үшін фотоакустикалық жасушаның цилиндрлік құрылымына артықшылық беріледі. Басқа факторларды талдаумен қатар, фотоакустикалық ұяшықтағы газ шығаратын қысымның ауытқуы математикалық түрде анықталады.

Кілт сөздер: фотоакустикалық дабыл, қатты кристалл, энергияны беру, жарық пен заттың өзара әрекеттесуі, сәулеленбеген козуды өшіру, фотоакустикалық ұяшық, фотоакустикалық түрлендіргіштер.

А.П. Сароде, О.Х. Махаджан

Теоретические аспекты генерации фотоакустических сигналов с твердыми кристаллами

Сегодня методы неразрушающего анализа играют важную роль в промышленном применении, в научных, а также технологических исследованиях. Фотоакустический метод является одним из таких неразрушающих методов, при котором генерация акустических волн происходит за счет поглощения модулированного падающего излучения. Фотоакустический сигнал является основой для фотоакустических исследований. Генерация фотоакустического сигнала связана с природой ячейки, используемой для исследования. Хотя многие исследователи сообщают о различных объяснениях генерации сигналов при фотоакустическом взаимодействии, некоторые аспекты еще предстоит детально изучить. При исследовании твердого кристалла в фотоакустике для анализа генерации фотоакустического сигнала учитываются такие факторы, как режим работы, схема возбуждения, форма ячейки и колебания давления в ячейке. Кроме того, оптимизация конструкции и производительности фотоакустической ячейки играет важную роль в определении эффективности генерации соответствующего сигнала. Авторами статьи представлены теоретические аспекты генерации фотоакустического сигнала для твердых кристаллов. При получении выражения для флуктуаций давления с твердым кристаллом предпочтительна цилиндрическая конфигурация фотоакустической ячейки, чтобы получить лучшее соотношение «сигнал/шум». Наряду с анализом других факторов, математически определяются колебания давления, создаваемые заключенным газом в фотоакустической ячейке.

Ключевые слова: фотоакустический сигнал, твердый кристалл, передача энергии, взаимодействие света и вещества, безызлучательное отключение возбуждения, фотоакустическая ячейка, фотоакустические преобразователи.

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