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Mustapha Abarkan, Anush Danielyan, Sasha Sewastianow, Michel Aillerie, Nicéphore Théofanous, et al. The  $r = 22$  electro-optic coefficients in indium-doped congruent lithium–niobate crystals. NAMES'16 - New Achievements in Materials and Environmental Sciences, Nov 2016, Nancy, France. 10.1088/1742-6596/879/1/012005 . hal-01609060

**HAL Id: hal-01609060**

**<https://hal.science/hal-01609060>**

Submitted on 3 Oct 2017

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2017 J. Phys.: Conf. Ser. 879 012005

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# The $r_{22}$ electro-optic coefficients in indium-doped congruent lithium–niobate crystals

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**Abstract.** The high- and low- frequency electro-optic coefficients  $r_{22}$  of In-doped lithium niobate (LN) and the corresponding dielectric permittivity as well, have been experimentally determined and compared with the results obtained in undoped congruent LN crystals. Compared to pure congruent lithium niobate, a low acoustic contribution of the electro-optic and dielectric properties are originally found in indium (In)-doped congruent lithium niobate (LN:In) crystals in the low indium concentration range [0.12-1.7 mol%]. All reported results confirm that the LN:In is a very promising candidate for several non-linear devices as Pockels cells for laser Q-switching.

## 1. Introduction

It is now well established that the electro-optic effect is one of the most convenient properties that could be exploited to obtain fast Q switching. Although,  $\text{LiNbO}_3$  (LN),  $\text{LiTaO}_3$  (LT),  $\text{KTiOPO}_4$  (KTP) and  $\text{BaB}_2\text{O}_4$  (BBO) are widely used as Pockels cells for laser Q-switching [1, 2].  $\text{LiNbO}_3$  offers good transmission and high extinction ratio with modest half-wave voltage in transverse configuration  $r_{22}$  and it is not hygroscopic [3]. When it is grown in its usual composition, i.e. congruent composition, it suffers of a relatively low optical damage threshold being a major drawback of a LN Pockels cell [4]. It can be noted that this low optical damage threshold is related to high photorefractive sensitivity interesting for holographic applications. Thus, for the realization of integrated devices based on this property, LN crystals in congruent composition or doped with metallic ions are good candidate [5-9]. On contrary, the low optical damage threshold generally observed in LN crystals limits the use of LN in high power laser systems [10]. Nevertheless, doping by ions of transitions elements or rare earths makes possible to modify the physical properties and seems to be a way to mitigate this problem. The optical damage threshold is considerably increased in stoichiometric LN and in congruent LN doped with specific ions, as it depends on the amount of intrinsic defects present in the lattice. Some



researches point out that divalent ion such as  $Mg^{2+}$  [11-16] and  $Zn^{2+}$  [17-19] and more recently tetravalent ions as Hf [20-26] and Zr [27-32] can improve the optical damage resistance of LN crystals for specific concentrations. It was found a threshold concentration around  $[Hf] = 2 \text{ mol\%}$  in the melt to lead a significant increase of the photorefractive resistance and it has been found that the light-induced birefringence changes of  $LiNbO_3$  crystal doped with 4 mol% of  $HfO_2$  were comparable to that of 6 mol%  $MgO$  doped crystals [23]. It was also shown the existence of a threshold concentration around 2.0 mol% in zirconium doped LN crystals. Moreover, hafnium and zirconium ions have the advantage to possess a distribution coefficient near one at the threshold concentration. Therefore, high quality crystals can be grown easier than the usual LN:Mg crystal with 6% of  $MgO$  having a distribution coefficient closer to 1.2 [26]. This fact suggests that Hf and Zr might represent an excellent alternative for obtaining higher optical damage resistance crystals with high optical quality [27-32]. It was proved that the EO coefficients reveal a small dependence on the Hf and Zr content introduced in the LN lattice with just a kink at 2.0 mol% attributed to the strain contribution related to the introduction of Hf and Zr ions, respectively [23, 27, 28]. Otherwise, if compared to undoped congruent crystal, the hafnium and zirconium doped lithium niobate crystals present the advantage to have smaller optical damage, especially for concentration equal to 2.0 mol%, and therefore should be more suitable for EO and NLO applications [26, 29]. In these divalent and tetravalent-doped LN crystals, electro-optic behaviors were studied and the obtained results largely confirm the threshold in dopant concentrations mentioned above and as observed experimentally by other techniques. Nevertheless, only few studies mentioned the doping of LN with trivalent ions such as  $Sc^{3+}$  and  $In^{3+}$  in the aim of increasing the damage threshold for EO and NLO applications [33, 34].

