Nonlinear Refractive Index for Crystals at Terahertz Frequencies

(Invited Paper)

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***Abstract*—The lecture is devoted to the analysis of the nonlinearity of the refractive index for crystals at terahertz frequencies. We develop a simple analytical model for calculating the vibrational contribution to the nonlinear refractive index n_2 of a crystal in terms of known crystalline parameters such as the linear refractive index, the coefficient of thermal expansion, atomic density, and the reduced mass and the natural oscillation frequency of the vibrational modes of the crystal lattice. Significant part of the lecture is devoted to the theoretical methods of analysis and features of self-action of few-cycle terahertz waves in various nonlinear media.**

I. Introduction

RESEARCH in the field of terahertz (THz) radiation has celebrated the beginning of its second decade. Since their first demonstrations, THz techniques continue to find new applications from medical diagnostics and therapy to the detection of hidden substances, including explosives and drugs [1]. In recent years, pulsed sources of the THz radiation with sufficient intensity for the observation of nonlinear optical effects have appeared in a number of laboratories [2], [3]. Investigation of the nonlinear optical effects in the THz spectral range has now become feasible.

In this lecture, we discuss a method for calculating the coefficient n_2 of crystals in the THz spectral range. It is shown that the vibrational contribution to the nonlinear response in the far infrared spectral range can be several orders of magnitude larger than the electronic nonlinearity, which is the dominant contribution for ultrashort pulses in the visible and near-infrared spectral ranges [4], [5].

We analize the benefits of different theoretical methods for the investigation of intensive few-cycle THz wave propagation in an optical medium and demonstrate the features of self-action of such waves with a small number of optical field oscillations. It is shown that the decrease in the number of field oscillations in pulse leads to a change of the dominant effects in the radiation self-action phenomenon. For instance, the effect of high frequency generation in nonlinear isotropic media dominates over the self-focusing effect for initially single-cycle pulse.

II. Estimation of Nonlinear Refractive Index for Crystals at THz Frequencies

There are different mechanisms contributing to the nonlinear refractive properties of an optical material. In the visible and near-infrared spectral ranges, the dominant contribution to the nonlinear refractive index is of electronic nature [4], [5]. In the far-infrared range, one expects the dominant mechanism of the nonlinearity to be associated with anharmonic vibrations of the crystalline lattice.

Let us analyze the vibrational nonlinearity of a crystalline material by considering the dynamics of ions in the lattice resulting from the force induced by the electromagnetic field. In this analysis, we make use of a classical model of the anharmonic oscillator:

$$\ddot{x} + 2\gamma\dot{x} + \omega_0^2 x + ax^2 + bx^3 = \alpha E. \quad (1)$$

Here x is the deviation of an ion from its equilibrium position, E is the applied electric field, ω_0 is the central frequency, γ is the damping coefficient, a and b are the nonlinear coefficients, $\alpha = q/m$, where q is the ionic change, and m is the reduced mass of the vibrational mode.

Within this study, we assume that the electric field interacting with the ions is monochromatic with the frequency ω $E(\omega) = E_\omega \mathrm{e}^{-i\omega t} + \mathrm{c.c.}$ We treat the nonlinear optical effects that occur without change in the frequency spectrum. We thus consider the oscillations of the crystalline ions at the fundamental frequency only: $x = x_\omega \mathrm{e}^{-i\omega t} + \mathrm{c.c.}$ Making use of a classical anharmonic oscillator model (1), we find the expression for the deviation of an ion from its equilibrium position x_ω, and then relate this quantity to the part of the amplitude of the polarization component, oscillating at the frequency ω and including both linear and nonlinear contributions $P_\omega = \chi^{(1)} E_\omega + 3\chi^{(3)}|E_\omega|^2 E_\omega$ to obtain the nonlinear susceptibility $\chi^{(3)}$, and hence to obtain the expression

$$n_2 = \frac{\pi q N}{n_0} \frac{\alpha^3}{(\omega_0^2 - \omega^2 - 2\gamma i\omega)^4} \times \left[2a^2 \frac{3\omega_0^2 - 8\omega^2 - 8\gamma i\omega}{\omega_0^2(\omega_0^2 - 4\omega^2 - 4\gamma i\omega)} + 3b\right] \quad (2)$$

for the nonlinear refractive index $n_2 = 3\pi\chi^{(3)}/n_0$.

