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## ON THE CONTRIBUTION OF THE QUADRATIC ELECTROOPTIC EFFECT TO THE NONLINEAR RESPONSE PROPORTIONAL TO THE FOURTH POWER OF MODULATING FIELD

*It is shown that the fourth-order electrooptic effect cannot be measured without a background of the contribution that is proportional to the square of the quadratic electrooptic coefficient. The contribution manifests on the fourth power of the modulating field and may be calculated. Thus, if the contribution is found to be comparable with the measured fourth-order electrooptic response, this apparent fourth-order effect should be taken into account in the interpretation of the results of the measurement.*

**Keywords:** quadratic electrooptic effect, fourth-order electrooptic effect.

### 1. INTRODUCTION

The results of measurements of the fourth-order electrooptic coefficients have been previously reported for some crystals including BaTiO<sub>3</sub> [1] and members of the KH<sub>2</sub>PO<sub>4</sub> (KDP) family crystals (see, e.g. [2-4]). The first published results for the fourth-order effect in KDP type crystals have been obtained employing static fields and indicated very large values of the effective coefficient  $|K_{111111} - K_{221111}|$  of the order of magnitude  $10^{-30} \text{ m}^4\text{V}^{-4}$  [2-4]. However, these data have not been confirmed by further attempts to measure the fourth-order electrooptic effect [5,6]. For example, the coefficient  $|K_{111111} - K_{221111}|$  measured in NH<sub>4</sub>H<sub>2</sub>PO<sub>4</sub> (ADP) by dynamic means has been found to be not

higher than  $10^{-33} \text{ m}^4\text{V}^{-4}$  [6]. One notes that somewhat similar situation was observed for the quadratic electrooptic effect. It has been found that apparent quadratic coefficients obtained by static methods and due in fact to imperfectly cut or aligned investigated crystal samples, are sometimes, three orders of magnitude larger than those, measured employing dynamic methods [7,8]. The theoretical results presented recently for the quadratic electrooptic effect have shown that the sensitivity of measurements performed by static techniques for the inaccuracies is much higher than in methods employing a sinusoidal electric modulating field. Corresponding nonlinearities may be interpreted in terms of apparent quadratic electrooptic coefficients [9-12].

The fourth-order effect is important, e.g. from the point of view of the relationship between the spontaneous birefringence and spontaneous antipolarization of antiferroelectrics, for example, in the low-temperature phase of ADP [13]. Nonlinear electrooptic effects may also lead to nonlinear responses of various technical devices employing electrooptic crystals, therefore, are significant in applications.

The aim of this work is to show that when in measurements the fourth-order electrooptic response appears, this does not necessarily mean that the true fourth-order electrooptic effect is responsible for the nonlinearity.

## 2. RESULTS

Traditionally, the quadratic and the fourth-order electrooptic tensors, respectively, are defined by

$$g_{ijkl} = \frac{1}{2} \left( \frac{\partial^2 \eta_{ij}(\omega)}{\partial E_k \partial E_l} \right), \quad (1)$$

and

$$K_{ijklmn} = \frac{1}{24} \left( \frac{\partial^4 \eta_{ij}(\omega)}{\partial E_k \partial E_l \partial E_m \partial E_n} \right), \quad (2)$$

where  $\eta_{ij}$  is the impermeability tensor. Thus in a principal axes system, the electric-field-induced changes in the impermeability tensor are given by

$$\eta_{ii}(E) = \frac{1}{n_i^2(0)} + g_{iikl} E_k E_l + K_{iiklmn} E_k E_l E_m E_n, \quad (3)$$

where  $n_i(0)$  is the refractive index of unperturbed crystal. The matrices of the fourth-order electrooptic effect have been obtained previously [14]. However,

in this work we will neglect its contribution and take into consideration only the quadratic electrooptic one. Therefore, it follows from Eq. (3) that the new refractive indices  $n_i$  observed in the crystal subjected to a modulating electric field depend on the field as

$$n_i(E) = \left( n_i^{-2}(0) + g_{iikl} E_k E_l \right)^{-1/2}, \quad (4)$$

or

$$n_i(E) = n_i(0) \left( 1 + n_i^2(0) g_{iikl} E_k E_l \right)^{-1/2}. \quad (5)$$

After expanding  $n_i(E)$  in the power series of the type  $(1+x)^{-1/2}$  up to the term proportional to the fourth power of  $\mathbf{E}$  the changes in the refractive index may be expressed as

$$n_i(E) \approx n_i(0) - \frac{n_i^3(0)}{2} \left[ g_{iikl} E_k E_l - \frac{3}{4} n_i^2(0) g_{iikl}^2 E_k^2 E_l^2 \right]. \quad (6)$$

Equation (6) shows that the term proportional to the square of  $g_{iikl}$  behaves as the fourth-order electrooptic coefficient. As an example we consider ADP, for which values of some quadratic electrooptic coefficients have been measured at room temperature [7,8]. Among them,  $g_{1111} = -7.4 \cdot 10^{-20} \text{ m}^2 \text{V}^{-2}$  and  $g_{2211} = -1.6 \cdot 10^{-20} \text{ m}^2 \text{V}^{-2}$  are known. Taking the value of the ordinary refractive index, i.e.  $n_1(0) = 1.5222$  [15] one obtains the fourth-order electrooptic modulation suggesting the magnitude of the effective fourth-order electrooptic coefficient to be about  $|K_{111111} - K_{221111}| = 6 \cdot 10^{-39} \text{ m}^4 \text{V}^{-4}$ .

### 3. CONCLUSIONS

The findings that the square of the quadratic electrooptic coefficients always contributes to the forth-order response may be useful in investigations of the fourth-order effect. Certainly the contribution cannot explain previously reported values of the order of magnitude  $10^{-30} \text{ m}^4 \text{V}^{-4} - 10^{-33} \text{ m}^4 \text{V}^{-4}$  [2-4,6]. However, the orders of the fourth-order coefficients are still uncertain and may be highly overestimated. Therefore, if in a measurement a response corresponding to the value of the fourth-order effect comparable to the square of the quadratic one is obtained, the previously neglected terms due to the quadratic coefficients that may result in the apparent fourth-order effect should be taken into consideration.

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**POZORNY ELEKTROOPTYCZNY EFEKT CZWARTEGO  
RZĘDU MAJĄCY SWOJE ŹRÓDŁO  
W KWADRATOWYM EFEKCIE ELEKTROOPTYCZNYM**

**Streszczenie**

Pokazano, że prawdziwemu efektowi elektrooptycznemu czwartego rzędu zawsze towarzyszy efekt pozorny, mający swoje źródło w efekcie kwadratowym. Dotychczas zaniedbywany wkład pozornego efektu można obliczyć i jeśli jest on porównywalny z mierzoną odpowiedzią przypisywaną efektowi elektrooptycznemu czwartego rzędu, należy go uwzględnić przy interpretacji wyników pomiarów.