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ON THE NATURE OF TEMPERATURE DEPENDENCE OF INTRINSIC QUADRATIC ELECTROOPTIC COEFFICIENTS IN KDP-TYPE CRYSTALS

It is shown that the fourth-order electrooptic effect may be responsible for the temperature dependence of the quadratic electrooptic coefficients defined in terms of polarization. An example is given for the ammonium dihydrogen phosphate (ADP) crystal.

Keywords: quadratic electrooptic effect, fourth-order electrooptic effect, ammonium dihydrogen phosphate.

1. INTRODUCTION

Crystals having a structure isomorphous with KH_2PO_4 (KDP) are known for their marked nonlinear properties. Experimental results have been published for a variety of nonlinear phenomena exhibited by the compounds. Among them several results comprising nonlinear electrooptic phenomena have been reported (see, e.g. [1-6]). However, in respect of the nonlinear electrooptic effects a large spread is observed in the relevant coefficients of a given crystal. The first published results for nonlinear electrooptic effects in KDP type crystals have been obtained employing static fields.In some measurements, for a strong field, deviations from the linear relationship between the induced birefringence and the square of the electric field have been reported. These nonlinearities have been explained in terms of the fourth-order effect. The results obtained at room temperature employing static fields for the effect, indicate very large values of the fourth-order coefficient $|K_{111111} - K_{221111}|$ of the order of magnitude 10^{-29}-10^{-33} m^4V^{-4} [1-3]. However, these data have not been confirmed by measurements performed employing more precise dynamic methods [4]. Further theoretical findings have shown that the errors in measurements of electrooptic coefficients are noticeably greater when a static electric field is used than in the case of an alternating field [5,7].

The knowledge of the magnitude of the fourth-order effect in crystals is important. For example, the effect may be partially responsible for the spontaneous birefringence in the low-temperature phase of ferroelectrics and antiferroelectrics [6,8,9]. In applications, nonlinear electrooptic effects may lead to nonlinear distortions in devices employing electrooptic crystals.

The aim of this work is to analyze if the fourth-order electrooptic effects may be partially responsible for the temperature dependence of the quadratic electrooptic coefficients. An example of ADP is considered.

2. TEMPERATURE CHANGES IN INTRINSIC QUADRATIC ELECTROOPTIC COEFFICIENTS

Pockels [10] introduced electrooptic coefficients defined as terms in a power-series expansion in the low-frequency electric field ***E*** of the optical frequency impermeability tensor $\eta_{ij}(\omega)$. The tensor $K_{ijk_1 \ldots k_f}$ that describes the electrooptic effect of the order f is given by

$$K_{ijk_1 \ldots k_f} = \frac{1}{f!}\left(\frac{\partial^f \eta_{ij}(\omega)}{\partial E_{k_1} \ldots E_{k_f}}\right) \tag{1}$$

It was also Pockels who first noticed that electrooptic and dielectric properties are closely connected and electrooptic coefficients follow the dielectric properties of crystals. He introduced the so-called intrinsic electrooptic coefficients $M_{ijk_1 \ldots k_f}$ defined in terms of the induced polarization ***P***

$$M_{ijk_1 \ldots k_f} = \frac{1}{f!}\left(\frac{\partial^f \eta_{ij}(\omega)}{\partial P_{k_1} \ldots P_{k_f}}\right) \tag{2}$$

The intrinsic coefficients are widely assumed to be roughly constant and what is more they are nearly equal both in the paraelectric and polarized phases [10-13]. Despite the fact that the assumption of constancy of the intrinsic coefficients is only an approximation, it should be stressed that, contrary to the coefficients defined in terms of the electric field, their changes with temperature are weak [9].

In this work we focus our attention on a possibility that higher-order electrooptic effects contribute to the temperature dependence of the quadratic intrinsic coefficients. According to the definition of electrooptic coefficients, in centrosymmetric media or in those configuration in noncetrosymmetric ones when the linear electrooptic effect is forbidden by crystal symmetry, the impermeability tensor may be expressed as

$$\eta_{ii}(\omega)=\frac{\delta_{ij}}{n^2}+M_{iikl}P_kP_l+M_{iiklmn}P_kP_lP_mP_n\ , \tag{3}$$

where δ_{ij} is the Kronecker delta, n is the refractive index of unperturbed crystal, and M_{ijkl} and M_{ijklmn} stand for the quadratic and fourth-order intrinsic electrooptic effect, respectively.

