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THz Generation By Optical Rectification And Competition With Other Nonlinear Processes

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Abstract. We present a study of the competition between THz generation by optical rectification in ZnTe crystals with two-photon absorption and second harmonic generation. The incident pump field for optical rectification is shown to be depleted by two-photon absorption and the generated THz field is shown to be significantly absorbed upon tight focusing by free-carrier absorption.

Keywords: Terahertz, ZnTe, optical rectification, nonlinear optics.

PACS: 42.65.-k, 42.65.Ky, 78.20-e.

INTRODUCTION

Ultra-fast terahertz (THz) spectroscopy is a powerful technique for studying a wide variety of materials [1]. In this context, a now widely used technique consists in generating THz pulses by optical rectification of femtosecond laser pulses [2,3]. A laser pulse is focused on a nonlinear crystal, which then radiates a THz pulse with a duration of few cycles of the electromagnetic field. In the spectral domain, the radiation is broadband, from about 100 GHz to typically 3 THz for a 100 fs pulse. For an excitation with a typical Ti:sapphire mode-locked laser, the generated THz power is only about few tens of nW, which stresses the importance of understanding and optimizing the THz generation.

Efficient optical rectification requires using materials with large second-order nonlinear susceptibilities and well suited phase-matching properties. ZnTe crystals with $\langle 110 \rangle$ orientation offer a very good compromise [4]. Unfortunately, when a ZnTe crystal is irradiated with a high-power laser pulse, other competing nonlinear processes such as second harmonic generation (SHG), two-photon absorption (TPA) and free-carrier absorption (FCA) also occur, reducing the THz generation [5,6,7]. Additionally, upon tight focusing, the size of the THz source (the laser spot) becomes smaller than the typical THz wavelength, resulting in a decrease of the THz generation due to diffraction [8]. Nevertheless, to date, existing analysis are not comprehensive or sometimes even contradictory [6,7,8].

In the present work, we study all these nonlinear mechanisms and show their interdependence. In particular, we show that free-carrier absorption cannot be neglected, as often assumed in the past.

EXPERIMENT

The excitation of the $\langle 110 \rangle$, 2 mm-thick, ZnTe crystal is provided by a 800 nm Ti:sapphire laser (100 fs) at 300 mW, using a 4 cm focusing lens. The ZnTe crystal is virtually transparent at this wavelength. The detection of the THz field is performed with a liquid helium cooled bolometer or with a free-space electro-optic sampling set-up [1].

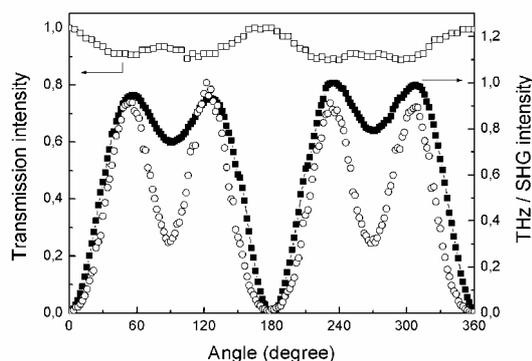


FIGURE 1. Closed symbols: THz intensity as a function of the ZnTe azimuthal angle. Open circles: SHG intensity upon focusing. Open squares: transmission at 800 nm close to focusing.

Figure 1 shows the THz emission as a function of ZnTe azimuthal angle. The THz intensity is proportional to the square of the nonlinear polarization which angle dependence is determined by the nonlinear susceptibility tensor. Figure 1 also displays the angle dependence of the SHG and of the transmission at 800 nm.

The competition between optical rectification, SHG, TPA and FCA was investigated using an open-aperture z-scan experiment [7]. The THz emission, SHG power and transmission of the 800 nm beam were measured as a function of the distance z between the beam waist and the crystal (Figure 2). As shown in Figure 2 (a), the THz emission drops significantly upon focusing. Moreover, TPA is responsible of a strong depletion of the 800 nm pump beam close to $z = 0$. Simple resolution of the propagation equation for TPA allows to extract the nonlinear absorption coefficient [7]. Here $\beta = 4.6 \text{ cm/GW}$.

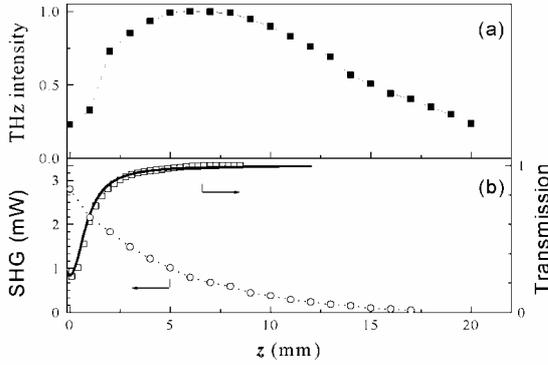


FIGURE 2. (a) THz intensity as a function of the distance to focus point. (b) Circles: SHG power. Squares: transmission at 800 nm. Solid line: fit of the TPA equation ($\beta = 4.6 \text{ cm/GW}$).

ANALYSIS

For large distances to the focus point (large z), the laser spot size $S \propto z^2$. Since the optical rectification is a second-order nonlinear process, the THz electric field is proportional to the laser intensity $I_0 = P_0 / S$, where P_0 is the input power. When diffraction effects can be neglected (spot size larger than the THz wavelength), the measured THz signal power is thus $P_{\text{signal}} \propto S (P_0 / S)^2 \propto S^{-1} \propto z^{-2}$ as can be seen in Figure 3 (thin solid line, large z). On the contrary, the situation is different when the THz emission is generated by a localized emitter of extension smaller than the THz wavelength [6,8]. The THz emission from the whole emitter section interfere constructively. As a consequence, the THz emitted power is expected [6] to become independent of the spot size close to

focus (dashed line). For $z < 5$ mm, other nonlinear process further reduce the THz emission and create a ‘z-hole’. Figure 3 (thick solid line) shows the calculated THz emission (in the diffraction limit), when TPA (only) is taken into account, which reduces the pump beam and thus the THz emission. Figure 3 also shows the calculated THz emission when FCA of the THz emission (only) is taken into account. Free carriers are generated by SHG (dotted line) and TPA (dash-dotted). The calculation in Figure 3 was performed using a value of 33 cm^{-1} for the absorption at 400 nm [7] and 6 cm^{-1} for the absorption in the THz range (as measured independently using a two-color experiment [9]).

In summary, THz generation by optical rectification is reduced upon focusing by depletion of the pump by TPA and by absorption of the THz radiation by FCA.

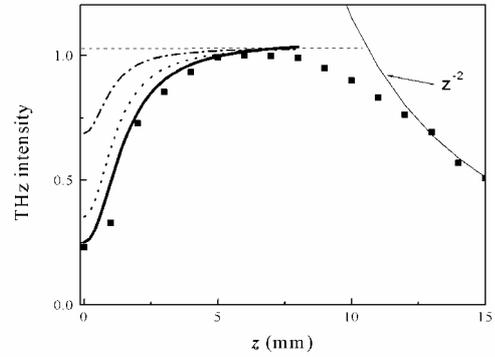


FIGURE 3. Thin solid line: THz emission for large z , when the THz source is larger than the THz wavelength. Dashed: THz emission in the diffraction limit (small spot size). Thick solid: THz emission including TPA only. Dotted: THz emission including FCA induced by SHG only. Dash-dotted: THz emission including FCA induced by TPA only.

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