Ultrafast Control of Even-Order Harmonic Generation from Solids by an Intense Terahertz Field

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***Abstract*— Nonperturbative even-order harmonics, physically forbidden in centrosymmetric crystals, are observed from both insulator (diamond) and semiconductor (silicon) samples driven by IR pulses, when in addition an intense THz electric field is applied to the solid. The time-resolved study of the high-harmonic spectra with the temporal profile of the THz electric field suggests a transient THz-field-induced symmetry reduction. We also investigate the harmonics' yields depending on the angles of the linear IR and THz polarizations with respect to the crystal axes. This work might pave the way for THz-based symmetry control in solids with important ramifications for petahertz electronics.**

I. INTRODUCTION

SINCE its first observation in 2011 [1,2], high-order harmonic generation (HHG) in bulk crystalline materials suggests a promising route towards a new generation of compact high- harmonic sources. Developing techniques to deliberately manipulate the harmonics' properties, such as the harmonics' yields and polarization states [3,4], becomes increasingly important for time-resolved spectroscopy of ultrafast electronic dynamics in the plethora of known solids and emerging quantum materials [5]. One viable approach is to control the selection rules of harmonic generation by reducing the symmetry of solids. This has been realized previously in conventional perturbative second-harmonic generation (forbidden in centrosymmetric crystals), e.g., by applying strain [6,7], DC electric fields [8] in silicon waveguides, or even exploiting a strong THz pulse in an inorganic ferroelectric [9] or semiconductors [10,11]. Extending symmetry control to *nonperturbative* HHG [10,11], we propose and demonstrate to use an intense THz electric field to break the centrosymmetry of cubic crystals and therefore transiently reduce the symmetry of these crystals on a sub-THz-cycle time scale.

II. RESULTS

The centrosymmetric crystal systems we study are the cubic systems diamond and silicon. Specifically, we choose free-standing, (100)-cut, 1-µm-thick and (110)-cut, 5-µm-thick Si crystals as well as a free-standing, (110)-cut, 20-µm-thick diamond crystal as samples. The experimental setup is shown in Fig. 1. The laser system employed for the experiments is a home-built infrared (IR) optical parametric amplifier (OPA) pumped by a Ti:sapphire chirped-pulse amplifier (40 fs, 4 mJ, 3 kHz). The signal and idler pulses of the OPA are centered at

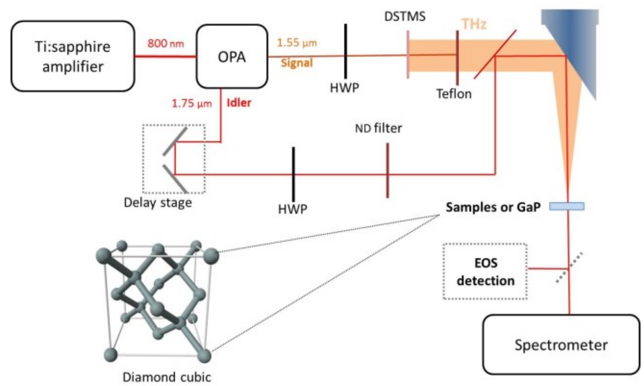

Fig. 1. Scheme of the experimental setup: OPA, optical parametric amplifier; HWP, half-wave plate; DSTMS, organic crystal [13]; ND filter, neutral-density filter; EOS, electro-optic sampling employing a GaP crystal.

