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► **To cite this version:**

Marc D. Fontana, Mustapha Abarkan, Jean-Paul Salvestrini. Calculation of the dispersion of the electro-optical and second harmonic coefficients from the refractive index dispersion. *Optical Materials*, 2014, 36 (4), pp.764-768. 10.1016/j.optmat.2013.11.020 . hal-01077689

HAL Id: hal-01077689

<https://hal.science/hal-01077689v1>

Submitted on 30 May 2024

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Calculation of the dispersion of the electro-optical and second harmonic coefficients from the refractive index dispersion

M.D. Fontana ^{a,b,*}, M. Abarkan ^{c,*}, J.P. Salvestrini ^{a,b}

^a Université de Lorraine, LMOPS EA 4423, 57070 Metz, France

^b Supelec, LMOPS EA 4423, 57070 Metz, France

^c Université Sidi Mohammed Ben Abdellah, Faculte Polydisciplinaire de Taza, LIMA0-Taza, Morocco

The relationships of the wavelength dependence of both the electro-optic and second harmonic generation coefficients are established within the same model applied to nonlinear optical materials. It is demonstrated that the dispersion of these coefficients can be obtained from the own dependence of the refractive indices only, without any fitted parameter. Solely the measurement of the coefficient at one wavelength is required within this approach. A very good agreement in the λ -dependence of electrooptic and second harmonic generation coefficients is achieved between calculated values and experimental data in SBN, DAST, KTP and KNbO₃ crystals.

1. Introduction

It is of a prime importance to get the knowledge of the λ -dependence of the nonlinear susceptibility $\chi^{(2)}$ tensor coefficients to find the best conditions to use nonlinear optical (NLO) materials in devices, or to optimize the properties of a material. Unfortunately the Pockels electrooptic (EO) and second harmonic generation (SHG) coefficients are generally measured at one or two particular wavelengths only. The measurements are time consuming and require special preparation of the crystals, such as cutting for a particular shape and face polishing. Therefore the prediction of the values via models is needed. The calculation of the second-order susceptibility (SOS) has been the object of many studies since several decades. After the earlier work of Bloembergen [1] who formulates at first the development of the NLO polarization, Kurtz and Robinson [2] or Garrett and Robinson [3] (see also the textbook of Boyd [4]) have expressed different formulae of the EO and SHG coefficients. At that time, experimental data in NLO materials were relatively rare and numerous results are available since 15 years only. Surprisingly, just a few studies were so far concerned by the comparison of the predicted values of NLO coefficients with the experimental data. Seres [5] reported a model of the wavelength dependence of the SHG coefficients and his calculations were compared with data in KTiOPO₄ (KTP) and LiNbO₃ (LN).

Furthermore, Wang [6] derived, from a model of the quadratic NLO susceptibility, the values of Pockels EO and SHG coefficients along c -axis in many ferroelectric materials. The results were in agreement within a factor 2 with experimental data. These both models therefore do not afford a completely suitable description of the λ -dependence of $\chi^{(2)}$ coefficients. In addition second-order susceptibilities have been computed within the framework of density functional theory. These calculations provided electronic band structure and optical properties which match the experimental data, in particular in borate crystals such as lanthanum calcium borate [7]. However EO coefficients are not given within this approach. Here we propose a way to calculate in any NLO crystal the dispersion of both EO and SHG coefficients from the λ -dependence of the refractive indices only. For this, by contrast with the studies of Seres [5] and Wang [6], we start from a formulation close to this used in pioneer works [1–3]. Thus, from an anharmonic-oscillator model we can derive the SOS and thus both the SHG and EO coefficients. For this, we successively consider the SOS in which, either the frequency of both interacting fields are identical (ω) and in the optical range (case of the SHG), and then one frequency (Ω) of one field is much smaller than the optical field frequency (ω) (case of the EO process). We demonstrate that the wavelength dependence of these coefficients depends only on the dispersion of the linear refractive indices. We show that solely one adjustable parameter is required to obtain the behavior of the EO and SHG coefficients as a function of the laser wavelength, or as well as the temperature dependence of the EO coefficient. The validity of our approach is shown by the good agreement which is achieved between our calculations and experimental data, in different NLO crystals.

