Frequency and Wavelength Dependences of the Electro-optic Coefficients $r_{63}$ and $r_{41}$ in Congruent KDP Crystals

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Abstract: The electro-optic coefficients $r_{63}$ and $r_{41}$ are determined in congruent KDP crystals, using an experimental method based upon the direct measurement of material. Sénarmont system for electro-optic coefficient measurement and characterization of crystals was modified. This modification allowed us to obtain on the frequency dispersion dependence of the electro-optic coefficients within a frequency range up to 20 MHz and on a new version of modulation depth method. To the best of our knowledge, by using this system, the electro-optic coefficients $r_{63}$ and $r_{41}$ in different configurations (transverse and longitudinal) have been measured for the first time within a frequency range up to 20 MHz. The measurements have been investigated as a function of laser wavelength ranging from 532 nm to 750 nm. While previously these coefficients were measured at only one wavelength, 632.8 nm, and the unclamped coefficient $r_{63}$ is known at just 3390 nm. From these measurements, the clamped and unclamped electro-optic coefficients as well as the acoustic contribution have been determined.

Introduction

Potassium Dihydrogen Phosphate (KDP) crystals are among the most widely used commercial nonlinear optical (NLO) materials. In addition, they are also excellent electro-optic crystals with high electro-optic coefficients that are widely used as electro-optical modulators, Q-switches, and Pockels Cells. The dependence of the electro-optic (EO) coefficients for crystals on frequency, reflects the various physical processes contributing to the EO effect according to the working frequency [1].

In the present work, a modified Senarmont arrangement was used with a modified modulation depth method to have more accurate measurements at low and high frequencies as well as at different wavelengths. The EO coefficients are measured below and above the piezo-resonances, that are the $r^T$ (unclamped) and (clamped) $r^S$ coefficients respectively. The knowledge of these two coefficients is useful for Pockels cells. The coefficient between $r^T$ and $r^S$, is called the $r^a$ coefficient, which corresponds to the acoustic contribution.

The accurate determination of the EO coefficients over a wide range of frequencies gives information on the physical mechanism for the origin of the EO properties and on frequency ranges for various applications.

Experimental Setup

The system used in this work is called the modified Sénarmont system which is shown in Figure 1. It mainly consists of a polarizer (P), the EO sample (S), the compensator and the
The analyzer (A). The polarizer and analyzer are placed at perpendicular crossed axes. The EO sample which is the EO crystal is placed between the polarizer and the compensator (which is represented by a quarter wave-plate (Q)). Both the polarizer and the compensator are rotated at an angle of 45° relative to the crystal axis, whereas the analyzer has a free angle $\beta$. The electric field is applied to the sample by means of a voltage $V$ that is supplied from a power supply. As a result, when a laser beam passes through the system, it is transformed into an intensity-modulated beam. This modulated beam is received by a photodetection system. The photodetection system consists of a photodiode (PD) and its associated high-gain photodiode amplifier (PDAM) followed by a band-pass filter (BPF).

The modification of photodetection system gave a new dynamic technique for modulation depth method (MDM) [2,3]. By means of the direct modulation depth method (DMDM) which depends on the Sénarmont setup (Figure 1), we have shown that this technique allows to obtain the frequency dispersion of the EO coefficient from DC up to high frequency (20 MHz) with high accuracy. The DMDM method based on the direct measurements of EO coefficients, depends on observing the output signal from photodetection system when the opto-electrical bias $\bar{\varphi}$ is applied on the EO sample under test. Also, this observation of the signal leads to the acoustic contribution $r^a$ measurement with a high precision regarding to the EO effect at the operating point $M_1$ of the transfer function (Figure 2). It is known that one of the quiescent points along the I-$\bar{\varphi}$ characteristic curve is the middle point $M_1$ corresponding to the midpoint of intensity, $I=I_0/2$, of the transfer function (Figure 2). This point is also called the