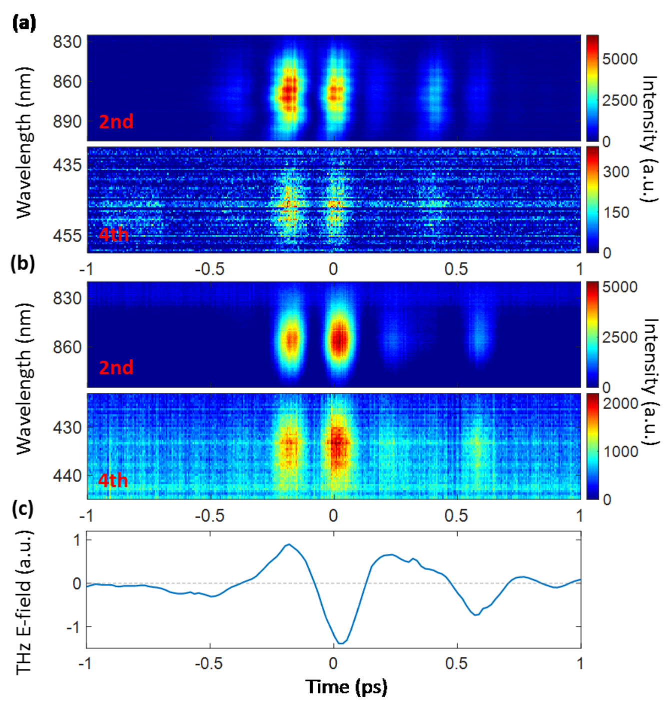

Fig. 2. Time-resolved even-order harmonics (2nd, 4th) from (a) diamond and (b) (100)-cut silicon, together with (c) temporal profile of the THz electric field, for parallel polarization configuration (IR || THz). The samples' rotation angles around their normal are chosen to maximize the even harmonics.

wavelengths of 1.55 µm and 1.75 µm, respectively [12]. By optical rectification of the signal pulse in a 0.6-mm-thick DSTMS [13] crystal, a ~4-µJ THz pulse (centered at ~3 THz) is generated. After being combined on a pellicle beam splitter, the THz and idler pulses are then focused onto the sample at normal incidence. A half-wave plate (HWP) is used to adjust the polarization direction of the IR idler driving field. The generated harmonics are recorded by an Ocean Optics USB2000 spectrometer. The incident peak intensities of the THz and IR pulses are 5×10^7 W/cm^2 and 1.5×10^{12} W/cm^2, respectively. The THz electric field is characterized via electro-optical sampling (EOS) with a 100-µm-thick GaP crystal (see Fig. 2(c)). Note that the generated intense THz field is still a comparably weak, perturbative dressing field.

Figure 2(a),(b) report measured high-harmonic spectra versus IR-THz delay, with the temporal profile of the THz electric field displayed in Fig. 2(c). We observe nonperturbative (as confirmed by the driving IR intensity scaling, not shown here) even-order harmonic generation (2nd, 4th) from both diamond and silicon. Here the polarizations of the THz and IR pulse are parallel, and the samples' rotation angles around their normal are chosen to maximize the even harmonics. The appearance of even-order harmonics coincides with the extrema of the THz electric field. This demonstrates the possibility to transiently control the cubic symmetry of silicon and diamond on a sub-THz-cycle time scale.

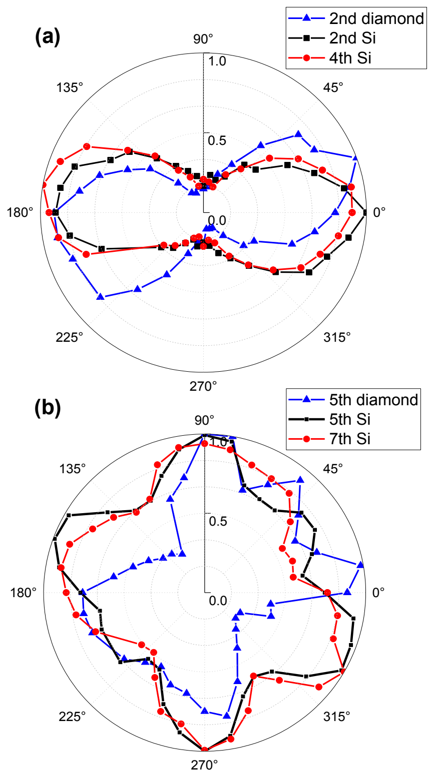

Fig. 3. Harmonic yields versus IR polarization angle. 0° corresponds to IR || THz, the samples' rotation angles around their normal are chosen to maximize the even harmonics. (a) Even harmonics (2nd, 4th in (110)-cut Si and 2nd in diamond) exhibiting a 2-fold symmetry; (b) odd harmonics (5th, 7th in (110)-cut Si and 5th in diamond).

In the measurements shown in Fig. 3, the THz polarization is kept fixed with respect to the crystal axes, and the harmonic yields are measured versus IR polarization angle, which is set by rotating the HWP shown in Fig. 1. From Fig. 3(a), it becomes clear that the THz-induced even harmonics in both samples exhibit a 2-fold symmetry, which is strongly connected to the THz electric field. This behavior is distinctly different from the pattern observed for the odd harmonics in Fig. 3(b).