However, it is not always straightforward to make use of the Eq. (2), because the values of the parameters appearing in this equation are not well known. We therefore use the model of Eq. (1) to calculate the values of other properties of the crystal that are known or easily measurable: linear refractive index and coefficient of thermal expansion. For instance, we find the relationship between parameters of Eq. (1) and the thermal expansion coefficient of the crystal:

$$\alpha_{\rm T} = -\frac{a k_{\rm B}}{m\omega_0^4 a_{\rm l}}, \tag{3}$$

where $k_{\rm B}$ is the Boltzmann constant and $a_{\rm l}$ is the lattice constant. Using these results, we derived the formula for vibrational contribution to n_2 at the terahertz frequency range (under the approximation $\omega \ll \omega_0$) in the form

$$\begin{aligned} \bar{n}_{2,\,{\rm v}}^{\omega\ll\omega_0} = \frac{3a_{\rm l}^2 m^2\omega_0^4\alpha_{\rm T}^2}{32 n_0\pi^2 q^2 N^2 k_{\rm B}^2}\left[\left(n_{0,\,{\rm v}}^{\omega\ll\omega_0}\right)^2 - 1\right]^3 \\ - \frac{9}{32}\frac{1}{\pi N}\frac{1}{n_0\hbar\omega_0}\left[\left(n_{0,\,{\rm v}}^{(\omega\ll\omega_0)}\right)^2 - 1\right]^2. \end{aligned} \tag{4}$$

Then we found that its value for the crystal quartz $n_2 = 2.2\times10^{-9}$ esu $= 4.4\times10^{-16}$ m^2/W is four orders of magnitude larger than its value in the visible range 3×10^{-20} m^2/W. Used for above estimation of n_2 values of the thermal expansion coefficient in the case of crystalline quartz are $\alpha_{\rm T} = 7.6\times10^{-6}\,(^\circ\,{\rm C})^{-1}$ parallel to the optic axis, and $\alpha_{\rm T} = 14\times10^{-6}\,(^\circ\,{\rm C})^{-1}$ perpendicular to the optic axis. We assumed that the vibrational mode is the Si-O stretch mode. The value for the fundamental vibrational frequency is 1242 cm^{-1} or 37.2 THz or 2.34×10^{14} rad/sec. The lattice constant of crystalline quartz is 4.91 $\AA$ along the c-axis, and is 5.40 $\AA$ along the a and b axes. We can define a mean lattice constant with the value 5.24 $\AA$ or 5.24×10^{-8} cm. The reduced mass m of the stretch mode was calculated as follows. The mass of the silicon atom is $m_{\rm Si} = 28.1$ amu, and that of the oxygen atom is $m_{\rm O} = 16$ amu. The reduced mass is thus $m_{\rm Si}m_{\rm O}/(m_{\rm Si}+m_{\rm O}) = 10.2$ amu or 1.69×10^{-23} g. The number density N of the vibrational units was calculated as follows. The specific gravity of crystal quartz is 2.65, and the formula weight of SiO_2 is $28+16\times2 = 60$. Each silicon atom is thus associated with the total mass of $60\times1.67\times10^{-24}$ g $= 1.00\times10^{-22}$ g. The number of silicon atoms in 1 cm^3 of quartz is thus $2.65/(1.00\times10^{-22}) = 2.65\times10^{22}$.

Our model also predicts a large variation of n_2 with frequency in the vicinity of the vibrational resonance, and the existence of the two-photon resonance in the spectrum of n_2 (see the figure below). It is evident from Fig. 1 that the resonant value of n_2 exhibits more than one order of magnitude of enhancement compared to its low-frequency value.

