Study on nonlinear processes affecting terahertz radiation generation from undoped GaP crystal

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Nonlinear processes associated with terahertz radiation generated via optical rectification from undoped GaP crystal, including second harmonic generation (SHG) and multiphoton absorption processes, are examined. Experimental results of polarization-resolved SHG are obtained.

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Terahertz (THz) technologies have a growing number of applications in medical imaging, standoff detection of bio-threat species, security screening, and time domain spectroscopy[1−5]. These applications call for the development of compact and efficient sources of high-power THz pulses. Recently, broadband THz pulse generation through optical rectification of a femtosecond laser of around 1 μm has been reported[6−8]. Femtosecond Yb-doped fiber lasers, especially femtosecond photonic crystal fiber (PCF) lasers at 1040 nm, have become an ideal pump source because they can offer extremely high output power while presenting environmental stability and compact size[9]. In a pump wavelength range around 1040 nm, undoped GaP is the most commonly used emitter that can fulfill the phase matching condition, and has very low absorption in the THz range[10]. Near single-cycle THz pulses of 150 μW at a repetition rate of 52 MHz are obtained from a 3-mm-thick bulk undoped GaP crystal pumped by high-power femtosecond pulses from an amplifier based on a large-mode-area (LMA) PCF[8]. In this letter, we focus on the concomitant nonlinear processes in THz radiation generation from undoped GaP crystal, including nonlinear absorption and harmonic wave generation.

An experimental setup was built to study polarization-resolved nonlinearities, as shown in Fig. 1. The pump source was an LMA PCF amplifier, which delivered femtosecond pulses at a repetition rate of 52 MHz with a pulse duration of 61 fs and a central wavelength of 1040 nm. The laser system can offer ultrahigh temporal and spatial resolutions in nonlinear experiments, and the thermal effects are very weak due to the short pulse duration and the absorption band of the GaP crystal. The pump power of the linearly polarized beam before lens L1 was set as 800 mW, and the polarization could be adjusted continually by a half-wave plate mounted on a motorized rotating stage (MRS). The focused beam was projected onto the <110>-cut GaP sample with a thickness of 1 mm. Then, the harmonic waves generated were filtered using a short pass filter (SF) from the transmitted fundamental beam and focused by a lens (L2) onto the detecting photodiode. The signals detected by the photodiode (PD) were dealt with using a lock-in amplifier (LIA) and then recorded by a computer. In our experiments, optical second harmonic generation (SHG) at a center wavelength of 520 nm and third harmonic generation (THG) at a center wavelength of 347 nm are observed, as shown in Fig. 2.

Figure 3 illustrates the SHG intensities as a function of the polarization of the pump laser beam by driving the MRS at a speed of 3 deg./s. As for THG, the intensities are so weak that anisotropy and polarization dependence are not available.

The polarization-dependent THz wave generation was carried out on a setup as described in Ref. [8]. The THz intensities were measured every 10 degrees using a Golay Cell detector. For zinc-blende crystals, both the optical SHG and THz generation via optical rectification belong to the $\chi^{(2)}$ process, and posses the same anisotropy[11]. The polarization dependence in a <110>-cut crystal can be expressed as

$$I_{110} = d_{14} I_0 [\sin^2\theta(1 + 3\cos^2\theta)], \quad (1)$$

where d_{14} is the nonlinear susceptibility coefficient for SHG and optical rectification, I_0 is the pump intensity, and θ is the angle between the linear polarization direction and the [001] axis in the (110) plane.

The experimental results shown in Fig. 3 agree very well with the theory, but the SHG efficiency of GaP is extremely low because of the sufficiently high absorption at the SHG frequency ($\lambda_{2\omega}$ = 520 nm is below the absorption band edge of 540 nm[12]). In spite of this, studying the SHG process in THz generation in GaP is necessary because the SHG process depletes pump power as a kind of dissipation factor.

The THz conversion efficiency based on broadband phase-matched different frequency generation in a second-order nonlinear medium has been theoretically

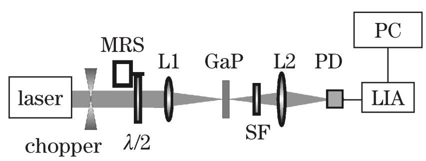

Fig. 1. Schematic of the experimental system.

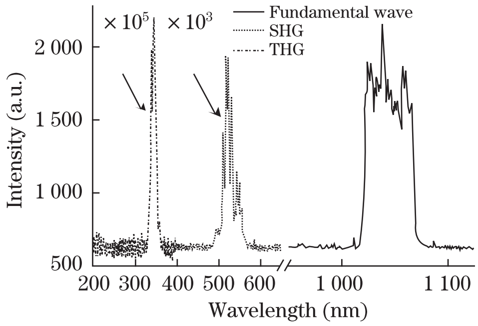

Fig. 2. Spectra of fundamental, second-, and third-harmonic waves.