Finally, we study the sample rotation dependence of the induced even-order harmonics in (100)-cut Si for parallel polarization configuration (IR || THz). As shown Fig. 4, we observe a four-fold symmetry of the 2nd, 4th, and 6th harmonics, compatible with the THz-dressed silicon crystal.

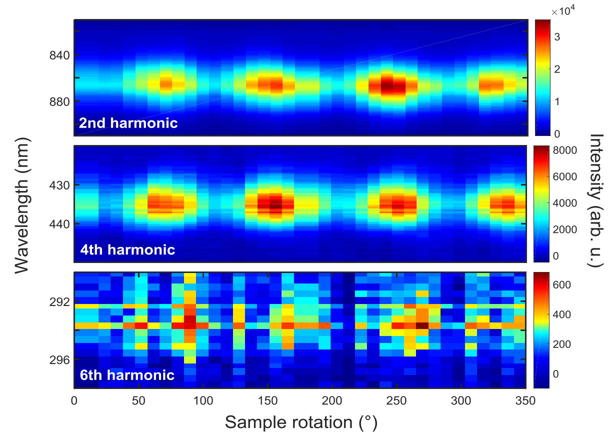

Fig. 4. Sample rotation dependence of the even-order harmonics from (100)-cut Si for parallel polarization configuration (IR || THz).

This work might pave the way for THz-based symmetry control in solids. More sophisticated control is possible by also employing elliptical/circular polarizations [3,4]. THz-dressing of crystals extends the toolbox for ultrafast manipulation of information for petahertz electronics and more generally for controlling nonperturbative light-matter interactions.

REFERENCES

[1] S. Ghimire *et al.*, "Observation of high-order harmonic generation in a bulk crystal," *Nat. Phys.*, vol. 7, pp. 138–141, 2011.
[2] O. D. Mücke, "Isolated high-order harmonics pulse from two-color-driven Bloch oscillations in bulk semiconductors," *Phys. Rev. B*, vol. 84, p. 081202(R), 2011.
[3] N. Tancogne-Dejean *et al.*, Ellipticity dependence of high-harmonic generation in solids originating from coupled intraband and interband dynamics," *Nat. Commun.*, vol. 8, p. 745, 2017.
[4] N. Klemke *et al.*, "Polarization-state-resolved high-harmonic spectroscopy of solids," *arXiv:1805.10453 [physics.optics]*, 2018.
[5] D. N. Basov, R. D. Averitt, and D. Hsieh, "Towards properties on demand in quantum materials, " *Nat. Materials*, vol. 81, pp. 1077–1088, 2017.
[6] R. S. Jacobsen *et al.*, "Strained silicon as a new electro-optic material," *Nature*, vol. 441, pp. 199–202, 2006.
[7] M. Cazzanelli *et al.*, "Second-harmonic generation in silicon waveguides strained by silicon nitride," *Nat. Materials*, vol. 11, pp. 148–154, 2012.
[8] E. Timurdogan *et al.*, "Electric-field-induced second-order nonlinear optical effects in silicon waveguides," *Nat. Photonics*, vol. 11, pp. 200–206, 2017.
[9] K. A. Grishunin *et al.*, "THz Electric Field-Induced Second Harmonic Generation in Inorganic Ferroelectric," *Sci. Rep.*, vol. 7, p. 687, 2017.
[10] H. Huang *et al.*, "High-Order Harmonic Generation from Solids Dressed by an Intense Terahertz Field," paper FM3F.3, *CLEO*, San Jose, CA, May 13-18, 2018, https://doi.org/10.1364/CLEO_QELS.2018.FM3F.3.
[11] G. Vampa *et al.*, "Strong-field optoelectronics in solids," *Nat. Photonics*, 2018, DOI: 10.1038/s41566-018-0193-5.
[12] C. Li *et al.*, "Generation of carrier-envelope phase stabilized intense 1.5 cycle pulses at 1.75 µm," *Opt. Express*, vol. 19, pp. 6783–6789, 2011.
[13] M. Shalaby and C. P. Hauri, "Demonstration of a low-frequency three-dimensional terahertz bullet with extreme brightness," *Nat. Commun.*, vol. 6, p. 5976, 2015.