As evident from Fig. 1, for the waves with the spectrum spanning the frequency range between 0 and 12 THz, the nonlinear refractive index doubles in value. One cannot neglect the dispersion of n_2 when analyzing the interaction of a broad-spectrum radiation with an optical medium. In the frequency range beyond 12 THz the dispersion is significant; the optical nonlinearity exhibits inertia.

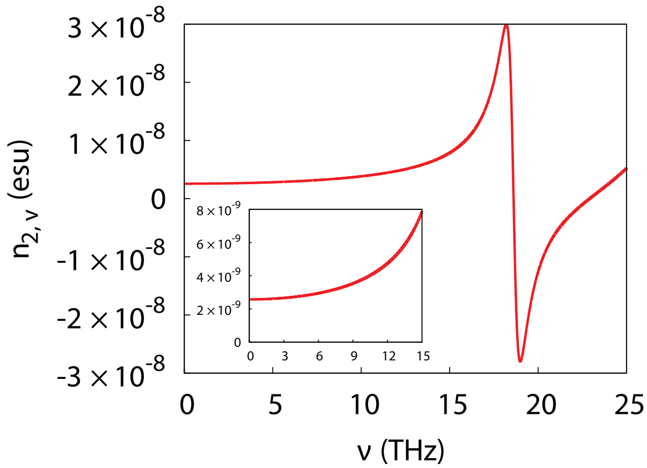

Fig. 1. Dispersion of the nonlinear refractive index in the vicinity of the two-photon resonance.

In the lecture we also discuss the applicability of the presented method of calculation of n_2 for the case of isotropic media.

III. Features of Self-action of Few-cycle THz Pulses

Main part of the sourses of intensive THz radiation generates few or even just one cycle pulses. So we then analyse the features of propagation of initially single-cycle optical pulses in isotropic dielectric media with instant cubic (Kerr) nonlinearity. Because it is evident from the inset in Fig. 1 that one can neglect the dispersion of a wide-spectrum pulse, as long as the entire pulse spectrum width lies in the range between 0 and 6 THz.

We consider unidirectional paraxial propagation corresponding to a beam width much larger than the optical wavelength, and assume that the wavelength spectrum is within the region of normal group-velocity-dispersion with parameters of crystal quartz. Under such conditions, the evolution of the electric field $E(z,x,y,t)$ of an optical wave can be modelled by the following equation [6], [7]:

$$\frac{\partial E}{\partial z} + \frac{N_0}{c}\frac{\partial E}{\partial t} - a_{disp}\frac{\partial^3 E}{\partial t^3} + gE^2\frac{\partial E}{\partial t} = \frac{c}{2N_0}\Delta_\perp\int_{-\infty}^{t} E dt', \tag{5}$$

where z is the distance along the propagation direction, $\Delta_\perp = \partial^2/\partial x^2 + \partial^2/\partial y^2$ is the transverse Laplace operator, t is time, c is the speed of light in vacuum. Parameters N_0 and a_{disp} define the medium dispersion as $n_0(\omega) = N_0 + a_{disp}c\omega^2$, where n_0 is the linear refractive index and ω is the optical frequency. The coefficient g characterizing the Kerr-type nonlinear optical response is related to the Kerr coefficient n_2 according to $g = 2n_2/c$. We underline that Eq. (5) is formulated for the electric field E of the optical wave, and it is suitable for theoretical modeling of ultra-short pulse evolution with very broad spectrum, including the case of pulses with single-cycle field oscillation.