**Fig. (1) : Sénarmont system**
maximum-linearity point [4], because if the ac field of frequency \( f \) is applied to the crystal a clear signal modulated at the same frequency \( f \) appears at the demodulated output. The so-called linear working point \( M_1 \) can be used to determine the EO coefficient. By measuring the peak-to-peak amplitude \( i_{pp} \) of the modulated signal at the point \( M_1 \), one can obtain the EO coefficient directly from the following equation [5]

\[
r(f) = \left( \frac{\lambda d}{\pi n^3 LV_m} \right) \frac{i_{pp}}{I_0}
\]

(1)

where \( \lambda \) is the wavelength of laser beam, \( d \) and \( L \) are the dimensions of the crystal, \( n \) is the refractive index, \( V_m \) is the amplitude of the ac field and \( I_0 = I_{\text{max}} - I_{\text{min}} \) represents the total intensity shift of the transfer function.

In Equation 1, the dimensionless ratio \( i_{pp}/I_0 \) is commonly noted and is called the intensity modulation (IM) depth.

![Image](image1.png)

**Fig. (2)**: Optical transmission of the system

**Experimental Results**

The measurements have been performed on the \( \text{KH}_2\text{PO}_4 \) (or KDP) crystal grown with water solution by slow evaporation technique. Three sample cuts were employed to explore two different configurations. The first and the second cut are 4x10x4 mm\(^3\) 45\(^0\) Y-cut and 5x10x4 mm\(^3\) Y-cut KDP crystals for the transverse EO coefficients \( r_{63} \) and \( r_{41} \), respectively. The third one was 10x5x4 mm\(^3\) 0\(^0\) X-cut KDP crystal involving the longitudinal EO coefficient \( r_{41} \). For each sample the dimension along the laser beam propagation is 10 mm. Table 1 summarizes the configurations (directions of the light propagation and the applied electric field and EO coefficient involved) that were studied.

The measurements were carried out by means of the MDM and DMDM methods by using a 15 mW He-Ne laser beam at \( \lambda=632.8 \) nm. Within experiments carried out to determine the \( r_{63} \) and \( r_{41} \) in transverse and longitudinal mode configurations. By using the DMDM method, the acoustic contribution was calculated first at \( \lambda=632.8 \) nm for KDP crystal. In addition, the unclamped EO coefficients \( r^T \) was measured at low frequency and clamped EO coefficients \( r^S \) at high frequency for frequency up to 10 MHz.

The acoustic contribution \( r_{63}^a \) was determined first by using the DMDM method with the transverse configuration. The light intensities \( I_i \) and \( i_{pp} \) were measured and Equation 1 was used to calculate the EO coefficient \( r_{63} \). The results obtained were plotted as a function of frequency \( f \) as shown in in Figure 3. This figure shows the unclamped \( r_{63}^T \) is practically constant under acoustic resonance region (under 200 kHz) with a value equal to 11.5 pm/V. But, also at high frequencies up to acoustic resonance (up to 850 kHz to 10 MHz), the clamped EO coefficient \( r_{63}^S \) has been found to remain also constant with a value of 9.7 pm/V. Accordingly, the acoustic contribution \( r_{63}^a \) to the EO coefficient, which is equal to the difference between unclamped and clamped values ( \( r_{63}^a = r_{63}^T - r_{63}^S \) ), we found to be equal to 1.8 pm/V.

![Image](image2.png)

**Fig. (3)**: Freq. dispersion of the EO coefficient \( r_{63} \)
In order to determine the acoustic contribution $r_{41}^a$ with the transverse and longitudinal configurations, the optical intensities $I_0$ and $i_{pp}$ were measured with $d=L$ for longitudinal configuration. The EO coefficients were calculated by using Equation 1 and were drawn as a function of frequency as shown in Figure 4. Figure 4 shows, the unclamped $r_{41}^T$ in transverse and longitudinal configurations was constant (stable) for low frequencies (below 100 kHz). It is equal to 8.56 pm/V and 8.2 pm/V respectively. Afterwards, for the high frequencies (from 900 kHz up to 10 MHz), the clamped EO coefficients $r_{41}^S$ found to be constant with a value of 4.53 pm/V and 4.1 pm/V for transverse and longitudinal configurations, respectively. In this case, the acoustic contribution to $r_{41}$ given by $r_{41}^a = r_{41}^T - r_{41}^S$. $r_{41}^S$ will be equal to $r_{41}^a = 4.03$ pm/V in transverse configuration and $r_{41}^S = 4.1$ pm/V in longitudinal configuration. Table 1 summarizes the value of the unclamped and clamped coefficient for all measurements.