* Corresponding authors. Addresses: Supelec, LMOPS EA 4423, 57070 Metz, France (M.D. Fontana), Faculte Polydisciplinaire de Taza, LIMA0-Taza, BP 1223 TAZA Gare, Morocco. Tel.: +212 660254892 (M. Abarkan).

E-mail addresses: fontana@supelec.metz.fr (M.D. Fontana), mustapha_abarkan@yahoo.fr (M. Abarkan).

2. Model

2.1. Model description

In the general formulation of second-order NLO process the mixing between two monochromatic waves with angular frequencies ω_1 and ω_2 both in optical range are considered and the induced polarization $P^{(2)}(t)$ as due to the second order susceptibility results [4] from the superposition of signals oscillating at frequency 0, $2\omega_1$, $2\omega_2$, $\omega_1 + \omega_2$, and $\omega_1 - \omega_2$. Within the microscopic mechanism generally used, the material is assumed to consist into independent electronic oscillators and the NLO polarization arises from an anharmonic cubic potential [2,8]. Therefore this model yields the description of the NLO response, and thus, the electronic (or direct) EO coefficients in terms of electronic distortion only. Here we aim to report on a model which should be able to provide the dispersive behavior of EO and SHG coefficients as well. As a consequence we afford two changes to the general formalism. First we consider the EO Pockels effect as a peculiar NL process where the frequencies are very different ($\omega_2 \ll \omega_1$) by contrast of the SHG effect, which is widely reported in the literature, for which the frequencies are equal $\omega_1 = \omega_2$. It is worth to note that whereas in the SHG process the sole contribution is electronic, in the EO effect, in addition to the direct electronic contribution, another contribution arises from indirect modulation by the external electric field of the outer electrons via the lattice vibration and mechanical deformation [8,9]. Therefore we extend the anharmonic model to take into account of the electron-ion interaction. It is the second change we made with respect to the general formalism. The potential of the electron is thus rewritten under the form

$$V(x) = \frac{m\omega_0^2}{2}x^2 + \frac{B}{3}x^3 + \frac{C}{2}x^2X \quad (1)$$

where ω_0 is the main electronic resonance frequency, m and x are respectively the mass and displacement of electron, and X is the ion displacement. B and C are constants for a material. The first and the second terms are the harmonic and anharmonic contributions of the electronic motion whereas the third term arises from the interaction of the electron with the ionic core. Thus the indirect EO contribution arising from the modulation via the ionic lattice can be considered by this last term. This means that we can express the EO coefficients at any frequency of the modulated applied field. As reflected by Eq. (1), it is assumed, as usually, that the linear susceptibility or the refractive index has a main contribution arising from one single oscillator with an angular frequency ω_0 lying in the visible (blue-green) or UV range. In a lossless material the SOS is generally assumed to be independent of the frequencies of the electric fields, so that the permutation can be done on the indices i, j, k denoting the field components. This is known as the Kleinman symmetry condition, leading to the contracted notation used in the description of the symmetry properties of the SOS. This assumption is not taken within our study since essentially the dispersion of the SOS is aimed, and our formulation explicitly separates the indices i, j and k . Consistently, we do not suppose that refractive indices have the same resonances along the principal axes. In our re-formulation of the dispersion of second-order coefficients we specify as explained above, the subscripts i, j and k and we stress the quantities which are depending on the frequency. At last, we do not consider here any local field correction which is generally needed to insure the link between microscopic and macroscopic formulations. This effect is not included in our equations since we are interested to the calculation of macroscopic linear and nonlinear susceptibilities, and their comparison with experimental values. Now we will derive expressions of the SOS susceptibility, and the EO Pockels and SHG coefficients as well.

2.2. Expression of the SHG coefficients

At first is expressed the dispersion of the SHG coefficients. In this case, only the two first terms of the potential in Eq. (1) are considered. From the polarization term at frequency 2ω the second order susceptibility and the SHG coefficients d , can be derived as

$$\chi_{ijk}^{SHG}(2\omega, \omega, \omega) = \frac{B}{(\omega_0^2 - (2\omega)^2)_i} \chi_j(\omega) \chi_k(\omega) \quad (2)$$

or

$$\chi_{ijk}^{SHG}(2\omega, \omega, \omega) = B \frac{(n_j^2(\omega) - 1)(n_k^2(\omega) - 1)}{(\omega_0^2 - (2\omega)^2)_i} \quad (3)$$

where n is the refractive index and χ the linear susceptibility, and

$$d_{ijk}(\omega) = \frac{1}{2} \chi_{ijk}^{SHG}(2\omega, \omega, \omega) \quad (4)$$

