Emission of Terahertz Radiation from SiC

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We report the emission of strong coherent broadband terahertz radiation from 6H-Silicon-Carbide (SiC) excited with optical pulses. The measured terahertz spectral signal-to-noise ratio is better than one thousand. The dependence of the terahertz signal on the optical pump polarization, the pump angle of incidence, and the pump power indicate that the terahertz radiation is generated via second order optical nonlinearity (optical rectification).

Silicon Carbide (SiC) is a wide bandgap semiconductor possessing high mechanical stability, chemical stability, and thermal conductivity. As a result, it is a promising candidate for high-field and high-power electronics [1, 2], including applications intended for high temperature environments. Recently, SiC has also been explored for terahertz (THz) applications. Electrically-pumped THz emitters based on electronic transitions between impurity states have been demonstrated [3]. SiC devices, such as IMPATT oscillators, are being explored for high power applications in the low THz region [4].

In this paper, we present for the first time results on the emission of coherent terahertz (THz) radiation from semi-insulating 6H-SiC excited with near-IR femtosecond optical pulses. Broadband THz generation and detection in semiconductors with femtosecond (fs) optical pulses is a powerful and well-studied mechanism with applications in spectroscopy, imaging, and sensing [5]. Many semiconductors, such as GaAs and InAs, emit coherent broadband THz pulses upon excitation with femtosecond optical pulses due to free-carrier generation and subsequent carrier dynamics in internal or externally applied electric fields [5, 6]. 6H-SiC has a large spontaneous polarization and, therefore, a large permanent bulk electric field [7]. However, the large indirect bandgap of SiC (> 3 eV [8, 9]) implies that free-carrier generation via direct interband absorption is not possible. Free-carrier generation through two-photon or defect absorption is possible. Semi-insulating compensted 6H-SiC has a large number of donor and acceptor like states in the bandgap [10, 11, 12]. A nonlinear mechanism, such as optical rectification [5], can also be responsible for the generation of THz radiation. The nonlinear optical properties of various SiC polytypes have been previously studied, and 6H-SiC is known to have a large second-order nonlinear susceptibility comparable to crystals such as lithium niobate and KTP [9, 13]. In this paper, we study the THz radiation dependence on the optical pump polarization, the pump angle of incidence, and the pump power. We show that second order optical nonlinearity, and not free-carrier dynamics, is responsible for THz emission. Our results demonstrate that SiC can be used for generating strong THz radiation.

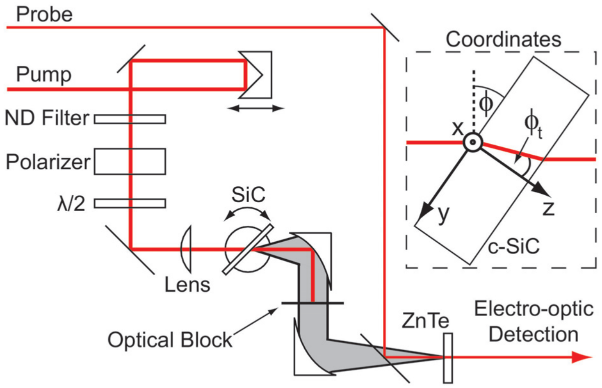

FIG. 1: Schematic of the experimental setup.

For our experiments, we used a mode-locked Ti:Sapphire laser system to produce optical pump pulses with center wavelength ∼780 nm, duration ∼90 fs, and repetition rate 81 MHz. Pulse energies varying from 1-15 nJ were used for excitation. Pump polarization angle (θ) was controlled with polarizers and a half-waveplate. Vanadium-compensated semi-insulating 6H-SiC (0001) dies, 370 μm thick, were mounted vertically on a rotation stage such that the crystals could be rotated about the vertical axis, thus controlling the angle of incidence ϕ of the pump beam. The pump beam was chopped at 2.5 kHz and focused onto the sample with a 25 mm focal length lens. The emitted THz radiation was collected in a nitrogen-purged environment with off-axis parabolic mirrors and detected in the time domain by means of a standard electro-optic detection setup using a (110)-ZnTe crystal [5, 14]. Strong THz pulses with maximum spectral power signal-to-noise ratios better than one thousand and detection-limited bandwidths wider than 3 THz were observed. A representative electric field transient and its accompanying spectrum are shown in Figure 2. The dip in the spectrum around 1.9 THz is discussed later in this paper.