![Acoustic Contribution](image)

**Table (1): The EO coefficient $r_{63}$ and $r_{41}$ in KDP obtained at 632.8 nm.**

<table>
<thead>
<tr>
<th>Configuration</th>
<th>Unclamped EO Coefficient $r^T$ (pm/V)</th>
<th>Clamped EO Coefficient $r^S$ (pm/V)</th>
<th>Acoustic Contribution $r^a$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Transverse</td>
<td>$r_{63}$: 11.5</td>
<td>$r_{41}$: 8.56</td>
<td>$r^a = r^T - r^S$</td>
</tr>
<tr>
<td></td>
<td>$r_{41}$: 8.56</td>
<td></td>
<td>$r_{41}^a = 4.03$ pm/V</td>
</tr>
<tr>
<td>Longitudinal</td>
<td>$r_{41}$: 8.2</td>
<td></td>
<td>$r_{41}^a = 4.1$ pm/V</td>
</tr>
</tbody>
</table>

All previous EO results were obtained at a wavelength of 632.8 nm only (by using a He-Ne laser beam) at room temperature, by using DMDM method. The values of the EO coefficients were obtained for different wavelengths also. Several lasers were used for this purpose. In addition to the He-Ne-laser, a diode-laser at $\lambda=532$ nm, and $\lambda=750$ nm wavelengths was used also. These different wavelengths allowed us to study the relation of unclamped $r^T$ and clamped $r^S$ linear EO coefficients of KDP crystal as a function of wavelength $\lambda$. The same setup of Figure 1 was used. The results are shown in

![Figure 4](image)

**Fig. (4):** Frequency dispersion of the EO coefficient $r_{41}$ in (a) transverse configuration (b) longitudinal configuration.
Figure 5. This wavelength dependence has been determined using the DMDM method.

This wavelength dependence has been determined using the DMDM method. The results obtained are numerically listed in Table 2.

Table (2): Values of unclamped $r_T^T$ and clamped $r_S^S$ EO coefficients at different wavelengths.

<table>
<thead>
<tr>
<th>$r$ (pm/V)</th>
<th>$\lambda = 532$ nm</th>
<th>$\lambda = 633$ nm</th>
<th>$\lambda = 750$ nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>$r_{63}^T$</td>
<td>12.32</td>
<td>11.5</td>
<td>10.8</td>
</tr>
<tr>
<td>$r_{63}^S$</td>
<td>10.5</td>
<td>9.7</td>
<td>9.04</td>
</tr>
<tr>
<td>$r_{41}^T$</td>
<td>9.33</td>
<td>8.56</td>
<td>7.88</td>
</tr>
<tr>
<td>$r_{41}^S$</td>
<td>5.4</td>
<td>4.53</td>
<td>3.9</td>
</tr>
</tbody>
</table>

Also, this wavelength dependences of the EO coefficients were fitted by using Sellmeier equation of the form [5],

$$ r^2(\lambda) = A + \frac{B}{\lambda^2 - C} - D\lambda^2 \quad (2) $$

Were A, B, C, and D are constants of the KDP crystal. The values of the parameters used are listed in Table 3.

Fig. (5) : The wavelength dependence of the clamped $r_S^S$ and unclamped $r_T^T$ EO coefficient (a) $r_{63}$ configuration (b) $r_{41}$ configuration
The frequency dependence for $r_{63}$ and $r_{41}$ EO coefficients are shown in Figures 3 and 4 respectively. The unclamped coefficient is nearly constant low frequencies (Table 1). But the clamped coefficient $r_{63}^s$ have been found equal to values found in reference [6]. These values may be attributed to the good accuracy of the method that was applied or differences in the temperature during the experiments. For the best of our knowledge the value of $r_{41}^s$ at transverse or longitudinal configurations are found for the first time in this work.