Since B is a material constant, Eq. (3) shows that the frequency dependence of the SHG coefficients depends on the dispersion of the linear refractive indices only. We can underline that this factor B introduced in the potential (electronic anharmonic coefficient) corresponds in fact to the well known Miller constant Δ [2-4] defined as

$$\chi_{ijk}^{SHG}(2\omega, \omega, \omega) = \Delta_{ijk}(2\omega, \omega, \omega) \chi_i(2\omega) \chi_j(\omega) \chi_k(\omega) \quad (5)$$

It is observed in Eq. (3) that the index i plays a different role from indices j and k . As $(\omega_0^2 - (2\omega)^2)_i$ differs from $n_i^2(\omega) - 1$, Eq. (2) therefore does not hold with Kleinman symmetry rule. This means that even if the invariance between the indices j and k holds, the permutation between (i, j, k) cannot be applied.

2.3. Expression of EO coefficients

To express the EO Pockels effect in terms of second order process, a wave with a frequency ω_2 much smaller than the other ω_1 is considered. Then the NLO polarization and therefore the SOS can be rewritten in terms of the electrical frequency $\omega_2 = \Omega$ and the optical frequency $\omega_1 = \omega$ as

$$\chi_{ijk}^{EO}(\omega, \Omega) = \frac{\chi_j(\omega)}{(\omega_0^2 - \omega^2)_i} (B\chi_k^E + C\chi_k^I(\Omega)) \quad (6)$$

where $\chi(\omega)$ is the linear susceptibility at optical frequency, χ^E and χ^I are respectively the electronic and ionic contributions of the linear susceptibility (or permittivity), as determined far from electronic and ionic resonances. As usually considered, the index k denotes the direction of the external electric field and i and j correspond to the polarization components of the laser field. It is worth to note that the second factor in Eq. (6) is independent of the optical frequency ω (or laser wavelength).

The EO Pockels coefficient is then derived from the SOS by

$$r_{ijk}(\omega) = 2 \frac{\chi_{ijk}^{EO}(\omega, \Omega)}{n_i^2(\omega) n_j^2(\omega)}. \quad (7)$$

The EO coefficient thus depends on both frequencies, i.e. the frequency Ω of the modulating electric field and the optical frequency ω of the laser field. The dependence of the EO coefficient can be derived separately for each frequency from Eqs. (6) and (7). Thus if Ω is fixed we get

$$\chi_{ijk}^{EO}(\omega) = \frac{n_j^2(\omega) - 1}{(\omega_0^2 - \omega^2)_i} a_{\Omega,ijk}^{EO} \quad (8)$$

where $a_{\Omega,ijk}^{EO}$ is a parameter independent of ω derived from the second factor of Eq. (6). Eq. (8) therefore provides the dependence of the EO coefficients on the optical frequency (or laser wavelength)

from the dispersion of the linear refractive indices only. A second form can be deduced from Eq. (6), to express the dependence of the EO coefficient on the second frequency, i.e. the modulation frequency Ω . Thus if ω is fixed, we get

$$\chi_{ijk}^{EO}(\Omega) = a_\omega + b_\omega \Delta \chi_k^i(\Omega) \quad (9)$$

where a_ω and b_ω are parameters independent of Ω . It is worth to have a look on the role of the indices i, j, k in all above equations. We can note that even if the refractive indices vary as $(\omega_0^2 - \omega^2)^{-1}$ the indices i and j are not always equivalent since the resonance frequency ω_0 is not necessarily lying at the same position for each principal direction. This point is in opposite to the usual statement for Pockels coefficients, for which i and j generally play the same role, so that the Voigt rule can be applied and i and j are thus replaced by a sole contracted index. In fact this holds any time except for the study of the dispersion of EO coefficients in the vicinity of resonances. Furthermore Eq. (6) can be expressed in terms of the addition of the electronic and ionic contributions of the EO susceptibility. The electronic or direct EO effect arises from the modulation of electron clouds directly by the applied electric field whereas the ionic EO contribution comes from the indirect modulation via the lattice [9]. These contributions are respectively proportional to the electronic and ionic contributions of the linear susceptibility, and therefore can be responsible for the dispersion of EO coefficients vs the frequency Ω . In organic materials it is known that generally the linear permittivity (or susceptibility) is nearly constant in a wide frequency range and therefore has a value close to the refractive index squared, so that Eq. (6) becomes

