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RESEARCH ARTICLE

Dispersion of optical constants of Si:PbGeO crystal in the terahertz range

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Abstract

Objectives. Advances in laser physics over the last decade have led to the creation of sources of single-period electromagnetic pulses having a duration of about 1 ps, corresponding to the terahertz (THz) frequency range and a field amplitude of several tens of MV/cm. This allows the electrode-free application of an electric field to a ferroelectric for observing not only the excitation of coherent phonons, but also ultrafast (at the sub-picosecond timescale) dynamic polarization switching. To detect polarization switching, a pump-probe technique is used in which a THz pulse is used with an optical probe. Since its intensity is proportional to the square of the polarization, the signal of the optical second harmonic is used to measure polarization switching under the action of a THz pulse. To evaluate switching efficiency, both linear (refractive index and absorption coefficient) and non-linear optical characteristics (quadratic and cubic susceptibilities) are required. For any application of ferroelectric crystals in the THz range, knowledge of the relevant linear optical characteristics is also necessary.

Methods. The technique of THz spectroscopy in the time domain was used; here, a picosecond THz pulse transmitted through the crystal is recorded by strobing the detector with a femtosecond optical pulse. The THz -induced dynamics of the order parameter in a ferroelectric was studied by detecting the intensity of a nonlinear optical signal at the frequency of the optical second harmonic.

Results. The transmission of a THz wave and the intensity of second harmonic generation on a lead germanate crystal doped with silicon in the time and spectral domains were measured. On this basis, the absorption coefficient dispersion and cubic nonlinear susceptibility were calculated in the range of 0.5–2.0 THz. The presence of a region of fundamental absorption near the phonon modes was confirmed along with a resonant enhancement of the cubic nonlinear susceptibility for two phonon modes Ω_1 = 1.3 THz and Ω_2 = 2.0 THz.

Conclusions. The proposed technique is effective for analyzing the dispersion of the optical characteristics of ferroelectric crystals. The significantly improved spectral resolution (0.1 THz) increases the accuracy of determining nonlinear susceptibility due to the detailed analysis of the linear and nonlinear contributions to the second harmonic intensity.

Keywords: terahertz radiation, ferroelectrics, spectroscopy, optical second harmonic generation

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НАУЧНАЯ СТАТЬЯ

Дисперсия оптических характеристик кристалла Si:PbGeO в терагерцовом диапазоне

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Резюме

Цели. Успехи лазерной физики последнего десятилетия привели к созданию источников однопериодных электромагнитных импульсов длительностью порядка 1 пс, что соответствует терагерцовому (ТГц) диапазону частот, с амплитудой поля в несколько десятков МВ/см. Это позволило приложить электрическое поле к сегнетоэлектрику без электродов и наблюдать не только возбуждение когерентных фононов, но и сверхбыстрое, в субпикосекундном масштабе времени, динамическое переключение поляризации. Для обнаружения переключения поляризации используется метод накачки-зондирования, где в качестве накачки используется ТГц-импульс, а зонд является оптическим. Мерой переключения поляризации под действием ТГц-импульса служит сигнал оптической второй гармоники, поскольку ее интенсивность пропорциональна квадрату поляризации. Для оценки эффективности переключения требуются как линейные (показатель преломления и коэффициент поглощения), так и нелинейные оптические характеристики (квадратичная и кубичная восприимчивости). Знание линейных оптических характеристик необходимо также для любых применений рассматриваемых кристаллов в ТГц-диапазоне.

Методы. Использована методика терагерцовой спектроскопии во временно́й области, в которой на вещество направляется пикосекундный ТГц-импульс, а регистрируется ТГц-импульс, прошедший через вещество, путем стробирования детектора фемтосекундным оптическим импульсом. Исследование ТГц-индуцированной динамики параметра порядка в сегнетоэлектрике проводилось путем детектирования интенсивности нелинейно-оптического сигнала на частоте второй оптической гармоники.

Результаты. На кристалле германата свинца, легированного кремнием, измерены пропускание ТГц-волны и интенсивность генерации второй гармоники во временно́й и спектральной областях, на основании чего рассчитаны дисперсия коэффициента поглощения и кубичной нелинейной восприимчивости в диапазоне 0.5–2.0 ТГц. Обнаружено наличие области фундаментального поглощения вблизи фононных мод, а также резонансное усиление кубичной нелинейной восприимчивости для двух фононных мод Ω_1 = 1.3 ТГц и Ω_2 = 2.0 ТГц.

