32nd URSI GASS, Montreal, 19-26 August 2017

High-order Nonlinearities in GaP and diamond driven by an ultrastrong THz Field

Carlo Vicario
SwissFEL, Paul Scherrer Institute, 5232 Villigen-PSI, Switzerland

Abstract

Intense THz electric fields are used to drive highly nonlinear optical phenomena in gallium phosphide and diamond. Here we report on a laser giant spectral broadening and ultrafast metallization in GaP stimulated by THz electric fields. Moreover we present THz-driven electro-optical gate with transition time orders of magnitude faster than conventional electronics switches.

1. Introduction

Ultrastrong Terahertz (THz) transients have the potential to control instantaneously macroscopic properties of the matter [1-2]. By employing our recently developed THz bullet configuration [3, 4] with extreme electric and magnetic field strength we could initiate ultrafast electronic and magnetic dynamics [5, 6].
Here we demonstrate high-order nonlinear optical effects driven by the above THz stimuli. As driving field we employ the THz single-cycle source available at the Paul Scherrer Institute which provides pulse energy up to 50 µJ and record-high electric field up to 83 MV/cm in the spectral range between 0.1 and 10 THz [7,8]. The source is based on optical rectification in organic crystals of a mid-infrared pulse from an optical parametric amplifier pumped by a TW Ti:sapphire laser system.

2. THz-driven extreme nonlinearity in GaP

We report on the experimental observation of extreme laser spectral broadening by cross-phase modulation (XPM) and change in optical transmission in gallium phosphite induced by the tens of MV/cm THz single-cycle field [7, 8]. In the experimental setup the ultra-strong THz and a delayed co-propagating laser probe are focused in 50 µm thin electro-optical GaP crystal. At high THz field large XPM between the beams occurs and the laser spectrum is suddenly broadened. The experimental results are summarized in Fig. 1. The single-cycle THz pulses are realized by optical rectification in diethylaminosulfur trifluoride organic crystal (DAST). The THz effective electric field with peak strength of 24.4 MV/cm is reported in Fig. 1 a). Shown in Fig 1b) is the two dimensional map of the copragating laser spectral intensity (vertical axis) as function of the delay with respect to the THz wave (horizontal axis). The laser spectrum is instantaneously broadened by the THz-driven XPM due to simultaneous Pockels and Kerr effects. At the maximum amplitude, the nonlinear change of the index of refraction results in 5-fold spectral broadening up to 180 nm.

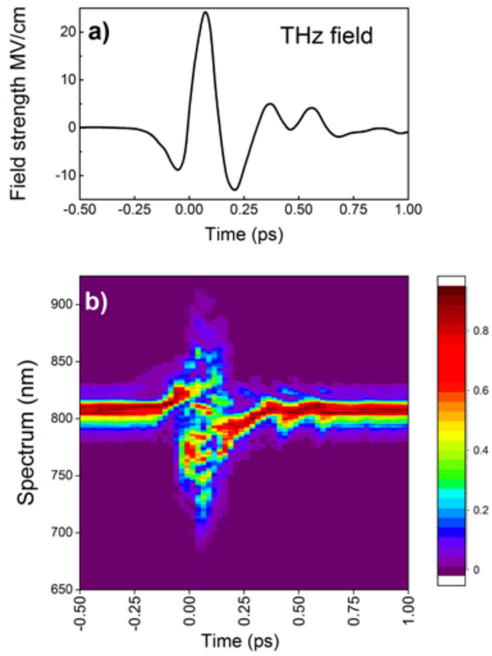

Figure 1. a) large THz electric field induces extreme spectral broadening on a co-propagating 800 nm laser probe spectra as function of the delay b).