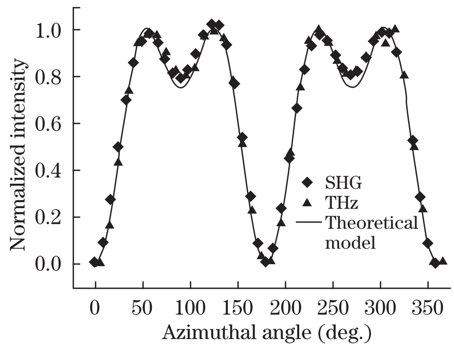

Fig. 3. Polarization-dependent SHG (diamonds), THz radiation (triangles), and theoretical model (solid line) for $\chi^{(2)}$ processes in <110>-cut GaP crystal.

studied[13], and the quadratic relationship between the generated THz pulse energy and pump pulse energy is well known. In our earlier work on high power THz wave generation, we found that the increase in THz output with pump power deviated from the quadratic rule and showed saturation effects (see Fig. 4). Therefore, we took nonlinear multi-photon absorption, which acted as an important source of pump power depletion, into consideration. We fitted the experimental data with.

$$\frac{\mathrm{d}I}{\mathrm{d}z} = -\left(\alpha + \sum_m \beta_m I^m\right) I, \tag{2}$$

$$P_{\mathrm{THz}} \propto \frac{d_{\mathrm{eff}}^2 L^\eta}{\tau^n}[1-(\alpha + k_2\beta_1 P_{\mathrm{opt}}^2 + k_3\beta_2 P_{\mathrm{opt}}^3)]P_{\mathrm{opt}}^2, \tag{3}$$

where β_m (integer $m \geqslant 1$) is the $(m+1)$-photon absorption coefficient; I denotes the pump beam intensity; P_{THz} and P_{opt} are the average powers of the THz radiation and pump beam, respectively; d_{eff} represents the effective nonlinear coefficient of the crystal; L^η is the working length with a factor $1 < \eta \leqslant 2$ depending on the phase matching condition achieved in the emitter; τ denotes the optical pump pulse duration; factor $1 < n \leqslant 3$ is related to the optical pulse chirp and the dispersion of the emitter. The linear, two-, and three-photon absorption coefficients are $\alpha = 0.3$ cm^{-1}, $\beta_1 = 0.05$ cm/GW, and $\beta_2 = 0.042$ cm^3/GW^2, respectively[14]. Coefficients k_2 and k_3 are weight factors normalized to the ratio of linear absorption. The second harmonic process affects THz generation in two aspects. On the one hand, SHG is a depletion of pump power, thereby directly reducing pump power, which is the second item in Eq. (3). On the other hand, the second harmonic wave generated is strongly absorbed by the GaP crystal[12], and produces free carriers, which absorbs THz photons during their recombination. This effect can be regarded as an equivalent two-photon absorption process, and corresponds to the third item in Eq. (3). The fitting results show that the weight factor for three photon absorption k_3 is larger than that for k_2 by 15 orders of magnitude, indicating that three photon absorption is the dominant factor in nonlinear absorption in bulk GaP. In our recent work, we also carried out Z-scan experiments[15]. Open aperture (OA) Z-scan curves show clear differences from the lower order two-photon absorption, providing evidences that under experimental conditions, no indirect two-photon absorption[16] occurs because of the lack of thermal effects, and direct three-photon absorption dominates the nonlinear absorption processes. These are in good accord with the results from the analysis of THz generation. By analyzing the differences between OA curves with and without the second harmonic wave, we know that although three-photon absorption dominates the absorptions, SHG also offers a contribution and cannot be disregarded. By measuring the absorption rate at the beam focus, which is the Position with both the maximum SHG conversion efficiency and nonlinear absorption, the difference caused by SHG depletion is about 4%. Compared with an absorption rate of 60% at the focus, the contribution of SHG can be estimated as about 7%. We also speculate that SHG may contribute to the optical Kerr-like effect through the cascaded second-order nonlinear effects, i.e., through the Kerr-like effect[17], and this serves as a highly important concomitant factor in THz generation.

In conclusion, SHG and THG are observed as THz radiation is generated from an undoped <110>-cut GaP crystal when pumped by high power femtosecond pulses. The polarization-dependent characteristic of the SHG process is studied, and the effects of SHG on THz conversion are discussed.

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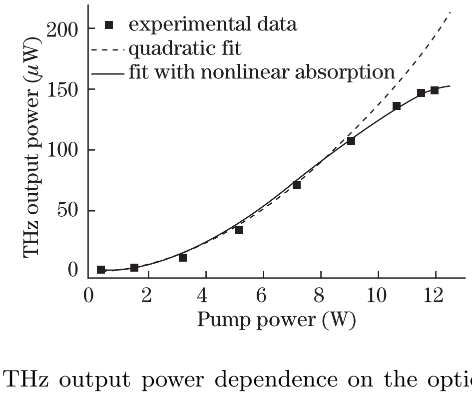

Fig. 4. THz output power dependence on the optical pump power. Solid squares are experimental data, the dashed line is the quadratic fit, and the solid line is fitted by Eq. (3).

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