Terahertz emission via the generation and subsequent motion of free-carriers has generally no strong dependence on the pump polarization or the pump angle of incidence (other than that which is related to the transmission/reflection of the pump and the emitted THz radiation at the crystal interfaces). In contrast, the emission of THz radiation from nonlinear optical rectification is strongly dependent on the pump polarization and angle of incidence as dictated by the form of the second-order

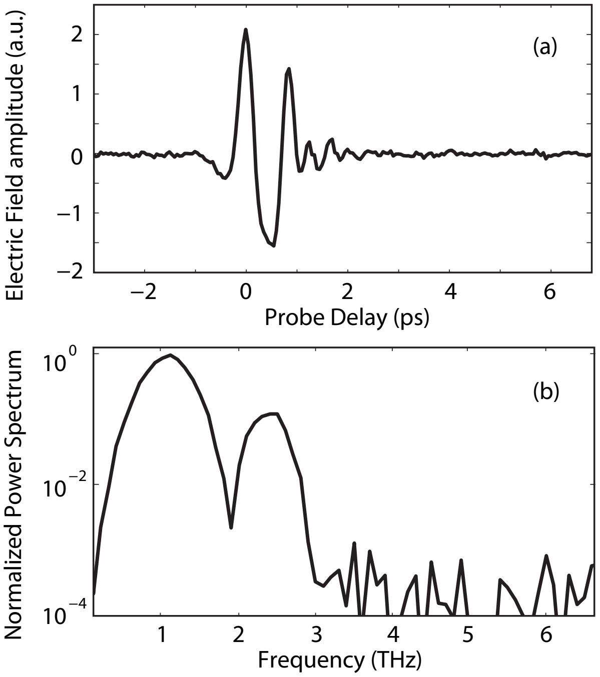

FIG. 2: (a) Observed electric field transient. (b) Corresponding power spectrum.

nonlinear susceptibility tensor $\chi^{(2)}_{ijk}$ [5, 15]. 6H-SiC has C_{6v} crystal symmetry and therefore, under Kleinman's condition, there are only two independent components of $\chi^{(2)}_{ijk}$ [15]. Assuming the z-axis to be oriented along the c-axis of the 6H-SiC crystal, one can write $\chi^{(2)}_{ijk}$ as follows [15],

$$\chi^{(2)}_{ijk} = \begin{bmatrix} 0 & 0 & 0 & 0 & \chi^{(2)}_{zxx} & 0 \\ 0 & 0 & 0 & \chi^{(2)}_{zxx} & 0 & 0 \\ \chi^{(2)}_{zxx} & \chi^{(2)}_{zxx} & \chi^{(2)}_{zzz} & 0 & 0 & 0 \end{bmatrix}. \tag{1}$$

In order to understand the dependence of the THz radiation on the polarization and the angle of incidence of the pump field, we describe the linearly polarized pump field by $E_o\,[\cos(\theta), -\sin(\theta)\cos(\phi), \sin(\theta)\sin(\phi)]$, as shown in Figure 1. The pump field inside the crystal is then,

$$\boldsymbol{E} = E_o \begin{bmatrix} t_{\mathrm{TE}}(\phi)\cos(\theta) \\ -t_{\mathrm{TM}}(\phi)\sin(\theta)\cos(\phi_t) \\ t_{\mathrm{TM}}(\phi)\sin(\theta)\sin(\phi_t) \end{bmatrix}, \tag{2}$$

where t_{TE} and t_{TM} are the ϕ-dependent TE and TM transmission coefficients, and $\phi_t = \arcsin[(n_{air}/n_{SiC})\sin\phi]$ is the propagation angle within the crystal relative to z. The induced nonlinear polarization $\boldsymbol{P}$ at terahertz frequencies is computed using,