Выводы. Предложенная методика эффективна для анализа дисперсии оптических характеристик сегнетоэлектрических кристаллов. Существенно улучшено спектральное разрешение, составляющее в данной работе 0.1 ТГц, а также точность определения нелинейной восприимчивости за счет детального анализа линейного и нелинейного вкладов в интенсивность второй гармоники.

Ключевые слова: терагерцовое излучение, сегнетоэлектрики, спектроскопия, генерация второй оптической гармоники

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INTRODUCTION

Lead germanate crystals ($Pb_5Ge_3O_{11}$, PGO) are uniaxial ferroelectrics having a Curie temperature T_C = 450 K [1]. Along with silicon-containing solid solutions based on them, these crystals exhibit features of spontaneous polarization switching as well as pyroelectric and photorefractive effects that have a number of potential applications. By changing the silicon concentration in the solid solution, the Curie point can be controlled in order to transfer the main features into the temperature range from room temperature to T_C, thus significantly expanding the potential areas of application of these crystals, including as elements of pyroelectric receivers and ferroelectric memory devices. While a number of works deal with the structure of these materials, as well as their dielectric, piezo- and pyroelectric, mechanical, and other properties, these have mainly been focused on the low-frequency region [2].

The frequency domain of 1–10 THz is of great interest for studying ferroelectrics, since it is in this domain that phonon modes, including soft phonon mode, occur. Until recently, the Raman spectroscopy technique has mainly been used to study incoherent processes in this domain [3, 4].

Advances in laser physics taking place over the last decade have led to the creation of sources of single-period electromagnetic pulses with a duration of about 1 ps, which corresponds to the terahertz (THz) frequency range and a field amplitude of several tens of MV/cm. By this means, an electric field can be applied to a ferroelectric without electrodes to observe not only the excitation of coherent phonons, but also ultrafast (at the sub-picosecond timescale) dynamic polarization switching. To detect polarization switching, a pump-probe method is used, in which a THz pulse is used as pump while the probe is either optical or X-ray. The theory of coherent oscillation generation in THz pump-probe spectroscopy is described in [5].

As well as being used to distinguish between linear and nonlinear modes of mode oscillations [6, 7], the optical probe is capable of detecting dynamic and permanent polarization switching if the latter occurs [8]. The measure of polarization switching under the action of a THz pulse serves as the optical second harmonic SHsignal, its intensity being proportional to the square of polarization. Both linear (refractive index and absorption coefficient) and nonlinear optical characteristics (quadratic and cubic susceptibilities) are required for evaluating switching efficiency. For any applications of the crystals under consideration in the THz range, it is necessary to obtain information concerning their linear optical characteristics.

THz time domain spectroscopy (THz-TDS), in which the passage of a picosecond THz pulse through the substance is registered by gating the detector with a femtosecond optical pulse, is applicable for determining both linear and nonlinear optical constants. Since the generation and detection schemes are sensitive to the effect of the sample on both the amplitude and the phase of the registered THz radiation, it can be used to obtain both real and imaginary parts of optical constants.

The THz-TDS technique is widely used to determine the dispersion of optical constants, especially in organic materials (see review [9]). While certain successes have also been achieved in the study of phonon modes of ferroelectric crystals [10, 11], the results obtained by the majority of studies are based on low-power THz sources with standard parameters providing so-called broadband spectroscopy, where the generated spectrum region when pumped by a picosecond pulse is 0.5–2 THz with a center in the 1 THz region. However, in many ferroelectric materials, the phonon spectrum—including the most interesting soft mode—lies in the region above 1.5 THz.

In this connection, PGO represents a truly unique material, its pronounced ferroelectric properties occurring at a relatively high phase transition temperature (about 450 K) with a soft-mode frequency close to 1 THz; thus, there is a fairly wide range of phonon modes falling within the operating range of desktop installations. Results of a previous study [12] into PGO crystal using broadband spectroscopy with spectral resolution of 0.25 THz include a determination of the spectral dependencies of the electrically-induced optical SH.

In the present paper, narrow-band THz spectroscopy is used for the first time to determine the absorption

coefficient and nonlinear (cubic) susceptibility of a silicon-doped PGO crystal. The accuracy of nonlinear susceptibility determination is substantially improved by carrying out a detailed analysis of linear and nonlinear contributions to the SH intensity obtained at a spectral resolution of 0.1 THz.