Concerning this last point, we remember that piezoelectric ringing in  $LiNbO_3$  crystal in  $r_{22}$  configuration is big, as evidenced by comparison to the  $r_c$  configuration. It has been proved that in  $LiNbO_3$  Pockels cells the applied voltage induces a mechanical compression of the crystal via the piezoelectric effect, which remains for some interval of time after the voltage has been removed, creating an additional birefringence modulation via the elasto-optic effect. A polarization sensitive element inside the cavity transforms this modulation into an additional time-dependent cavity loss. The acoustic oscillations following the initial relaxation of the compression were considered irrelevant for Q switching due to their long periods, on the order of  $2 \mu s$  [1]. Abarkan et al. [35, 36] showed that the resonances amplitude have diminished when the pulse duration is short (less than 100 ns).

In the present work we report, as function of indium concentration, experimental results and analysis obtained in the characterization of the EO coefficients  $r_{222}^T$  and  $r_{222}^S$  mainly involved in Q-switch laser applications. The coefficient  $r_{222}$  is obtained when a light-beam is propagating along the optical axis (c-axis) of the crystal and, therefore with an optical polarization of the transmitted beam in the isotropic plane, which renders it unaffected by the temperature dependence of the birefringence within this configuration. The EO results would be then discussed. To complete the study, we have measured the frequency dependence of the corresponding dielectric permittivity  $\epsilon_{22}$  and finally established the figure of merit allowing the comparison between materials, used as modulator, dedicated for Q-switching applications.

Five samples were grown by the Czochralski growing method from a congruent melt with various concentrations of  $In_2O_3$  (0.12%, 0.25%, 0.5%, and 1.7%) corresponding to 0.24, 0.5, 0.9, and 2.1 at% in the crystal [33]. These crystals were considered for comparison and discussion, with a congruent lithium niobate and a Zr doped lithium niobate crystals grown from a melt with a concentration of 2mol% of  $ZrO_2$  [27, 28]. It is to be of note that the Zr doped sample was chosen as its concentration of dopant, 2 mol% of  $ZrO_2$  corresponds to the threshold concentration where the optical damage is the smaller in Zr doped lithium niobate series [37]. All samples were prepared, cut and polished at optical grade.

## 2. Electro-optic measurements

According to [38, 39], we remind that the linear or Pockels EO coefficients in dielectric materials may be described as

$$r^T = r^a + r^S. \tag{1}$$

In this relation,  $r^T$  is the EO coefficient for the unclamped sample characterized under low-frequency electric field or DC electric field,  $r^a$  is the acoustic or piezo-optic contribution related to the crystal deformation via the piezo-electric and the elasto-optic effects, and  $r^S$  is the corresponding EO coefficient for the clamped sample measured at high frequency electric field.

For EO measurements, optical interferometric and ellipsometric arrangements can be used. In the current contribution, we used the ellipsometric one with an experimental setup based on the Sénarmont setup [38]. The transfer function is presented in figure 1.

The point  $M_1$  located at 50% transmission  $(I_{\max}-I_{\min})/2$  point corresponds to the so-called linear working point. When a sinusoidal electric field is applied on the sample under tests, this point is associated with the “Modulation Depth Method” (MDM) [38] and can be used to determine the EO coefficient as a function of frequency from DC to 1 MHz. This maximum frequency limit is defined by the specifications of power supply and signal acquisition electronic apparatus used for these experiments. In this method, the peak-to-peak amplitude  $i_{pp}$  of the modulated signal is measured at the point  $M_1$ , and the EO coefficient is directly obtained from the following equation [38]:

$$r_{\text{eff}}(\nu) = \frac{2\lambda d}{\pi n_{\text{eff}}^3 I_0 L} \frac{i_{pp}(\nu)}{V_{pp}(\nu)}, \tag{2}$$