$$P_i = \epsilon_0 \sum_{jk} \chi^{(2)}_{ijk} E_j E^*_k. \tag{3}$$

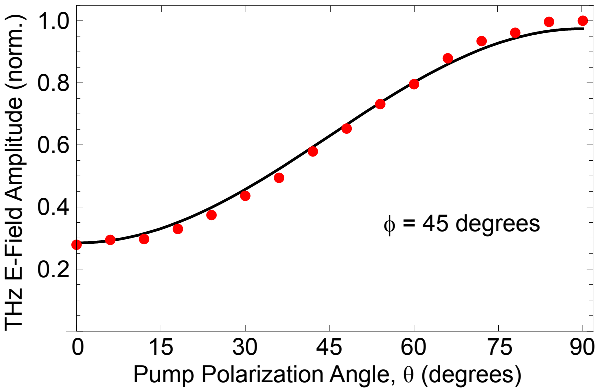

FIG. 3: Normalized peak-to-peak electric field versus excitation polarization. 0 degrees corresponds to TE polarization, 90 degrees to TM. Fit with $\chi^{(2)}_{zzz}/\chi^{(2)}_{zxx} = -2.1$.

Due to phase matching considerations, the radiated THz field $\boldsymbol{E}_{\mathrm{THz}}$ is proportional only to the component of $\boldsymbol{P}$ orthogonal to the direction of pump propagation inside the crystal,

$$\boldsymbol{E}_{\mathrm{THz}} \propto \hat{\boldsymbol{k}} \times (\boldsymbol{P} \times \hat{\boldsymbol{k}}) \tag{4}$$

where the unit vector $\hat{\boldsymbol{k}}$ equals $[0, \sin(\phi_t), \cos(\phi_t)]$. Finally, at the SiC-to-air interface, the THz field outside the crystal $\boldsymbol{E'}_{\mathrm{THz}}$ is related to the components of the THz field inside the crystal by,

$$\boldsymbol{E'}_{\mathrm{THz}} = \begin{bmatrix} t_{\mathrm{TE}}(\phi_t)\; E_{\mathrm{THz},x} \\ t_{\mathrm{TM}}(\phi_t)\cos(\phi)/\cos(\phi_t)\; E_{\mathrm{THz},y} \\ t_{\mathrm{TM}}(\phi_t)\sin(\phi)/\sin(\phi_t)\; E_{\mathrm{THz},z} \end{bmatrix}. \tag{5}$$

A few important conclusions can be drawn from Eqs. 1, 2, and 4. There is no THz emission at normal pump incidence ($\phi = 0$) regardless of the pump polarization (assuming a collimated pump beam). Also, if the pump beam is completely TE ($\theta = 0$) or completely TM ($\theta = 90^\circ$) polarized, the emitted THz will be completely TM polarized; otherwise, its polarization will be mixed.

Figure 3 shows the experimental results for the dependence of the THz electric field amplitude on the pump polarization for the pump angle of incidence equal to 45 degrees. The corresponding theoretical curve is also shown. Since the ZnTe-based electro-optic THz detection scheme is sensitive to the THz polarization, we have taken this dependence into account following the analysis of Chen *et al.* [14] in generating the theoretical curve. Figure 3 shows a strong dependence on the pump polarization and a good fit of the optical rectification theory to the experimental data. The ratio of $\chi^{(2)}_{zzz}/\chi^{(2)}_{zxx}$ is the only fitting parameter. A value of -2.1 was used to obtain the fit in Figure 3. This value of $\chi^{(2)}_{zzz}/\chi^{(2)}_{zxx}$ is in good agreement with the *ab initio* and the measured values [9, 13].