$$\chi_{ijk}^{EO}(\omega) = \frac{n_j^2(\omega) - 1}{(\omega_0^2 - \omega^2)_i} (B + C) n_k^2 \quad (10)$$

where B and C are the coefficients defined in Eq. (1). By contrast, in inorganic materials, the electronic susceptibility (or the refractive index squared) is generally much smaller than the susceptibility measured at lower frequency, so that Eq. (10) becomes

$$\chi_{ijk}^{EO}(\omega) = \frac{n_j^2(\omega) - 1}{(\omega_0^2 - \omega^2)_i} C \chi_{jk}^0 \quad (11)$$

where χ_k^0 is the static susceptibility. Therefore the enhancement of the EO coefficient in inorganic materials is generally attributed to a larger permittivity. However, this holds only if the coefficient C introduced in the anharmonic model and reflecting the modulation of the electronic cloud by the ionic motion is sufficiently large. For any material the electronic contribution of the EO susceptibility can be expressed from Eq. (6) as

$$\chi_{ijk}^{EO,E}(\omega) = \frac{n_j^2(\omega) - 1}{(\omega_0^2 - \omega^2)_i} B \chi_k^E \quad (12)$$

Since it is due to electronic processes, the electronic EO coefficient r_{ijk}^E can be linked to the NL susceptibility appearing to the SHG given by Eq. (3). If χ_k^E is no longer assumed to be constant, we find

$$r_{ijk}^{EO,E}(\omega) = \frac{d_{ijk}^{SHG}(\omega)}{n_i^2(\omega) n_j^2(\omega)} \frac{4(\omega_0^2 - (2\omega)^2)_i}{(\omega_0^2 - \omega^2)_i} \quad (13)$$

We can pay attention to the fact that $(\omega_0^2 - (2\omega)^2)$ and $(\omega_0^2 - \omega^2)$ are equal for frequency ω very far from resonances. Only in this case, Eq. (11) is equivalent to the usual form given in the literature [9] $r_{ijk}^{EO,E} = \frac{4d_{ijk}}{n_i^2 n_j^2}$.

3. Results

3.1. Calculations of the EO coefficients

We apply Eqs. (2) and (6) to various NLO crystals, to describe the wavelength dispersion of the second order coefficients and even the temperature dependence of the EO coefficient. It was shown above that the dispersion of SHG and EO coefficients are simply expressed as function of the dependence of the refractive indices. So we consider this dispersion of the refractive indices to be included in Eqs. (2) and (6), under the usual Sellmeier form

$$n(\lambda) = \alpha + \frac{\beta}{\lambda^2 - \lambda_0^2} - \gamma \lambda^2 \quad (14)$$

where α, β and γ are constants as determined from the fit of $n(\lambda)$. Then we can successively calculate the wavelength dependence of the EO and SHG coefficients in several crystals. The EO coefficient given by Eqs. (7) and (8), can be re-expressed as function of the wavelength, instead of the angular frequency, as

$$r_{ijk}^{T(S)} = -\frac{1}{2\pi^2 c^2} \frac{n_j^2(\lambda) - 1}{n_i^2(\lambda) n_j^2(\lambda)} \frac{\lambda_{0i}^2 \lambda^2}{\lambda^2 - \lambda_{0i}^2} a_k^{T(S)} \quad (15)$$

where the superscripts T and S denote the constant strain and constant stress coefficients, respectively, and $a_k^{T(S)}$ is a parameter related