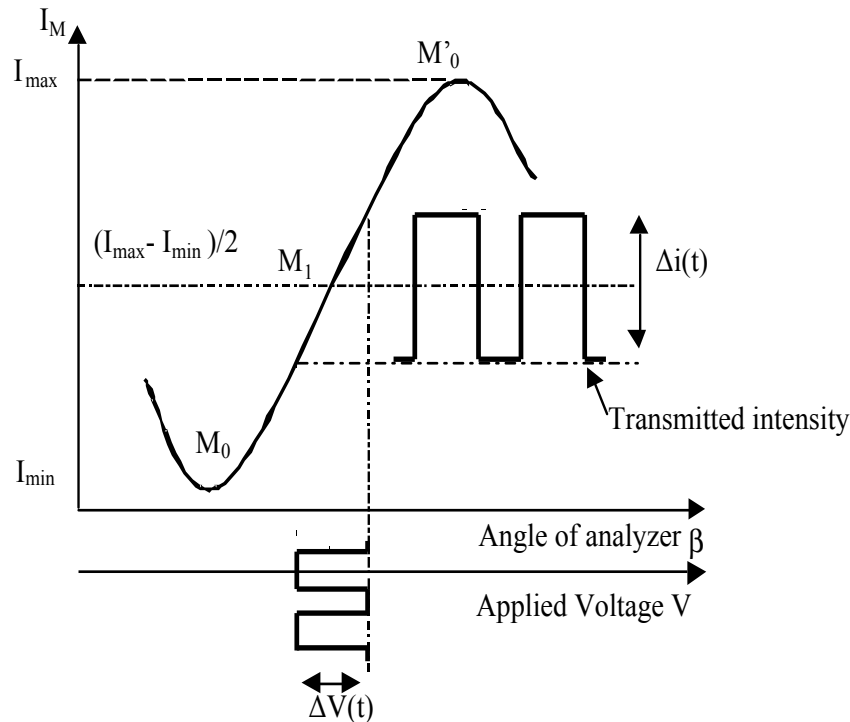
Here,  $I_0 = I_{\max}-I_{\min}$  represents the total intensity shift of the transfer function,  $L$  is the length of the crystal along the beam-propagation direction,  $d$  is the sample thickness along the applied electric field direction,  $n_{\text{eff}}$  is the effective refractive index,  $\lambda$  is the laser wavelength and  $V_{pp}$  is the peak-to-peak value of the applied ac field at the frequency  $\nu$ .

The  $M_1$  point can be also associated to a method called “Time Response Method” (TRM) [39]. The shapes of signals involved in this method are represented in figure 1. In this method, time signals  $\Delta i(t)$  and  $\Delta V(t)$  are displayed and measured on an oscilloscope screen. To obtain the various contributions of the EO coefficient this method is associated with performing power supply and oscilloscope i.e. apparatus allowing very short rising-time. At the working point  $M_1$ , the instantaneous variation of the transmitted beam intensity  $\Delta i(t)$ , induced by the applied voltage  $\Delta V(t)$  on the crystal, is given by:

$$\Delta i(t) = \frac{\pi n_{\text{eff}}^3 L I_0}{2\lambda D} r_{\text{eff}}(t) \otimes \Delta V(t), \tag{3}$$

where  $\otimes$  is the convolution operator and  $r_{\text{eff}}(t)$  is the instantaneous value of the EO coefficient.

The Z-transformation of these signals allows the determination of the frequency dependence of the response  $\Delta i(\nu)$  with the applied voltage  $\Delta V(\nu)$ . Thus, the EO coefficients can be derived within the use of the same equation 2 as established in the MDM method. This allows the full determination of the frequency dependence of the EO coefficients, possibly above their acoustic resonance if special attention is taken in the choice of apparatus. The optical response at short time leads to the clamped (high frequency) coefficient  $r^S$ , while the optical response at longer time provides the unclamped (low frequency) coefficient  $r^T$ .