After the main THz peak the original laser spectral shape is immediately restored. At the highest fields we observe also a phase transition of the intrinsic GaP from semiconductor to metal like with consequent drop of the nIR laser transmission, Fig. 2. The drop of the transmission occurs on the THz sub-cycle and scale nonlinearly with the stimulus peak field. At the THz maximum internal field of 24 MV/cm the laser transmission is reduced by more than 70%. This phenomenon is alleged to interband Zener tunneling and charge carrier density modification by impact ionization turning the semiconductor in a metal-like transient state at THz sub-cycle time scale [8].
The measurements indicate the coexistence of fast and retarded carrier dynamics. The fast one occurs on a temporal scale comparable with the driving THz field intensity. The quick reduction of the optical transmissivity at the THz peak is followed by a slow recovery.

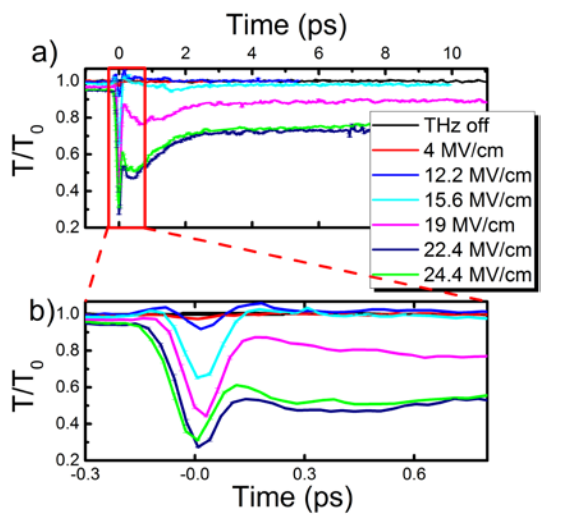

Figure 2. Dynamical change of the optical transmission in GaP induced by THz field at different strength on a) long and b) short time scale.

3. THz-driven ultrafast optical gate in diamond

Polarization switching of ultrashort laser pulses finds several applications in signal processing and laser technology. The maximum transition speed of the present Pockels switch is restricted to several nanoseconds due to the limited bandwidth of the driving high-voltage electronics. Here we show that by replacing the electric field provided by state of the art electronics by > 50 MV/cm field of a single-cycle THz transient the electro-optical gating process can be driven orders of magnitude faster [9]. As switching mechanism we employ a THz-induced Kerr polarization rotation in thin diamond window. Our THz pulse gating setup is schematically shown in Figure 3 a). The strong THz transient induces a polarization rotation and gates the incoming 50 femtosecond nIR laser pulse. The underline process is Kerr polarization rotation where the phase retardation is proportional to the which is proportional to THz pulse intensity $|E_{THz}(t)|^2$. Shown in Fig. 3 b) is $|E_{THz}(t)|^2$ (red curve) and the transmitted laser intensity after the polarizer at 1700nm (green curve) and 800 nm (blue curve). The gating temporal evolution is fully governed by the THz intensity shape. The THz-based Kerr switch exhibits ultrafast gating time of 125 fs and a rise/fall time of ≈120 fs. The gate driven at THz frequencies overcomes current electronics bandwidth limitations and undesired nonlinearities typically observed in semiconductor Kerr shutters at high laser intensities. While the presented proof-of-principle experiment has been performed at 800 nm and at 1700 nm, the exceptional broadband transmission of diamond makes the THz-driven switch applicable across spectral range from the deep-UV to the far infrared. In addition, diamond has exceptional heat conductivity and is thus ideal for high power all-optical modulators.

The polarization gating technique allows moreover for imaging the 3-dimensional THz beam in time by recording the THz-induced polarization rotation of the ultrashort probe beam on the 2-dim CCD sensor. Indeed, the polarizer transmission axis let pass only the near IR probe which underwent polarization rotation at a given time. A sequence of different beam slices recorded along the THz bullet at different times permit to reconstruct the THz 3-dimensional profile Fig. 3 c). The tomography shows the THz beam to exhibit a slight asymmetry at the head and the tail (at 190 fs and 80 fs, respectively).