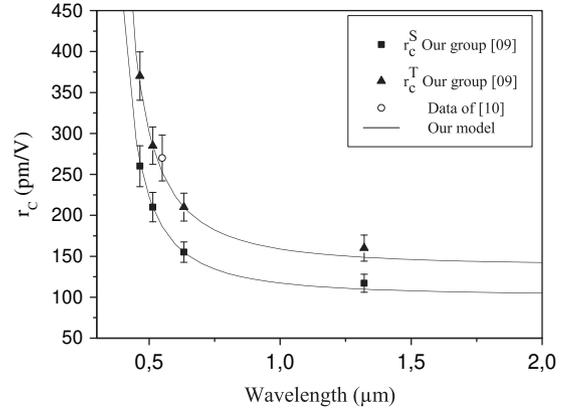


Fig. 1. Dispersion of the effective EO coefficient r_c^S and r_c^T in SBN:60 single crystal. The dots correspond to the measured values obtained by our group [10] and Trivedi et al. [11], and the line is the calculated dispersion obtained via our model.

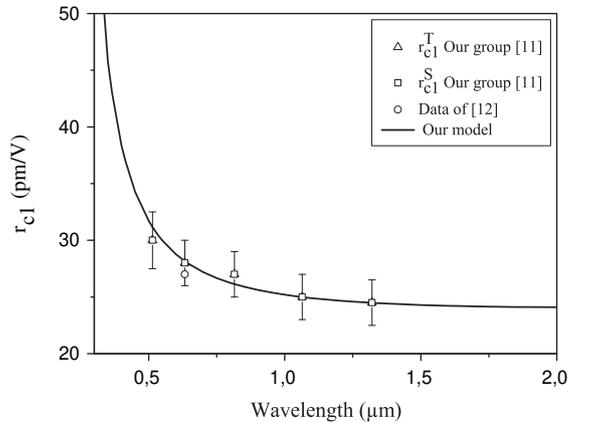


Fig. 2. Dispersion of the effective EO coefficient r_{c1}^S and r_{c1}^T in KTP crystal. The dots correspond to the measured values obtained by our group [12] and Boulanger et al. [13] and the line is the calculated dispersion obtained via our model.

Table 1
Values of the parameters used in our study. α , β , γ , and λ_{0i} are the Sellmeier coefficients given in Refs. [14,15] for SBN and KTP crystals, respectively. The terms a_{ijk}^T and a_{ijk}^S are the parameter values calculated from experimental value of the EO coefficient in the SBN:60 and KTP single crystals.

Crystal	Polarization direction	α	β (μm^2)	γ (μm^{-2})	λ_{0i} (μm)	a_{ijk}^T ($\text{mV}^{-1} \text{s}^{-2}$)	a_{ijk}^S ($\text{mV}^{-1} \text{s}^{-2}$)
SBN	X	5.002	0.1272	0.0520	0.352	1.4×10^{22}	1.03×10^{22}
	Z	4.8866	0.1187	0.0480	0.342		
KTP	X	2.9971	0.0410	0.0125	0.1958	2×10^{21}	2×10^{21}
	Y	3.0197	0.0440	0.0120	0.2050		
	Z	3.3055	0.0632	0.0139	0.2116		

to the constants B and C appearing in the anharmonic potential. In fact, as we are interested to describe the relative change of the EO coefficients versus λ , this parameter $a_k^{T(S)}$ at constant Ω , can be easily calculated from experimental value of the EO coefficient obtained at any wavelength λ' . The dispersion of the EO coefficient can be thus calculated via Eq. (15) from the dispersion of the refractive indices as given by experimental data and Eq. (14). This means that within our approach, only the measurement of the EO coefficient at one wavelength is necessary and the dependence of the EO coefficient on the wavelength in a wide range can be deduced from this value and the corresponding dependence of the indices. It was proved that the results are independent of the choice of λ' since the same dispersion was indeed provided by calculations via Eq. (15) for different λ' . It is worth to be noted that Eq. (15) is valid at low and high frequency Ω of the external electric field. As a consequence the clamped (or constant strain) r^S and unclamped (or constant stress) r^T EO coefficients can be expressed as function of λ . It is reminded that the coefficient r^S comes from the addition of the electronic and ionic contributions of the EO process discussed above, whereas r^T contains an additional contribution coming from the crystal deformation via the piezo-optic effect [9,8]. It is thus remarkable that the high frequency r^S and low frequency r^T EO coefficients can be described by the same λ -dependence. Only the fitted parameter $a_k^{T(S)}$ differs ($a^T \neq a^S$) and should be determined at a peculiar λ' . This calculation procedure was applied to three different and well known EO crystals. These materials are completely different from the point of view of the chemical structure and the crystal symmetry. The fact that our model can be successfully applied to them reinforces its validity.