EXPERIMENT

The silicon-doped PGO crystal $Pb_5(Ge_{0.74}Si_{0.26})_3O_{11}$ (made and provided by A.A. Bush, MIREA – Russian Technological University, Russia) is used for experimental studies. At this stoichiometric composition, the Curie temperature decreases to 346 K, as compared to the undoped crystal [2]. The thickness of the crystal under study is ~1.1 mm. The crystallographic orientation of the surface is (100); at this orientation, the polarization vector is aligned in the surface plane. The phonon spectrum of the crystal under study allows exciting effectively several frequencies of crystal lattice vibrations lying in the range of exciting THz radiation including the soft phonon mode frequency [13].

The narrow-band THz pulses generated using the Cr:forsterite laser system described in [12] have a wavelength of 1240 nm, pulse repetition rate of 10 Hz, and duration of 100 fs. The organic crystal OH1 serves as the generator of THz pulses [14].

For generating narrow-band THz pulses, the amplified laser pulse is divided into two, each passing through one of the arms of the Mach–Zehnder type interferometer. By varying the delay between these pulses before their subsequent compression in the compressor, beating optical pulses can be generated at a given frequency. The resulting frequency-modulated optical chirp irradiates the OH1 crystal to generate narrowband THz radiation [15]. The broadband pulse energy of 90 μJ is sufficient to generate narrowband radiation having a spectral width of ~200 GHz. According to measurements of the electric field strength profile using the THz-TDS method, such pulses, which comprise ~5 periods, have a duration of about 5 ps. The energy of the THz wave as measured by the Golay cell averages 4 μJ.

The THz-induced dynamics of the order parameter in the ferroelectric is studied by detecting the nonlinear optical signal intensity at the optical SH frequency [12].

Figure 1 depicts the typical time and spectral shapes of the incident THz wave, the THz wave passed through the sample, and the optical SH wave induced by the electric field of the THz pulse.

The energy of the incident wave changes (to a maximum at the frequency of 1.2 THz) when its frequency is changed. In the time shape (Fig. 1a), only the period changes, while the character of the dependence remains approximately the same for all frequencies in the range of 0.5–2.2 THz. For 0.9 THz, the pulse duration is ~8 ps. When trying to further increase the frequency, the signal becomes indistinguishable from noise. In the frequency domain (Fig. 1b), the pulse frequency shifts, its half-width changing weakly in the range of 0.25–0.3 THz (which corresponds to the observed pulse duration).

The crystal transforms the incident pulse significantly. The amplitude of the passed wave decreases sharply (at the incident wave frequency of 0.9 THz,