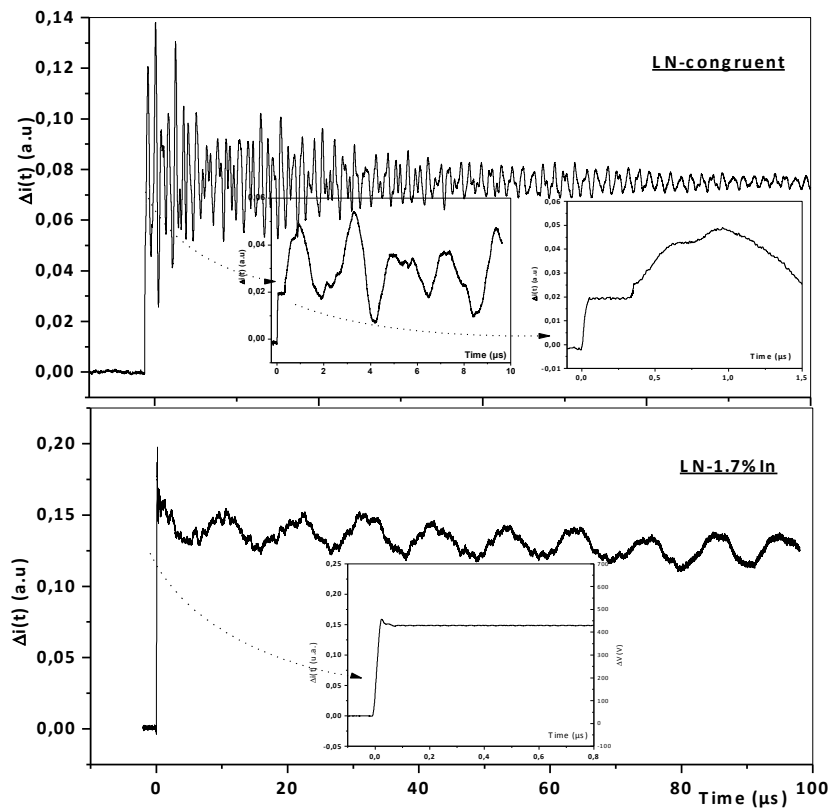


**Figure 1.** Optical transmission function of Sénarmont setup versus the analyzer  $\beta$  and applied voltage  $V$ . The point  $M_0$  is the minimum transmission point for which the output optical signal has a frequency twice the frequency of the applied electric field.  $M_1$  is the 50% transmission point yielding the linear replica of the ac voltage.

In practice, we have shown that this technique allows obtaining the frequency dispersion of the EO coefficient from DC up to at least 500 MHz, mainly limited by the rising time of the voltage pulse [38]. It is to be mentioned that the values of the coefficients obtained by these techniques are absolute values.

For the specific experiments that are concerned by this work, electro-optic measurements were carried out by both MDM and TRM methods presented above. Within experiments devoted to the determination of the EO coefficient  $r_{22}$  the beam propagates along the  $c$ -axis of the sample and an external electric field was applied along the  $a$ -axis (or equivalently the  $b$ -axis).

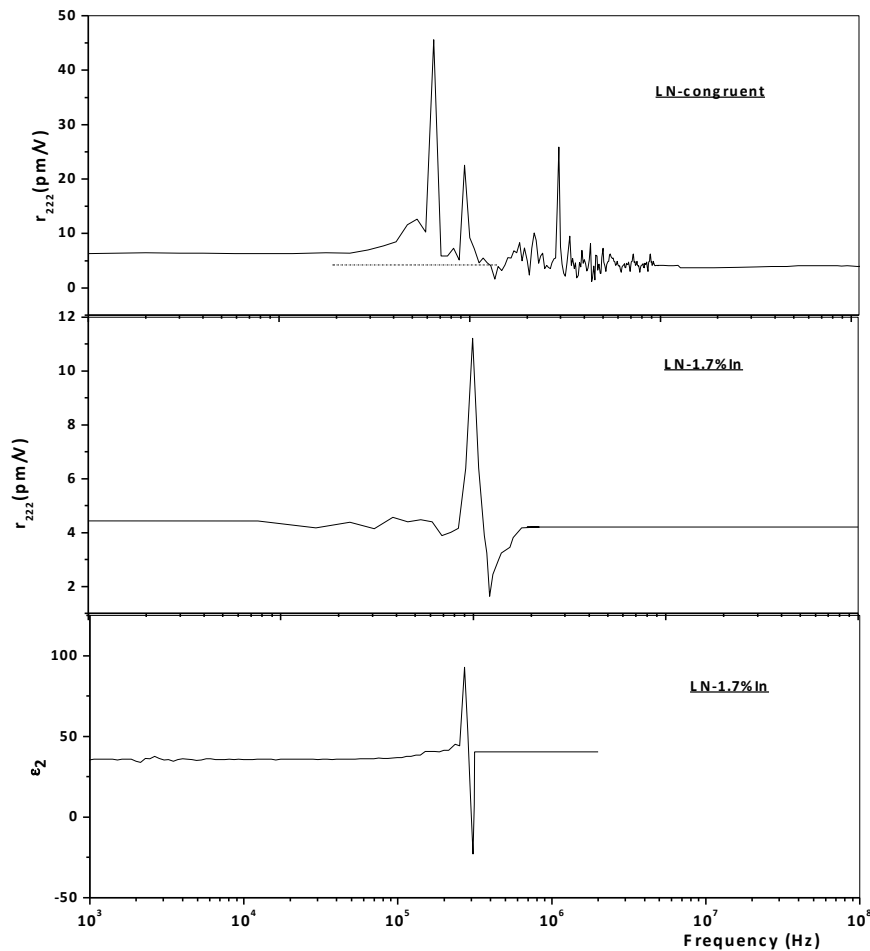
The measurements were carried out at room temperature using a He-Ne laser ( $\lambda = 633 \text{ nm}$ ). In the MDM method, we used an ac voltage of 220 V peak-to-peak at 1kHz, whereas in the TRM method, a pulse of amplitude equal to 700 V was applied onto the sample. Within these conditions, the sample dimensions and the performing optical and electric arrangements used for the experiment, the final uncertainty is in the order of 6% on the EO coefficients. Moreover, in the limit of the accuracy of the methods, no additional physical effects, as the distortion of the laser beam due to photorefractive effect or the presence of the quadratic electro-optic effect that could disturb the measurements were detected. Figure 2 shows the recording of the optical signal measured in the 1.7 mol%-In doped LN and LN in congruent composition samples at different time within the TRM method.



