THz Generation from Monoclinic Centrosymmetric GaTe Crystals Pumped across Bandgap due to Carrier Unidirectional Diffusion

Guibao Xu, Guan Sun, and Yujie J. Ding
Department of Electrical and Computer Engineering, Lehigh University, Bethlehem, PA 18015
yud2@lehigh.edu
Ioulia B. Zotova
ArkLight, P.O. Box 2, Center Valley, PA 18034
Krishna C. Mandal, Alket Mertiri, and Gary Pabst
EIC Laboratories, Inc., 111 Downey Street, Norwood, MA 02062
Nils Fernelius
Materials and Manufacturing Directorate, Air Force Research Laboratory, Wright-Patterson AFB, OH 45433

Abstract: We have observed unique polarization and azimuth-angle characteristics of THz waves, generated from anisotropic monoclinic GaTe when pump photon energy is below and above its bandgap, due to carrier unidirectional diffusion.

OCIS codes: (320.7130) Ultrafast processes in condensed matter, including semiconductors; (320.7100) Ultrafast measurements; (320.7080) Ultrafast devices

1. Introduction

There are two competing mechanisms for THz generation in semiconductors: optical rectification and photocurrent surge. All the binary compounds studied for THz generation in the past possess second-order nonlinearities. Therefore, when the pump photon energy is below the bandgap of the compound, optical rectification plays the dominant role for THz generation whereas above the bandgap photocurrent surge or combination of the two plays the primary role [1]. *There has been no report on the THz generation from any centrosymmetric (i.e. having vanishing second-order nonlinearity) binary compound semiconductor.*

Here, we report our new results on the THz generation from a series of GaTe crystals doped with Ge, Bi, and Pb at different levels. The highest output powers were measured to be 1.9 μW and 68 nW, corresponding to the pump photon energies of above and below the bandgap of the crystals, respectively. We have also observed unique polarization and azimuth-angle characteristics which are primarily dictated by the crystal symmetry.

Since GaTe (monoclinic, C2/m) has a layer-stacking structure with each layer being consisting of four monoatomic sheets in the sequence of anion-cation-cation-anion, it exhibits high anisotropy in terms of optical and electrical properties along intralayer and interlayer directions. Two-thirds of Ga-Ga dimers are oriented perpendicular to the layers while the remaining one-third of Ga-Ga dimers is almost parallel to the layers. Although such a structure is similar to that for GaSe, unlike GaSe, GaTe is centrosymmetric, and therefore, its second-order nonlinearity completely vanishes. In contrast, GaSe crystals have been used for THz generation primarily based on second-order nonlinear coefficients [2].

2. Crystal preparation and experiment description

Table 1. Summary measurements on GaTe crystals

Crystal ID	Dopant (ppm)	Highest Output Power	
		P1* (μW)	P2† (μW)
GTGB8601	Ge/Bi (50/50)	1.422	0.068
GTGP8901	Ge/Pb (50/50)	1.474	0.066
GTG8403	Ge (600)	1.863	0.029
GTG8101	Ge (3000)	1.645	0.073
GTG8112	Ge (4000)	1.789	0.037

Excitation wavelengths: * - 391 nm; † - 782 nm

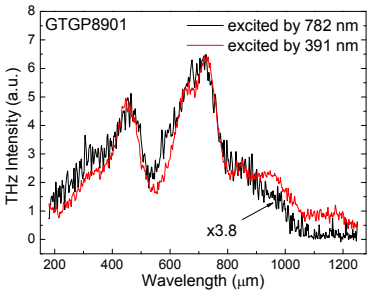

Fig. 1. THz spectra emitted from GaTe crystal.

Each undoped GaTe crystal was synthesized by melting mixture of stoichiometric amounts of high-purity constituent elements. The obtained ingot was placed in a conically-tipped quartz ampoule with appropriate high purity dopant elements, which was evacuated and sealed off at ~ μTorr. The ampoule was then heated by a two-zone horizontal furnace in order to introduce different dopants into GaTe. X-ray diffraction confirmed the monoclinic

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structure of all grown GaTe crystals. Due to the unique layer structure of GaTe, they can be easily cleaved along the (100) plane normal to the crystal surface. Broadband THz pulses were generated by using a Ti:sapphire regenerative amplifier at 782 nm below the bandgap of the crystals and a frequency-doubled amplifier at 391 nm above the bandgap (~180 fs, 250 kHz). When the excitation beam was focused onto each GaTe crystal, the THz radiation was collimated and then focused onto a pyroelectric detector in either transmission or reflection geometry.