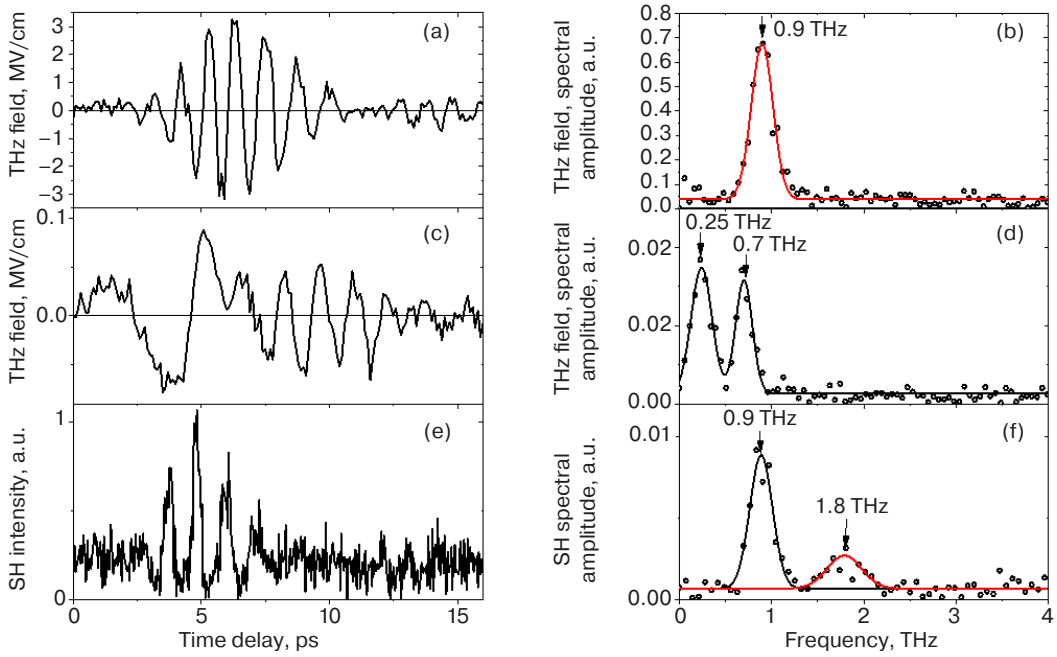

Fig. 1. For the 0.9 THz frequency, time (left) and spectral (right) dependencies of the incident THz wave (a), (b); of the THz wave passed through the sample (c), (d); of the optical SH wave induced by the electric field of THz pulse (e), (f)

the field amplitude is reduced by 30 times). Within the time dependence, the periodic function resembling the incident pulse is shifted by half of the pulse at its beginning while the aperiodic signal dominates. Accordingly, the frequency of 0.7 THz, i.e., decreased in comparison with the incident field frequency, appears in the spectrum of the passed THz pulse. In addition, the low-frequency component of 0.25 THz appears. It should be noted that 0.9 THz is the frequency of a sharp change in the transmittance character of THz radiation. At lower frequencies, a higher-frequency maximum dominates, its frequency approaching that of the incident wave. At higher frequencies, the pulse amplitude drops sharply, and only a low-frequency peak remains in the transmitted radiation. This corresponds to the absorption boundary found in broadband THz radiation in [12].

While the intensity of the SH looks like the incident pulse in the time domain, the two frequencies appearing in the frequency domain are those of the incident wave and its upper octave. This corresponds to quadratic dependence of the SH intensity on external field (on polarization in ferroelectrics) in the presence of the significant inactive by field contribution (non-switching polarization in ferroelectrics).

The SH intensity in the THz field can be represented as the decomposition either by the THz field E_Ω in the case of a non-ferroelectric crystal, as follows:

$$I^{2\omega}(E_\Omega) \propto (\chi^{(2)} + \chi_E^{(3)} E_\Omega)^2 (I^\omega)^2, \quad (1)$$

or by polarization $P(E_\Omega)$ in the case of ferroelectric crystal, as follows:

$$I^{2\omega}(P(E_\Omega)) \propto (P_0 + \chi_P^{(3)} P(E_\Omega))^2 (I^\omega)^2, \quad (2)$$

where $\chi^{(2)}(2\omega, \omega, \omega)$ is crystallographic quadratic susceptibility; $\chi_E^{(3)}(2\omega, \Omega, \omega, \omega)$ is cubic susceptibility.

Obviously, relations (1) and (2) are identical in the case of linear dependence of $P(E_\Omega)$, for example, in weak fields. In general, the dependencies of the SH intensity on the THz field should be studied for distinguishing (1) and (2).

When decomposing the square of sum, two field-dependent terms that are linear I_2 and quadratic I_3 appear, as follows:

$$I_2 \propto \chi^{(2)} \chi_E^{(3)} E_\Omega, \quad (3a)$$

$$I_3 \propto (\chi_E^{(3)})^2 (E_\Omega)^2. \quad (3b)$$

It is these in the Fourier decomposition that give, respectively, the signals at the fundamental Ω and doubled 2Ω frequencies of the incident wave.

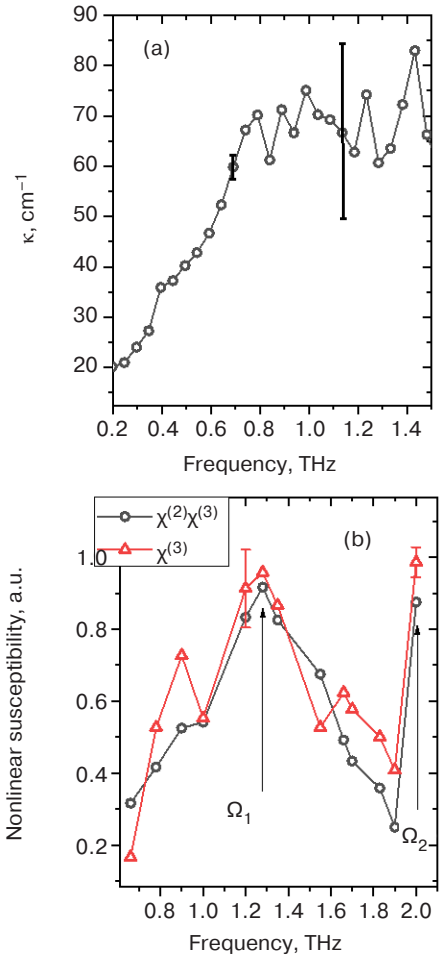

Fig. 2. Dispersion of the absorption coefficient and nonlinear susceptibility of PGO crystal