At first the dispersions of the EO coefficient $r_c = r_{33} - \frac{n_0^3}{n_c^3} r_{13}$ in SBN and KTP are calculated and plotted in Figs. 1 and 2 respectively. The parameter values used in our study are reported in Table 1. In SBN (Fig. 1), both r_c^T and r_c^S values are well described within our model, in the whole wavelength range, even in the vicinity of

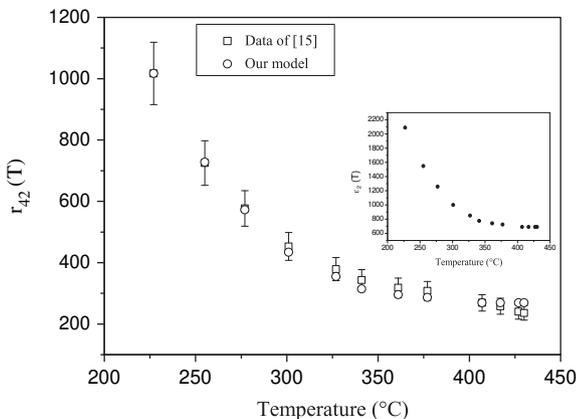


Fig. 3. Temperature dependence of the EO coefficient r_{42} in KNbO_3 crystal given by our model using the temperature dependence of the dielectric permittivity ϵ_2 from reference [16]. The parameter values used are: $a_T = 0.535 \text{ pm/V}$ and $b_T = -100.72 \text{ pm/V}$.

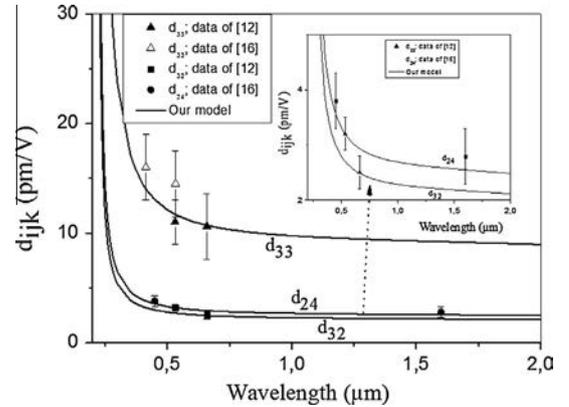


Fig. 4. Calculated dispersion of the second-order nonlinear optical coefficients d_{33} , d_{32} and d_{24} of KTP and the measured values reported in literature according to references [13,17].

the absorption edge. The agreement between experimental data and our calculations is achieved as well in KTP (Fig. 2). In this last case the clamped and unclamped coefficients do not differ notably.

In addition to the dispersion on the modulation frequency, Eq. (9) can be exploited for describing the temperature dependence of the EO coefficient. Since χ^{EO} and thus r are linked via Eq. (9) to the linear electric susceptibility χ , at same frequencies ω and Ω , any change of the value of this susceptibility, due to the temperature by example, will be reflected by a corresponding similar variation of the EO coefficient. As a consequence we get the temperature dependence of the EO coefficient as

$$r_{ijk}^{T(S)}(T) = a_T + b_T \epsilon_k(T) \quad (16)$$

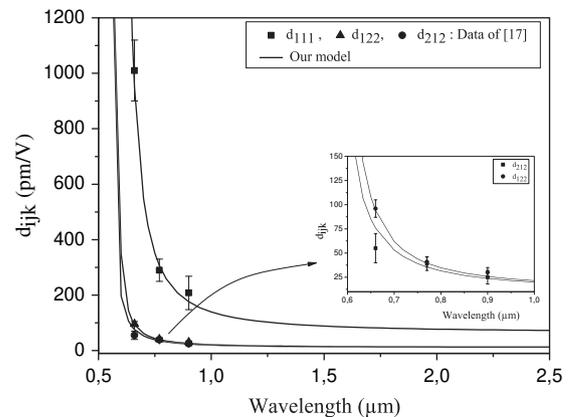


Fig. 5. Calculated dispersion of the second-order nonlinear optical coefficients d_{111} , d_{122} and d_{212} of DAST compared with the measured values reported in Ref. [18].

