Terahertz field-induced second harmonic generation through Pockels effect in a zinc telluride crystal

M. Cornet, A. Ould Hamouda, J. Degert, E. Abraham and E. Freysz
Université Bordeaux, LOMA, UMR 5798, F-33400 Talence, FRANCE
CNRS, LOMA, UMR 5798, F-33400 Talence, FRANCE

***Abstract*— We report on the second harmonic generation of a near-infrared pulse in a Zinc Telluride (ZnTe) crystal through the Pockels effect induced by an intense terahertz pulse, demonstrating cascading second-order nonlinear phenomena in the terahertz range.**

I. Introduction

The recent development of high power terahertz (THz) sources makes it possible to perform nonlinear optics in this spectral range. THz Kerr effect has been reported in liquids [1], ferroelectrics and amorpheous glasses. THz field-induced second harmonic generation (TFISH) has also been observed in a beta barium borate crystal [2] or liquids [3]. As expected for this third-order nonlinear phenomenon, the TFISH intensity is proportional to the THz intensity, I_{THz}, the square of the fundamental laser intensity, I_ω, and the third-order nonlinearity, $\chi^{(3)}$, of the material, so that:

$$I_{2\omega}^{TFISH} \propto (\chi^{(3)})^2 I_\omega^2 I_{THz} .$$

However, it is well-known that a cascade of two second-order nonlinear phenomena can also mimic a third-order process. We report hereafter on such a cascading phenomenon in zinc blende <110> ZnTe crystal.

II. Results

The experiment is the following: a 1.5 mJ, 800 nm linearly polarized laser beam is splitted into a pump beam used for THz generation and a probe for second harmonic generation (SHG). The pump pulse generates an intense THz radiation (E_{THz}= 20 kV/cm) through optical rectification in a $LiNbO_3$ crystal thanks to a tilted pulse front scheme [4]. Using two off-axis parabolic mirrors, the linearly-polarized THz beam is then focused into a 200 µm thick ZnTe crystal where it interacts collinearly with the IR probe pulse. Both THz and probe intensities can be adjusted by a pair of polarizers. The THz pulse polarization is set orthogonal to the c-axis of the crystal whereas a half-wave plate allows us to continuously rotate the probe polarization. The angle between the probe pulse polarization and the c-axis of the ZnTe crystal is denoted by θ hereafter. Finally, SHG is detected at 400 nm using a photomultiplier tube coupled to a lock-in amplifier.

First, performing the study with a <100> cut crystal, no SHG signal was recorded, whatever the configuration of both THz and probe polarization states. This indicates that TFISH is not visible in ZnTe under our experimental conditions. Furthermore, as expected in a <110> cut crystal, with a probe beam polarized along the c-axis of the crystal, no SHG was recorded in the absence of THz radiation. However, in its presence, a SHG signal was easily observed, whose temporal shape closely follows the evolution of $|E_{THz}(t)|^2$. In this experimental configuration (θ = 0°), it is well-known that the THz field-induced Pockels effect is maximized [5]. Actually, as the THz propagates through the crystal, it induces a Pockels effect, which rotates the probe beam polarization, enabling the latter to be frequency-doubled by the intrinsic second order susceptibility $\chi^{(2)}$ of the crystal. As a result, this Pockels-induced SHG signal can be expressed as:

$$I_{2\omega}^{Pockels} \propto (\chi^{(2)})^4 I_\omega^2 I_{THz} .$$

The two recorded intensity studies are displayed in the inset on Fig. 1. They clearly show the linear and quadratic dependences with respect to the THz and IR probe pulse intensities, respectively.

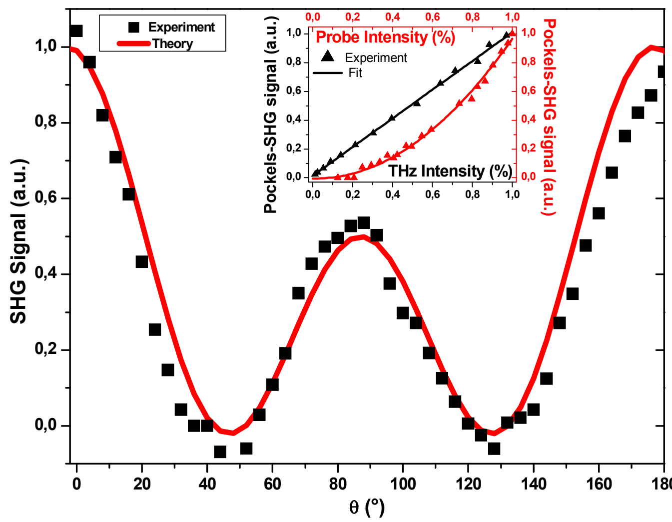

Fig. 1. Angular and intensity dependences of the Pockels-SHG signal.

We performed as well the θ angular dependence study of the Pockels-SHG signal. The experimental results (■) show a good agreement with the theoretical calculation (▬).

References

[1] M.C. Hoffmann, NC. Brandt, H.Y. Hwang, K.L. Yeh and K.A. Nelson, "Terahertz Kerr effect," *Appl. Phys. Lett*. vol. 95, 231105 (2009).
[2] J. Chen and X.-C. Zhang, "Terahertz-field-induced second-harmonic generation in a beta barium borate crystal and its application in terahertz detection," *Appl. Phys. Lett*. vol. 95, 011118 (2009).
[3] D.J. Cook, J.X. Chen, E.A. Morlino and H.M. Hochstrasser, "Terahertz-field-induced second-harmonic generation measurements of liquids dynamics," *Chem. Phys. Lett.* vol. 309, 221-228 (1999).
[4] J. Fülop, L. Palfalvi, S. Klingebiel, G. Almasi, F. Krausz, S. Karsch and J. Hebling, "Generation of sub-mJ terahertz pulses by optical rectification." *Opt. Letters* vol. 37, 557-559 (2012).
[5] P.C.M. Planken, H.K. Nienhuys, H.J. Bakker and T. Wenckebach, "Measurement and calculation of the orientation dependence of terahertz pulse detection in ZnTe," *JOSA B* vol. 18, 313-317 (2001).