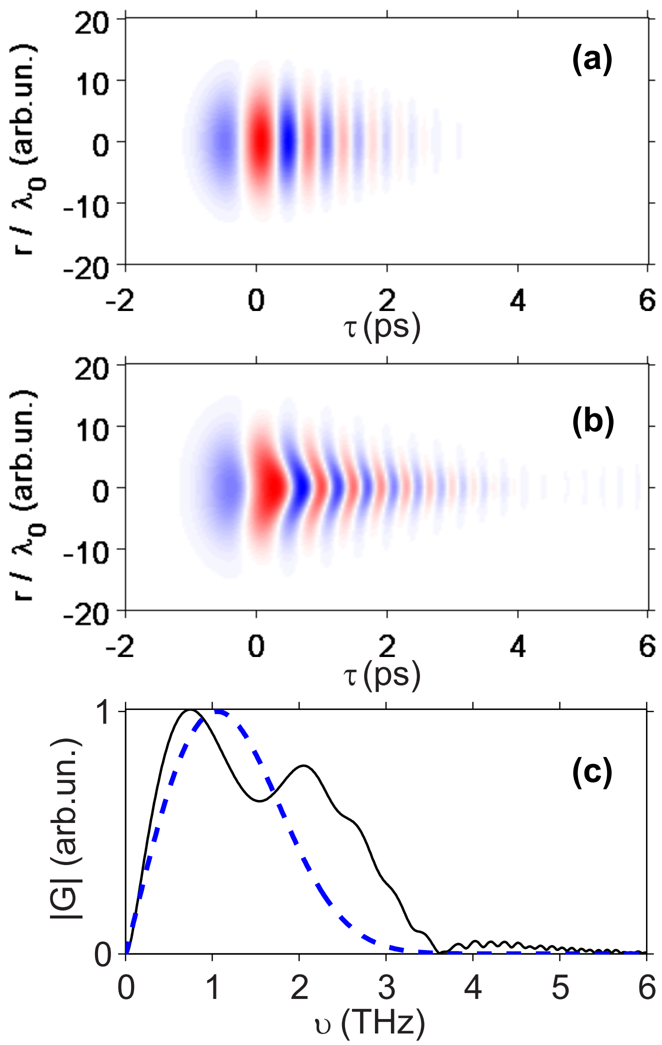

Fig. 2. Spatiotemporal electric output field profiles with initially Gaussian transverse distribution for (a) weak nonlinear ($I = 5.0 \times 10^8$ W/cm^2) and (b) strong nonlinear ($I = 5.0 \times 10^9$ W/cm^2) medium with the same nonlinearity as crystal quartz $n_2 = 4.4 \times 10^{-12}$ cm^2/W and (c) corresponding to strong nonlinear case modulus of the output spectrum at the beam axis. The spectrum of input single-cycle wave at the beam axis is shown by a dashed line. The length of medium is 10 mm. $r = \sqrt{x^2 + y^2}$, $\tau = t - (c/N_0)z$ is the retarded time.

In Fig. 2(a,b), we show two-dimensional contour plots of the axisymmetric electric field of a single-cycle wave propagation in (a) weak nonlinear medium at the intensity level $I = 5.0 \times 10^8$ W/cm^2 and (b) strong nonlinear dielectric medium at the intensity level $I = 5.0 \times 10^9$ W/cm^2 with the length 10 mm and the same nonlinearity as crystal quartz $n_2 = 4.4 \times 10^{-12}$ cm^2/W. Red and blue areas correspond to maximum positive and negative values, respectively. The transverse beam width at the input of the medium is $10\lambda_0$, $\lambda_0 = 300$ μm. In Fig. 2(c), we show the input and output spectra with the dashed and solid lines, respectively. It is evident from the graph that, for the higher input intensity, the combined action of the nonlinearity and dispersion lead to the formation of the high-frequency tail that spans up to five times the central frequency.

A part of the lecture will be devoted to the complex dynamics of the electric field of a single-cycle pulse with the spectrum at the frequency range of the two-photon resonance.

IV. Conclusion

We have deduced the relationship between the vibrational contribution to n_2 and other readily measurable parameters of a crystal. Using our model, we have performed an estimate of the value of n_2 for crystalline quartz in the THz spectral range and found that in the low-frequency limit it is four orders of magnitude larger than the value of n_2 in the visible range. Our model also predicts a large variation of n_2 with frequency. In the calculations of n_2, we have assumed that the THz radiation is quasimonochomatic. Main part THz experiments are conducted with intensive very short pulses, which may contain only a few (or even just one) optical cycles. We demonstrate that the nature of the nonlinear phenomena under these conditions can differ drastically from the case of quasimonochromatic radiation.

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