**Figure 2.** Responses  $\Delta i(t)$  to a step voltage  $\Delta V(t)$  at different time scales in the case of the EO coefficient  $r_{222}$  in 1.7%-In doped LN single crystal and in LN in congruent composition. Measurements were performed at the wavelength of 633nm.

In the long-time range, the optical signal oscillates with periods corresponding to the main piezo-electric frequency resonances. In the short time, the inset of figure 2 illustrates the EO response for a small time scales. For this time the oscillations do not exist since the acoustic waves need more time to propagate across the crystal. Contrary to LN in congruent composition, no difference in the level of the signal in the two time ranges is observed with In doped LN sample pointing out the negligible amplitude of the acoustic contribution.

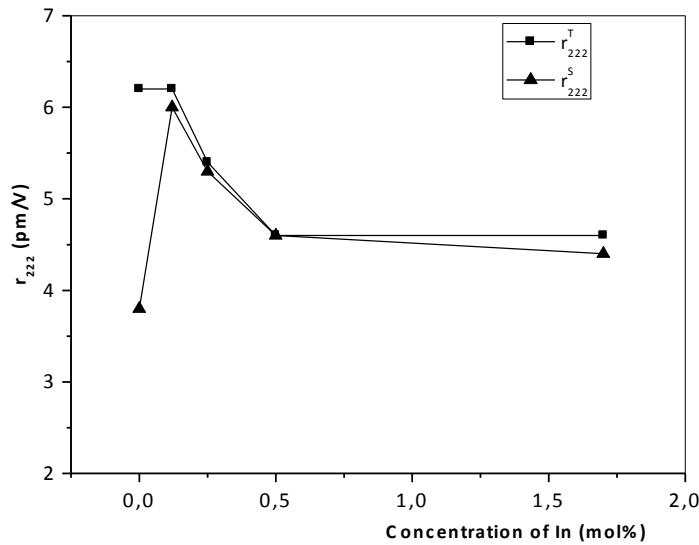
The frequency dispersion of the EO coefficient  $r_{222}$  is calculated according to equation 2. As shown in figure 3, the frequency dependence of the EO coefficient  $r_{222}$  of the crystal is flat on both sides of the piezo-resonances, giving, for this crystal doped with 1.7 mol% of indium the values of the EO coefficient at high frequency  $r_{222}^S = 4.4 \pm 0.4$  pm/V which is identical to the value at low frequency  $r_{222}^T = 4.6 \pm 0.4$  pm/V which are also in a good agreement with those measured with the MDM method (see table 1).



**Figure 3.** The frequency dispersions of the EO coefficient  $r_{222}$  in the 1.7 mol% In-doped LN crystal and Congruent LN. Comparison of the frequency dispersions of the dielectric permittivity  $\epsilon_2$  and of the EO coefficient  $r_{222}$  in the 1.7 mol% In-doped LN crystal.