3. Results and discussions

At an average power of 352 mW of the 391-nm excitation beam, the highest average THz output powers were well above 1 μW from all five samples, see Table 1. *To the best of our knowledge, this is the first demonstration of THz emission from a monoclinic centrosymmetric crystal.* When these crystals were excited by the 782-nm amplifier beam, the highest average THz output powers were approximately 20 times lower. The bandgap of the undoped GaTe crystal is about 1.7 eV (729 nm). When each crystal was pumped below its bandgap, free electron and holes can be generated by donor-acceptor pair absorption located at 1.57 eV (790 nm) [3], followed by ionization of bound electrons and bound holes by the surface electric field. In such a case, the free-carrier densities are significantly lower than those generated by the 391-nm excitation beam. This is the primary reason why the THz output powers generated by the 782-nm beam are much lower. Among all the crystals with different dopants or doping concentrations, the highest output power was measured to be 1.863 μW on GTG8403 under the pump wavelength of 391 nm. The output power was significantly increased as the doping concentration of Ge was increased, see Table 1. Since the undoped GaTe crystals are p type, Ge dopants can be used to reduce the free-hole density. As a result, the mobility of the GaTe crystal is increased, and therefore, the THz output power is increased accordingly. Since the output powers measured under 782 nm were measured under a large incident angle in the reflection geometry, not all the emitted powers were collected. This is the reason why the dependence of the output power on the Ge concentration is not consistent with that under 391 nm. Based on Fig. 1, THz power spectra cover the frequency range of 0.5 - 4.0 THz. When the pump intensity is ≤ 44 W/cm^2, the power dependence is well fitted by a square power law, whereas for the pump intensity of > 44 W/cm^2, the dependence exhibits an increasing deviation from the square power law, see Fig. 2(a). Such a behavior is primarily caused by the slowdown the carrier diffusion by the photo-Dember field. The THz output power exhibits periodic oscillations as either pump polarization angle or azimuth angle is increased. After taking into consideration Fresnel reflections at the crystal/air interfaces, our data are well fitted by respective theoretical curves, see Figs 2(b) and 2(c). The visible deviations are caused by spatial inhomogeneity of the crystal. On the other hand, one can see from Fig. 2(b) that the THz polarization angle was a constant as the pump polarization angle was increased. Moreover, the THz polarization angle linearly increased with increasing the azimuth angle through crystal rotation, see Fig. 2(c). These behaviors can be well understood by considering photocurrent surge as the mechanism for THz generation. After considering electrical anisotropy of GaTe [4], it turns out that the photocurrent surge primarily originated from the diffusion of the photogenerated carriers along the [001] direction, which is perpendicular to *a* and *b* axes of the GaTe crystal within the (100) plane of the crystal surface. *We believe this is the first observation of the THz generation induced by the carrier unidirectional diffusion within the crystal surface.* This work has been supported by U.S. AFRL.

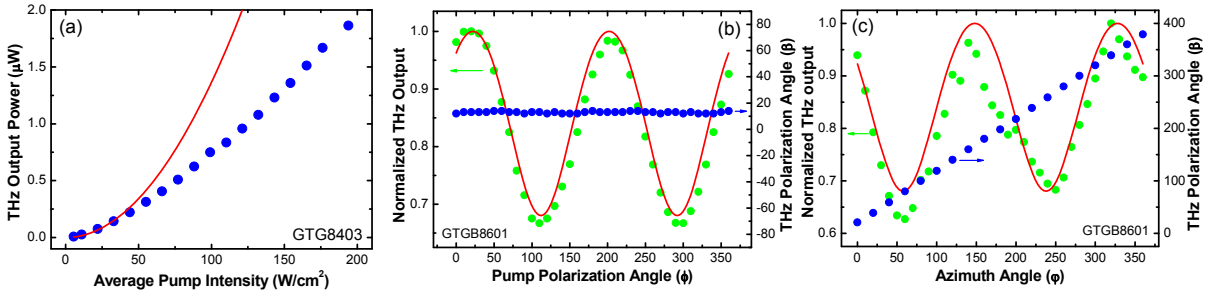

Fig. 2. Dots: data measured at pump wavelength of 391 nm. Solid curves: theoretical results for (a) quadratic fit, (b) and (c) after taking into consideration Fresnel reflections at crystal surface.

4. References

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