The matrices for quadratic electrooptic tenors are widely known (see, e.g. [14]). The matrices for the fourth-order electrooptic effect are given in Ref. [15]. Here, as an example, we consider the crystal of symmetry 42 m and the experimental configuration with the vector of induced polarization given by $\boldsymbol{P}$ = [P,0,0] and the light beam propagating along the crystal optical axis. It follows from Eq. (3) that the new refractive indices n_i depend on the induced polarization as

$$n_a=\left(n_o^{-2}+M_{1111}P^2+M_{111111}P^4\right)^{-1/2} \tag{4}$$

and

$$n_b=\left(n_o^{-2}+M_{2211}P^2+M_{221111}P^4\right)^{-1/2}, \tag{5}$$

where n_o is the ordinary refractive index. Expanding expressions (4) and (5) in power series to the fourth power in P gives

$$n_a=n_o-\frac{n_o^3}{2}\left[M_{1111}P^2+M_{111111}P^4-\frac{3}{4}n_o^2\left(M_{1111}\right)^2P^4\right] \tag{6}$$

and

$$n_b=n_o-\frac{n_o^3}{2}\left[M_{2211}P^2+M_{221111}P^4-\frac{3}{4}n_o^2\left(M_{2211}\right)^2P^4\right]. \tag{7}$$

Thus, the induced birefringence is

$$\Delta n = n_a - n_b = -\frac{n_o^3}{2}\Big[(M_{1111} - M_{2211})P^2 + (M_{111111} - M_{221111})P^4 + \\ -\frac{3}{4}\mathrm{n}_\mathrm{o}^2\left((\mathrm{M}_{1111})^2 - (\mathrm{M}_{2211})^2\right)\mathrm{P}^4\Big]. \tag{8}$$

Equation (8) predicts the following changes in the intrinsic quadratic electrooptic coefficient $M_{1111} - M_{2211}$ due to the changes in the induced polarization amplitude from the magnitude P^{ampl}(A) to P^{ampl}(B)

$$\Delta M^{quadr} = \Delta\left| \; M_{1111} - M_{2211} \right|_{P^{ampl}(B)}^{P^{ampl}(A)} = \\ = \frac{1}{2}\left[(M_{111111} - M_{221111}) - \frac{3}{4}n_o^2\left((M_{1111})^2 - (M_{2211})^2\right)\right]\left(P^{ampl}(A)^2 - P^{ampl}(B)^2\right) \tag{9}$$

The factor of ½ before the square bracket in Eq. (9) appears because an alternating modulating field is assumed to be employed and the changes in $\Delta M^{\mathrm{quadr}}$ are proportional to the square of the field. The amplitude of the induced polarization P^{ampl} is simply related to the amplitude of the modulating electric field E^{ampl}

$$P^{ampl} = \varepsilon_0(\varepsilon - 1)E^{ampl}, \tag{10}$$

where, ε_0 is the permittivity of free space and ε is the low-frequency dielectric constant. Thus, even when in measurements the amplitude of the modulating electric field is constant, the changes in temperature affecting the dielectric constant lead to changes in the polarization induced in the crystal.

The temperature dependence of ε_{11} in ADP has been reported in Ref. [16]. These data may be fitted using a Curie-Weiss-type formula [19]

$$\varepsilon_{11} = \varepsilon_{11}^{\infty} + \frac{C_1}{T - T_1}, \tag{11}$$

with $\varepsilon_{11}^{\infty}$= 19.8, C_1 =10160 K and T_1 =22.7 K. In ADP the quadratic electrooptic coefficients $g_{1111} = -7.4 \cdot 10^{-20}$ m^{2}V^{-2} and $g_{2211} = -1.6 \cdot 10^{-20}$ m^{2}V^{-2} have been measured at room temperature [17]. Taking the ordinary refractive index n_0 = 1.522 one obtains that for ADP the second term in the square bracket in Eq. (9) which is of the order of magnitude $6 \cdot 10^{-39}$ m^{4}V^{-4} (what gives at room temperature 0.1 m^8C^{-4}) may be neglected [18]. Thus in Eq. (9) the first term, related to the fourth-order effect may be important.

The coefficient $|K_{111111} - K_{221111}|$ measured in ADP by dynamic means has been found to be not higher than $3{\cdot}10^{-33}$ m^4V^{-4} [4] what gives for room temperature $|M_{111111} - M_{221111}| < 5{\cdot}10^4$ m^8C^{-4}. Taking as example $|M_{111111} - M_{221111}| = 3{\cdot}10^4$ m^8C^{-4} and assuming the amplitudes of the modulating electric field $6{\cdot}10^5$ Vm^{-1} and $2{\cdot}10^5$ Vm^{-1} one obtains that between 290 K and 340 K the changes $\Delta M^{\mathrm{quadr}}$ in ADP should be about $2{\cdot}10^{-4}$ m^4C^{-2}. In our opinion such a difference is measurable.

3. CONCLUSIONS

The temperature dependence of the intrinsic quadratic electrooptic coefficients may partially result from a contribution of the fourth-order effect. The contribution may be determined by measurements of the dependence performed employing alternating field of different strengths in a crystal for which the temperature behaviour of the low-frequency dielectric constant is known.

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ANALIZA MECHANIZMU ZALEŻNOŚCI TEMPERATUROWEJ WSPÓŁCZYNNIKÓW WŁAŚCIWEGO KWADRATOWEGO EFEKTU ELEKTROOPTYCZNEGO W KRYSZTAŁACH GRUPY KDP

Streszczenie

Pokazano, że efekt elektrooptyczny czwartego rzędu może być odpowiedzialny za zmiany temperaturowe współczynników właściwego kwadratowego efektu elektrooptycznego. Rozważono przypadek kryształu ADP.