The measurements of the coefficient  $r_{222}$  were performed for each crystal under investigation. All crystals present the same behavior as function of frequency. The dependence of the clamped and unclamped EO coefficients,  $r_{222}^T$  and  $r_{222}^S$  on the molar Indium concentration is shown in figure 4 and presented in table 1.





**Figure 4.** EO coefficient  $r_{222}^T$  and  $r_{222}^S$  versus In concentration in congruent LN.

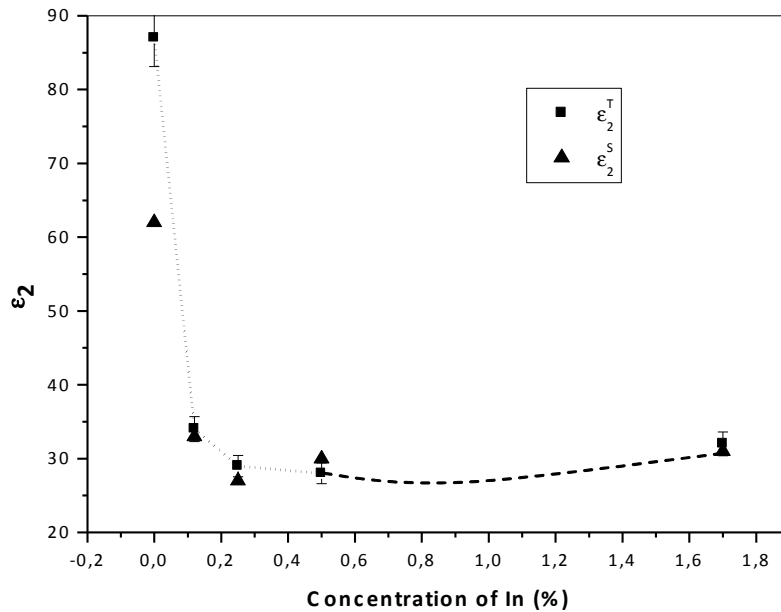
We can see in figure 4 that both clamped and unclamped EO coefficients  $r_{222}$  decrease versus indium concentration and present a kink probably at around 1.0 mol% of In in the crystal. The values of the low- and high- frequency  $r_{222}$  coefficients in In-doped LN in the case of 1.7 mol% are equal to  $r_{222}^T = 4.6 \pm 0.4$  pm/V and  $r_{222}^S = 4.4 \pm 0.4$  pm/V respectively. It corresponds to a decrease of about 25% of the value obtained in the 0.1 % In doped sample. Moreover, this result also prevents that the high frequency electro-optic coefficient value in all In doped samples remains higher than in the case of pure congruent LN pointing out a lower acoustic contribution attributed to indium incorporation.

We note that contrary to Hf-doped LN crystals [13-19], the value of  $r_{222}$  coefficient versus dopant concentration in LN:In crystals has to be emphasized since this coefficient presents a non-monotonous dependence as already observed for crystals doped with other ions such as Zr, Zn or Mg [27, 28, 37, 40-42].

### 3. Dielectric measurements

EO properties are linked to the linear dielectric properties in ferroelectric inorganic materials. The frequency dependence [43] of an EO coefficient reproduces the behavior of the corresponding dielectric permittivity  $\epsilon$  and a link between  $\epsilon$  and  $r$  still exists in their dependence on the doping composition.

Using a low voltage equal to 1 V, inducing an electric field in sample under test equal to 1 kV/m, the measurements of the dielectric permittivity  $\epsilon_{22}$  as function of frequency for all crystal-samples were done by means of two impedance analyzers HP4151 and HP4191A in frequency ranges from 1 Hz to 13 MHz and from 1 MHz to 1GHz, respectively. The frequency dispersion of dielectric permittivity  $\epsilon_{22}$  of the 1.7 mol%-In doped LN crystal measured along the c-axis is shown in figure. 5. All samples present the same general response with the frequency being such that the In-concentration dependence of the clamped and unclamped permittivities  $\epsilon_{22}^T$  and  $\epsilon_{22}^S$  present a same kink-behavior as the EO coefficient  $r_{222}$  shown in figure 4.



**Figure 5.** Dielectric constant  $\epsilon_{22}^T$  and  $\epsilon_{22}^S$  versus In concentration in congruent LN.

All data are reported in table 1. As expected, the behavior of  $\epsilon_{22}$  with frequency is very similar to this of  $r_{222}$ . In figure 3, we can see that  $\epsilon_{22}$  remains constant and displays only a very small jump between both sides of the piezoelectric resonances. We remind that this jump corresponds to the small electromechanical contribution  $\Delta\epsilon$  to the static permittivity  $\epsilon^T$ . This will be commented and discussed below.

