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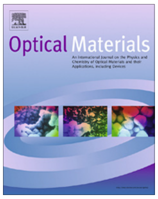

Calculation of the dispersion of the electro-optical and second harmonic coefficients from the refractive index dispersion

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ABSTRACT

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 $KNbO_3$ 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 $KTiOPO_4$ (KTP) and $LiNbO_3$ (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.

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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

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

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

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

or

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

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

$$d_{ijk}(\omega) = \frac{1}{2}\chi^{SHG}_{ijk}(2\omega,\omega,\omega) \tag{4}$$

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

$$\chi^{SHG}_{ijk}(2\omega,\omega,\omega) = \Delta_{ijk}(2\omega,\omega,\omega)\chi_i(2\omega)\chi_j(\omega)\chi_k(\omega) \tag{5}$$

It is observed in Eq. (3) that the index i plays a different role from indices j and k. As $\left(\omega_0^2-(2\omega)_i^2\right)^{-1}$ 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

$$\chi^{EO}_{ijk}(\omega,\Omega) = \frac{\chi_j(\omega)}{(\omega_0^2-\omega^2)_i}\left(B\chi^E_k + C\chi^I_k(\Omega)\right) \tag{6}$$

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

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

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

$$\chi^{EO}_{ijk}(\omega) = \frac{n_j^2(\omega)-1}{(\omega_0^2-\omega^2)_i}a^{EO}_{\Omega.ijk} \tag{8}$$

where $a^{EO}_{\Omega.ijk}$ 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)