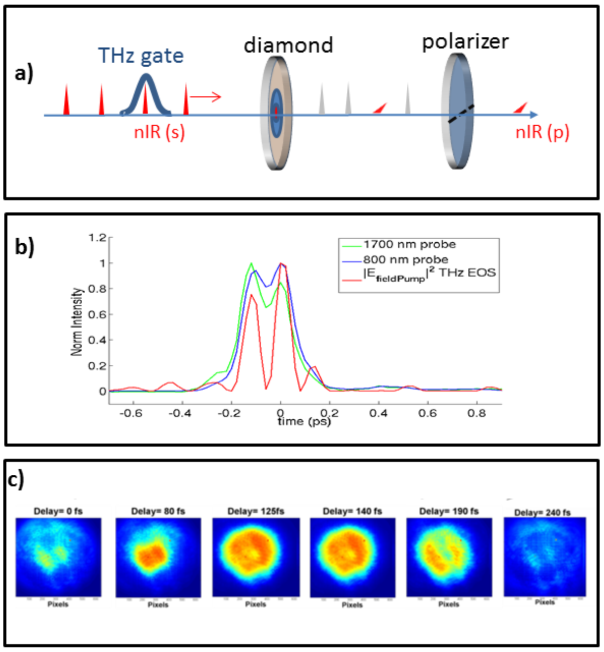

Figure 3. a) Schematic of the THz-driven optical modulator. b) the optical transmission for laser pulse at 1700 nm (green curve) and 800 nm (blue curve) is governed by the THz intensity shape (in red). c) the optical gate can be used for tomographic reconstruction of the THz pulse in the three dimensions.

The presented ultrafast THz-based all-optical switch discloses new applications in signal processing and pulse gate as well as advanced spatio-temporal THz beam diagnostics.

In conclusion, we have shown coherent high-order nonlinearities induced by strong THz field. Extreme spectral broadening and ultrafast metallization is realized in GaP. We also demonstrate novel Kerr-based optical gate in diamond window order of magnitude faster than conventional switching.

4. Acknowledgements

I acknowledge financial support from Swiss National Science Foundation SNSF (200021_146769, IZLRZ2_164051 and IZKSZ2_162129) and association to NCCR-MUST.

5. References

1.M. Tonouchi, “Cutting-edge terahertz technology”, Nature Photonics 1, 97 (2007).

2. T. Kampfrath, K. Tanaka and K. A. Nelson, “Resonant and nonresonant control over matter and light by intense terahertz transients”, Nature Photonics 7, 680 (2013).

3 M. Shalaby and C.P. Hauri, “Demonstration of a low-frequency three-dimensional terahertz bullet with extreme brightness”, Nature Commun. 6, 5976.

4. C. Vicario et al., “Generation of 0.9-mJ THz pulses in DSTMS pumped by a Cr:Mg_2SiO_4 laser”, Opt. Lett., 39, 6632, (2014).

5. C. Vicario et al., “Off-resonant magnetization dynamics phase-locked to an intense phase-stable terahertz transient”, Nature Photonics 7, 720 (2013).

6. M. Shalaby, C. Vicario and C.P. Hauri, “High-performing nonlinear visualization of terahertz radiation on a silicon charge-coupled device”, Nature Commun. 6:8439 doi:10.1038/ncomms9439 (2015).

7 Y. Shen et al., “Nonlinear Cross-Phase Modulation with Intense Single-Cycle Terahertz Pulses”, Phys. Rev. Lett. 99, 043901(2007).

8 C. Vicario, M. Shalaby and C. P. Hauri, “Subcycle Extreme Nonlinearities in GaP Induced by an ultrastrong THz Field”, accepted for publication in Phys. Rev. Lett. (2017).

9. Mostafa Shalaby, Carlo Vicario, and Christoph P. Hauri, “Extreme nonlinear Terahertz electro-optics in diamond for ultrafast pulse switching”, submitted to APL Photonics (2017).