Generation of high power terahertz pulses and applications

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Here we report on our latest results in creating high-power terahertz pulses by means of optical rectification technique and their application in various fields. We investigated terahertz generation in organic crystals DSTMS, DAST and OH1 directly pumped by a Cr: forsterite laser at central wavelength of 1.25 μm. This pump laser technology provides a laser-to-THz energy conversion efficiency higher than 3%. Phase-matching is demonstrated over a broad 0.1–8 THz frequency range. In our simple setup, we achieved hundred μJ pulses in tight focus resulting in electric and magnetic field larger than 10 MV/cm and 3 Tesla [1].

We've applied a novel method for measuring the THz spatial distribution of in the focal plane of a parabolic mirror based on generation of the second-harmonic radiation in the optical region of spectrum in centrosymmetric crystals under action of a powerful terahertz pulse with subsequent image transfer to iCCD camera (Fig. 1).

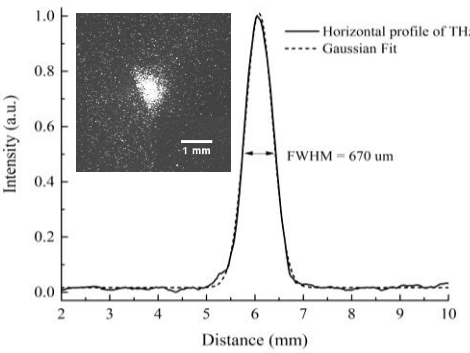

Fig. 1. THz intensity distribution for OH1 crystal

Application of different techniques of spatial profile transformation using interference of two chirped pulses in Mach Zehnder-type interferometer [2] as well as acoustooptical dispersion delay line [3] made it possible to generate THz pulses with a tunable center frequency in the range 0.5–2.5 THz (Fig. 2).

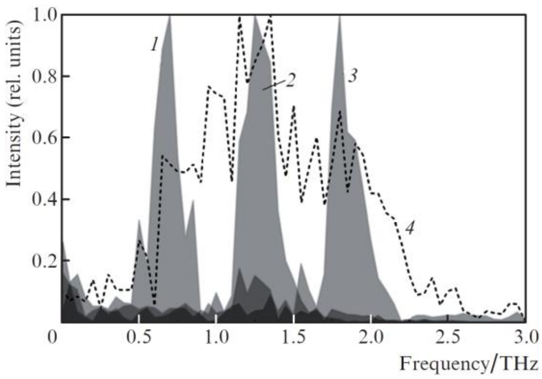

Fig. 2. Spectra of THz radiation under pumping of the OH1 crystal with laser pulses of different temporal shape and the delay between two chirped pulses (*1*) 0.5, (*2*) 1 and (*3*) 1.5 ps; (*4*) is the spectrum in the case of pumping with a single transform-limited pulse

Special attention is devoted to THz electric field-induced second harmonic generation in inorganic ferroelectric and centrosymmetric antiferromagnet as well. Second Harmonic Generation induced by the electric field of a strong nearly single-cycle terahertz pulse with the pick amplitude of 300 kV/cm is studied in a classical inorganic ferroelectric thin film of BaSrTiO3. The dependences of the SHG intensity on the polarization of the incoming light is revealed and interpreted in terms of electric polarization induced in the plane of the film. As the THz pulse pumps the medium in the range of phononic excitations, the induced polarization is explained as a change of the ferrolectric order parameter. It is estimated that under action of the THz pulse the latter acquires an in-plane component up to 3% of the net polarization.

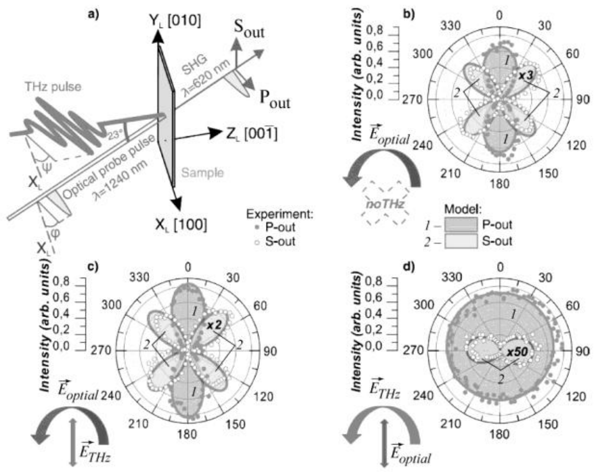

Fig. 3. Experimental geometry and polarization diagrams of the SHG intensity for various experimental geometries. (a) Experimental geometry. The axes of the chosen laboratory frame X_L, Y_L, Z_L correspond to [100], [010] and [001] crystallographic directions, respectively. ϕ – the angle between the electric field of near-infrared probe and the X_L-axis. ψ – the angle between the electric field of the THz pump pulse and the X_L-axis; (b) dependence of the SHG signal on ϕ without any THz pump; (c) the same, when the THz field is applied parallel with respect to the X_L-axis. The polarization of the SHG signal was set either in the P_{out} or S_{out}-state; (d) dependence of the SHG signal on ψ when the probe polarization was set to the P-state ($\phi = 0$). The polarization of the SHG signal was set either in the P_{out} or S_{out}-state. Dots correspond to experimental data and lines are fits. Values for S-state multiplied by 3, 2, and 50 for (b–d) respectively

Hence the data show that the THz electric field clearly affects the process of the second harmonic generation. To reveal ultrafast dynamics of these electric field induced changes, we performed pump-probe measurements of the SHG signal. In particular, the signal was measured as a function of the delay between the THz-pump and near-infrared probe pulses. Fig. 4a shows time-domain trace of the electric field of the THz pulse obtained with the help of electro-optical sampling. The measurements of the SHG signal from the BST film (Fig. 4b) reveal a similar dynamics during the overlap of the probe and THz-pump pulses. It points out that the nonlinear response is proportional to the THz electric field.