The dispersion dependencies of the absorption coefficient and nonlinear susceptibility of PGO crystal are shown in Fig. 2. The absorption coefficient (Fig. 2a) grows significantly up to the frequency of 0.7 THz where the accuracy of its determination does not exceed 5%. This is close to the value obtained earlier using the broadband THz spectroscopy technique. At higher frequencies, the error increases significantly. This is due to the fact that in narrow-band sensing the spectral lines of radiation are significantly deformed as they pass through the crystal (Fig. 1d). Finding the ratio of spectral amplitudes of the transmitted and incident radiation beyond the FWHM (full width at half maximum is pulse duration at half amplitude level) of the incident wave results in “division by 0”, or rather, by the noise signal. Similarly, beyond the FWHM of the passed wave, the division of the noise signal by a non-zero spectral amplitude occurs in the region of the incident wave spectral amplitude maximum. It is also possible to find spectral regions where both signals are noisy. At frequencies above 1.4 THz, the signal does not stand out against the noise.

The nonlinear susceptibility is calculated on the basis of spectral dependencies of the SH intensity (Fig. 1g) for

maxima at fundamental frequency by formula (3a) and at the doubled frequency by formula (3b). The result is shown in Fig. 2b. The dependencies are normalized to the maximum nonlinear susceptibility. It should be noted that the dependencies $\chi^{(2)}\chi_E^{(3)}$ and $\chi_E^{(3)}$ practically coincide. This implies the absence of spectral features in the THz range in the crystallographic susceptibility $\chi^{(2)}$, which corresponds to the nature of this susceptibility being the electronic type susceptibility. Thus, $\chi^{(2)}$ is simply a constant in the problem.

Cubic susceptibility $\chi_E^{(3)}$ clearly shows two maxima at Ω_1 = 1.3 THz and Ω_2 = 2 THz. While it has not yet been possible to measure the second maximum in more detail, attention is drawn to the measurement error at this point. The observed maxima correspond to the phonon modes of PGO crystal.

The studies show that in the case of THz spectroscopy in the area of phonon resonances, the generation technique of the SH induced by the narrow-band THz field is more informative compared to the commonly used TDS technique due to its significantly higher spectral resolution. This is due to the presence of the resonance denominator at the frequencies of the phonon modes $\chi_E^{(3)} \propto (\Omega - \Omega_0 - i\gamma\Omega)^{-1}$ in the expression for the cubic nonlinear susceptibility [16]. In addition, the distortion of the spectrum in propagation of the THz wave does not play a significant role, since the resonance itself "selects" the frequencies at which the resonance amplification occurs.

CONCLUSIONS

The presented investigation of the spectral characteristics of the absorption coefficient and nonlinear susceptibility based on the techniques of TDS and THz induced electric field generation of the optical SH demonstrates the presence of the region of fundamental absorption near phonon modes, as well as the resonance amplification of the cubic nonlinear susceptibility for two phonon modes Ω_1 = 1.3 THz and Ω_2 = 2 THz. The results are consistent with both earlier results from broadband TDS spectroscopy (absorption coefficient) and the results of Raman spectroscopy to determine the phonon mode frequencies. The spectral dependence of the cubic nonlinear susceptibility in the THz range is obtained for the first time. The results are important for understanding the physics of interaction processes of THz radiation with ferroelectric crystals. The obtained values of the absorption coefficient can be used for developing THz devices on PGO crystals.

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Authors' contributions

V.R. Bilyk—experimental studies of the THz-TDS of the PGO crystal; analysis of experimental data.

K.A. Brekhov—experimental studies of the PGO crystal nonlinear response dynamics.

M.B. Agranat—definition of the research concept; writing the initial text of the manuscript.

E.D. Mishina—definition of the research concept; writing the initial text of the manuscript; analysis of experimental data, calculation of the PGO crystal optical constants.

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