**Table 1.** Absolute values of the  $r_{222}$  EO coefficient and related parameters of LN:Zr crystals as function of zirconium concentration: The EO coefficients at constant stress ( $r^T$ ) were obtained by both MDM and TRM methods and at constant strain ( $r^S$ ) by the TRM method, at 633 nm and at room temperature. The dielectric permittivities  $\epsilon_{22}^T$  and  $\epsilon_{22}^S$  were measured at room temperature. The figure of merit  $F = n^7(r_{222}^S)^2/\epsilon_{22}^S$  was calculated from experimental values.

	MDM method	TRM method		Acoustic contribution	Dielectric constants		Figure of merit
	$r_{222}^T$ (pm/V)	$r_{222}^T$ (pm/V)	$r_{222}^S$ (pm/V)	$r_{222}^T - r_{222}^S$ (pm/V)	$\epsilon_{22}^T$	$\epsilon_{22}^S$	$n^7(r_{222}^S)^2/\epsilon_{22}^S$ (pm/V) <sup>2</sup>
LN-Cg	6,4±0.4	6,6±0.4	3,9±0.3	<b>2.7±0.4</b>	87±5	62±3	<b>79</b>
2%ZrO <sub>2</sub>	5.2±0.4	5±0.4	2.9±0.3	<b>2.1±0.4</b>	72±4	51±3	<b>157</b>
1.7%In	4.8±0.3	4.6±0.3	4.4±0.3	<b>0.2 ±0.4</b>	32±1	31±1	<b>200</b>
0.5%In	4.6±0.3	4.6±0.3	4.5±0.3	<b>0.1 ±0.4</b>	32±1	30±1	<b>216</b>
0.25%In	5.5±0.4	5.4±0.4	5.3±0.4	<b>0.1 ±0.4</b>	29±1	27±1	<b>333</b>
0.12%In	6.4±0.4	6.2±0.4	6 ±0.4	<b>0.2 ±0.4</b>	34±2	33±2	<b>349</b>

#### 4. Discussion

In inorganic crystals the contribution arising from the optical phonons is responsible for a large value of the EO coefficient at high frequency  $r^S$  [43]. The acoustic contribution can be thus derived from the experimental values of the EO coefficients measured below and above the piezo-electric

resonances. Compared to congruent composition where we found  $r_{222}^a = r_{222}^T - r_{222}^S = 2.7 \pm 0.4$  pm/V, we can observe a small dependence of the acoustic contribution in the case of the In-doped LN crystals, which is found  $r_{222}^a = r_{222}^T - r_{222}^S = 0.2 \pm 0.4$  pm/V.

The dependence of all physical properties on indium concentration in lithium niobate is still not available. Nevertheless, we are able to estimate the acousto-optic contribution to the electro-optic effect,  $r^a$  from the elasto-optic (Pockels)  $P_{ijkl}^E$  at constant electric field, and from the piezoelectric  $d_{kij}$  tensors and the electromechanical contribution to the static permittivity, i.e. the difference  $\Delta\epsilon_{ij}$  between the low- and the high- frequency values ( $\epsilon^T$  and  $\epsilon^S$ , respectively) of dielectric permittivity recorded on both sides of the acoustic resonances [43]

$$r_{ij,k}^a = \sum_{lm} P_{ij,lm}^E d_{lm,k}, \quad (4)$$

$$\Delta\epsilon_{ij} = \epsilon_{ij}^T - \epsilon_{ij}^S = \sum_{kl} d_{ij,k} e_{kl} = \sum_{kl} d_{ij,k} C_{ijkl}^E d_{l,ij}, \quad (5)$$

where  $e$  is the piezoelectric stress tensor and  $C^E$  is the tensor of the elastic constants at constant electric field or elastic stiffness.