When using KDP crystal as an element for an IM system, it is necessary to know the values of its unclamped $r^T$ and clamped $r^S$ EO coefficients at low and high frequencies, respectively. For this reason, these values have been recorded in Table 1. The knowledge of these coefficients is indispensable, specifically for practical reasons, as e.g. for determination of the appropriate driving voltages.

Also, the difference between the levels of $r^T$ and $r^S$ is attributed to the existence of the acoustic contribution $r^a$ to the EO effect. The acoustic contribution derives from an indirect EO effect arising from the change of the refractive index caused by the deformation in piezoelectric crystals. Theoretically, the coefficient $r^a$ with the value of the product $pd$ of the elasto-optic and piezoelectric coefficients $p$ and $d$.

The knowledge of the magnitude of $r^a$ is essential for having information about the role of the piezoelectric effect in the EO modulator crystal, which is directly connected with the acoustic resonances. The experimental value for $r^a$ can be a good corresponding with the calculated values from $p$ and $d$. This comparison made only for $r_{63}^T$ (Table 1).

The corresponding results, which are presented in Table 1 show that the KDP crystal has a high value of $r_{63}^T$ with small acoustic contribution $r_{63}^a$ (180 % of the total value), while $r_{41}^T$ has a low value with large acoustic contribution $r_{41}^a$ (403 % of the total value). Accordingly, we have interest to employ the $r_{63}$ configuration in the EO modulator. Also the KDP crystal have for $r_{63}^T$ a clamped coefficient $r_{63}^S$ lower than the corresponding unclamped coefficient $r_{63}^T$. On the other hand, the EO coefficients $r_{41}^T$, $r_{41}^S$, $r_{41}^a$ have values nearly similar to that of transverse and longitudinal configurations, therefore the KDP crystal can be used in EO modulation system.

From Figure 5, the wavelength dependence of the clamped $r^S$ and unclamped $r^T$ EO coefficient have the same performance. The values increase when wavelength decreases. Also, that the acoustic contribution $r^a$ is independent of the wavelength. The large increase of the effective EO coefficient with wavelengths probably originates from the increase of both refractive indices and EO coefficient due to optical absorption. To the best of our knowledge, no measurements of this kind has been made for wavelength dependence of $r_{63}$ and $r_{41}$ in KDP crystal. The only measurement recorded was for 3390 nm which was reported in [7].
Summary and Conclusions

In this study, the unclamped values of the electro-optic coefficient $r_{63}$ and $r_{41}$ in congruent KDP crystal with new orientation were determined. Also, the Sénarmont system for electro-optic (EO) of crystal which is based on the direct measurement of material was modified. This modification allowed to obtain accuracy values of coefficients using the direct modulation depth method DMDM. This method allows to measure the EO coefficients at low and high frequencies for a range up to 10 MHz. The DMDM depends on direct measurement by observing the output signal from photodetection system, so the frequency dependence of $r_{63}$ and $r_{41}$ coefficients of KDP crystal were studied for transverse and longitudinal configurations with He-Ne laser beam at $\lambda = 632.8$ nm. In addition, the modified system allowed to measure the acoustic contribution $a_{\sigma r_{63}}$ and $a_{\sigma r_{41}}$ for crystal with high accuracy. It was found that $r_{63}^T$ and $r_{63}^S$ have large value from $r_{41}^T$ and $r_{41}^S$.

In contrast, in $r_{63}$ configuration, the acoustic contribution $a_{\sigma r_{63}}$ have small value from the EO effect. But at $r_{41}$ configuration, the acoustic contribution $a_{\sigma r_{41}}$ have large value. Therefore, the experiment gives best results for the configuration with null acoustic resonances distribution at resonance point to a void the deformation in transmitted signal and modulation.

References