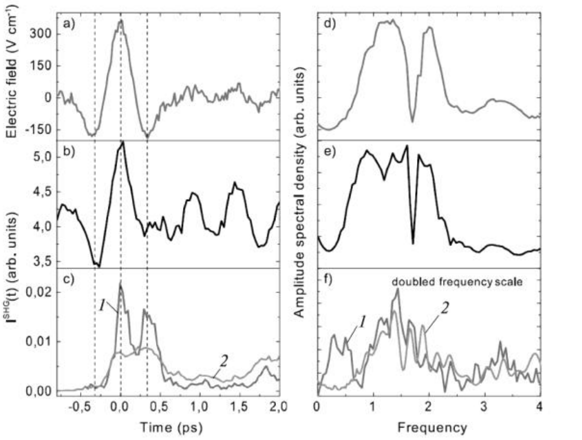

Fig. 4. THz-induced dynamics of nonlinear-optical response of the crystals. (a) Time trace of the electric field of the THz pump pulse; intensities of the SHG-signal: (b) for BST, (c) for STO (line 1) and Si (line 2); (d–f) Fourier transform spectra of the time traces shown in panels (a–c), respectively ((f) plotted in doubled frequency scale)

For comparison, we also measured temporal evolution of the SHG signals from centrosymmetric crystals excited by the intense THz pulse. The SHG transients for $SrTiO_3$ (STO) and Si are shown in Fig. 4c (lines 1 and 2, respectively). First of all, for the both cases of the centrosymmetric crystals the SHG signal of the unperturbed media are zero. Secondly, in the maximum of THz modulated signal the absolute values of the intensity of the SHG from BST is two orders of magnitude higher than those for the centrosymmetric crystals. Thirdly, the shape of the SHG response differs from the shape of the THz pulse.

Optical second harmonic generation at the photon energy of 2 eV in the model centrosymmetric antiferromagnet NiO irradiated with picosecond terahertz pulses (0.4–2.5 THz) at room temperature is detected (Fig. 5). The analysis of experimental results shows that induced optical second harmonic generation at the moment of the impact of a terahertz pulse arises through the electric dipole mechanism of the interaction of the electric field of a pump pulse with the electron subsystem of NiO. Temporal changes in optical second harmonic generation during 7 ps after the action of the pulse are also of an electric dipole origin and are determined by the effects of propagation of the terahertz pulse in a NiO platelet. Coherent oscillations of spins at the antiferromagnetic resonance frequency induced by the magnetic component of the terahertz pulse induce a relatively weak modulation of magnetic dipole optical second harmonic generation.

We have demonstrated that impact of electric component of picosecond single-cycle terahertz pulse (0.4–2.5 THz) leads to dynamic loss of the inversion center in centrosymmetrical (in terms of crystallographic and magnetic symmetry) antiferromagnet NiO.

This is accompanied by enablement for electrodipole SHG. This process takes place at the time of the THz pulse impact and is comparable in intensity to the spontaneous generation of SHG of the magnetodipole type allowed for symmetry at a pulse intensity of I^{THz} = = 0.2 mJ/cm^2 [4].

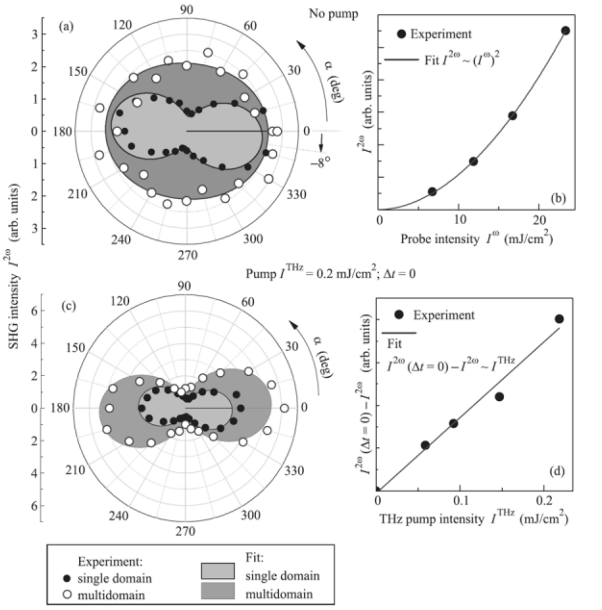

Fig. 5. (a, c) Experimental azimuthal dependencies of SHG intensity in single-domain (●) and multi-domain (○) states obtained in the absence (a) and under action (c) of THz pump pulse (I^{THz} = 0.2 mJ/cm^2) Radiation intensity at the fundamental frequency was I^{w} = 14 mJ/cm^2. Filled areas are theoretical calculations. (b) Experimental dependence (●) of SHG intensity I^{2w} as a function of pulse intensity at fundamental frequency in the absence of pump pulse and its approximation (curve). (d) Experimental dependence (●) of induced SHG intensity $I^{2w}(\Delta t = 0) - I^{2w}$ as a function of pump pulse intensity I^{THz} and its approximation (curve). Experimental dependencies (c) and (d) are obtained in multi-domain state at analyzer position α = 0

The experiments were conducted using Unique Facility "Terawatt Femtosecond Laser Complex" in the Center for Collective Usage "Femtosecond Laser Complex" of JIHT RAS.

The study was supported by the Russian Science Foundation (project No. 17-19-01261).

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