It is to be noted that the coefficients  $P_{ij,lm}^E$  can be also expressed with coefficients  $C_{ijkl}^E$  and  $e_{kl}$  using the thermodynamic relations. According to the point group 3m of LN crystal, [44] and by application of Neumann's principle to the  $p$ ,  $d$  and  $C^E$  tensors followed by the use of the reduced-subscript notation, we obtain the piezo-optic contribution to  $r_{222}$  and to  $\epsilon_{22}$  from Eqs. 4 and 5 as

$$r_{222}^a = -(p_{11}^E - p_{12}^E)d_{22} + p_{14}^E d_{15}, \quad (6)$$

$$\Delta\epsilon_{22} = 2d_{22}^2 (C_{11}^E - C_{12}^E) - 4d_{22}d_{15}C_{14}^E + d_{15}^2 C_{44}^E, \quad (7)$$

Using the values of piezo-electric and elasto-optic coefficients, available in literature [45, 47] for the pure congruent composition only, we found (Eq. 6)  $r_{222}^a = 2.7$  pmV<sup>-1</sup> in a good agreement with the experimental value, within the experimental error (10%). This piezo-optic contribution is relatively large in LN since it constitutes nearly 40% of the total value  $r_{222}^T \sim 6.6$  pmV<sup>-1</sup>. Within Eq.7, also evaluated for pure congruent lithium niobate, we found  $\Delta\epsilon_{22} = 35$ , which is close to the step directly detected in the experiments between both sides of the piezo-resonances.

We have demonstrated in our experiments that both the unclamped and clamped EO coefficient  $r_{222}$  and the dielectric permittivity  $\epsilon_{22}$  decrease and present probably a kink around In-doped LN 1 mol%. From the above consideration, the small step between low- and high-frequencies in both the EO coefficient  $r_{222}$  and the dielectric permittivity  $\epsilon_{22}$  in LN:In is mainly due to the quasi-absence of electromechanical contribution in LN:In, which constitutes the large contribution in the congruent material. We also note that the term included  $C_{44}$  is the largest in the congruent crystal even if it is too small to explain itself the piezo-optic contribution to  $\epsilon_{22}$ . Thus, the important changes in the electro-optic coefficient and dielectric constant can be attributed to large changes in piezo-electric and elasto-optic coefficients in doped crystals and generally to the strain effects along a (or b) axis.

Furthermore, indium is an important optical damage resistance dopant in lithium niobate, which would improve the photoconductivity  $\sigma_{ph}$  of the crystal and we can note that indium impurity is the most “efficient” among optical damage resistant ions due to the lowest threshold concentration, which was believed about 1.5–2 mol % [34]. It is important to emphasize that incorporation of In impurity ions do not affect the optical absorption of lithium niobate compared to the spectrum of pure congruent lithium niobate grown from the same charge [34].

In addition, as concerns optoelectronic applications of lithium niobate crystals, we have calculated the figure of merit linked to the driving voltage and the power (switching speed) of an EO modulator [44] or of a Pockels cell [37]. This figure of merit qualify EO devices used as modulators or Q-switches and is defined as  $F = n^7(r_{222}^S)^2/\epsilon_{22}$ . The values of F obtained in the present study for the LN:In samples are listed in table 1. LN:In crystals exhibit higher values of the figure of merit F compared to crystals of pure congruent LN composition. Thus, LN:In is a good candidate for Pockels cell [23]. Moreover, in LN:In series, the figure of merit F is improved for crystal having a concentration of 1.7 mol% of indium. This concentration corresponds also to an increase of the optical damage resistance threshold [34]. In the other hand, comparing to the latter, the indium-doped crystal possesses several advantages, such as larger photoconductivity, lower absorption coefficient, and higher photorefractive sensitivity [34, 48]. So, the current results and analyzes point out the high potential of LN:In to be a promising material for EO modulation and Q-switching applications.

## 5. Conclusion

In a series of Indium doped lithium niobate crystals, the frequency dependence of the EO coefficient  $r_{222}$  and of the associated permittivity  $\epsilon_{22}$  are experimentally measured and the piezo-optic contributions were calculated. The piezo-optic contribution to the EO coefficient was found much smaller in  $r_{222}$  compared to congruent composition. This is mainly related to the smaller deformation along the a (b) axis induces by the applied electric field as reflected by the difference between the low-and high-frequency values of  $\epsilon_{22}$ . Both EO and dielectric coefficients reveal a dependence on the In content introduced in the LN lattice with a kink probably located at 1.0 mol% and attributed to the introduction of In ions and to the related strain contribution. Compared to undoped congruent crystal, the indium doped lithium niobate crystals present the advantage to have smaller optical damage, especially for concentration equal 1.7 mol%, and therefore is a promising material for EO modulation and Q-switching applications.

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