Table 2

Values of the parameters used in our study. α , β , γ , and λ_{0i} are the Sellmeier coefficients given in Refs. [19] for KTP crystal and [20] for DAST crystal. The term A_{ijk} is the parameter value of the model used to fit the wavelength dispersion of the second-order nonlinear optical coefficient d_{ijk} .

Crystal	Polarization direction	α	β (μm^2)	γ (μm^{-2})	λ_{0i} (μm)	d_{ijk} (pm V^{-1})	A_{ijk} ($\text{mV}^{-1} \text{s}^{-2}$)
KTP	X	2.9971	0.0410	0.0125	0.1958	–	–
	Y	3.0197	0.0440	0.0120	0.2050	d_{223} (d_{24})	4.3×10^{19}
	Z	3.3055	0.0632	0.0139	0.2116	d_{322} (d_{32}) d_{333} (d_{33})	4.4×10^{19} 16×10^{19}
DAST	X	4.3180	0.046732	0	0.533	d_{111}	7.5×10^{19}
	Y	2.5122	0.11913	0	0.504	d_{122}	6.5×10^{19}
	Z	2.4492	0.05873	0	0.501	d_{212}	3.12×10^{19}

where ϵ is the dielectric permittivity. The close link between r and ϵ can be especially pointed out by the respective temperature dependencies of these quantities, on approaching the phase transitions. In Fig. 3 is plotted the EO coefficient r_{42} of the tetragonal KNbO_3 versus the temperature, on approaching the tetragonal – orthorhombic transition (488 K). Calculated values derived via Eq. (16) from the dependence of ϵ (reported in the insert) are found in the good agreement with the experimental data in the whole temperature range.

3.2. Calculations of the SHG coefficients

The wavelength dependence of SHG coefficients is deduced from Eq. (3) as

$$d_{ijk} = \frac{A_{ijk}}{4\pi^2 c^2} \frac{(n_j^2(\lambda) - 1)(n_k^2(\lambda) - 1)}{\lambda^2 - 4\lambda_{0i}^2} \lambda_{0i}^2 \lambda^2. \quad (17)$$

It is demonstrated that the wavelength dependence of the SHG coefficients depend on the dispersion of the linear indices only and A_{ijk} is the solely needed parameter which is determined at a particular wavelength. This dependence is calculated and plotted in two well known nonlinear optical (NLO) materials: inorganic KTP, and organic DAST. Results are reported in Figs. 4 and 5 respectively. A fairly good agreement is obtained between calculated and experimental data for both materials, and for the different considered coefficients. In KTP, d_{33} (d_{333}) and d_{24} (d_{223}) exhibit a different behavior as function of λ , reflecting respectively the difference in the dispersion of n_1 and n_3 , and n_2 and n_3 respectively. Moreover the slight difference between the coefficients d_{24} and d_{32} shows the limits of the Kleinmann rule which assumes the equality of these coefficients.

In DAST, d_{111} is much larger than d_{122} and d_{212} , which are different only on approaching the resonance frequency.

In Table 2 are reported the parameter values used to obtain the wavelength dispersion of the different NLO coefficients presented in Figs. 4 and 5.

4. Conclusion

The expressions of the dispersion of the EO and SHG coefficients have been derived from a simple model based upon the anharmonic electronic potential. It was underlined that our formulation can be applied to each component of the second-order susceptibility without any assumption on the dependence of the refractive indices. In the earlier studies it was indeed assumed that (i) the dispersion of the refractive indices is not very large and therefore independent of the axis and (ii) Kleinman rules hold so that permutation relation on the subscripts can be used.

We show that the dispersion of the second order susceptibility depends on the frequency of the linear susceptibility only. Our calculations describe the behavior of SHG and EO coefficients, with only one adjustable parameter. In other terms this means that

the complete λ -dependence can be derived from the value of the coefficient measured at one wavelength only, and the available dispersion of the appropriate refractive index. A good agreement was achieved between our calculations and experimental data in several NLO materials. It was pointed out that the frequency dispersion of unclamped, clamped and electronic EO coefficients can be satisfactorily described by the model. This reinforces the validity and interest of our approach.

Acknowledgment

We would like to thank Prof. Germano Montemezzani for useful discussions.

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