To further confirm the optical rectification process, we study the THz electric field amplitude dependence on the

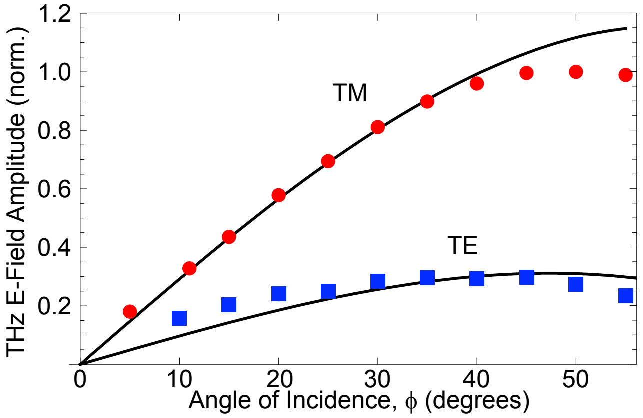

FIG. 4: Normalized peak-to-peak electric field versus angle of incidence for TE (filled square) and TM (filled circle) pump polarizations.

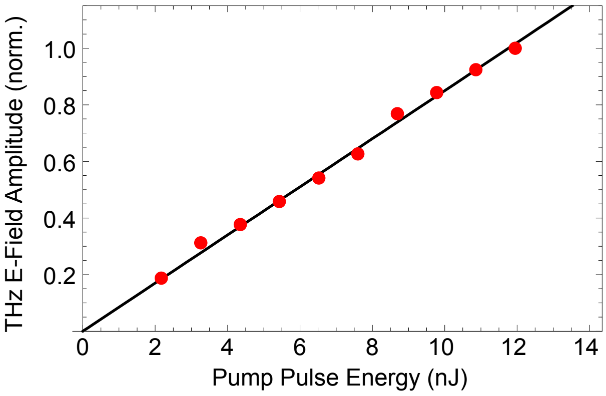

FIG. 5: Normalized peak-to-peak electric field versus excitation pulse energy (at 780 nm).

pump angle of incidence. The experimental results for TE and TM pump polarizations are shown in Figure 4 along with the theoretical curves. The theory is seen to fit the data well except at angles larger than ∼50 degrees. At large angles of incidence, total internal reflection of the THz radiation cone becomes significant, making it difficult to collect the emitted THz radiation efficiently and obtain accurate data. As expected from theory, E_{THz} approaches zero at small angles of incidence. Since the pump beam is not strictly collimated within the crystal, a small amount of THz radiation is detectable at angles near zero.

For a second order nonlinear emission process, the terahertz field amplitude is expected to be proportional to the pump power (or pump energy). In Figure 5, we plot the maximum THz pulse amplitude versus the pump pulse energy. The observed linear dependence is in agreement with THz generation via optical rectification.

The results presented here indicate that the mechanism responsible for THz emission in SiC is nonlinear optical rectification. The ratio of $\chi^{(2)}_{zzz}/\chi^{(2)}_{zxx}$ in 6H-SiC, the fitting parameter used in Figures 3 and 4, has been the subject of several theoretical and experimental investigations (see Table.I). The Equations given above show that the ratio of the THz field amplitudes for the TM and TE pump polarizations is linearly dependent on $\chi^{(2)}_{zzz}/\chi^{(2)}_{zxx}$ with slope and constant offset determined solely by the SiC index of refraction (∼2.55) and the angle of incidence ϕ. Measurements of the TM/TE ratio at $\phi = 40$ degrees yielded a value of $\chi^{(2)}_{zzz}/\chi^{(2)}_{zxx}$ equal to -2.1 with a 95% confidence interval of ± 2.5. Table.I shows a good agreement between the measured value of $\chi^{(2)}_{zzz}/\chi^{(2)}_{zxx}$ and the theoretical values to date.

		$\chi^{(2)}_{zzz}/\chi^{(2)}_{zxx}$, 6H-SiC
Theory	Wu *et al.* [16]	-1.89
	Adolph *et al.* [17]	-1.85
	Rashkeev *et al.* [18]	-1.84
	Chen *et al.* [19]	-1.84
Experiment	Lundquist *et al.* [9]	-10
	Niedermeier *et al.* [13]	-6
	This work	-2.1 ± 2.5

TABLE I: Summary of calculated (Wu *et al.* [16], Adolph *et al.* [17], Rashkeev *et al.* [18], Chen *et al.* [19]) and measured (Lundquist *et al.* [9], Niedermeier *et al.* [13]) values of the ratio $\chi^{(2)}_{zzz}/\chi^{(2)}_{zxx}$.

For THz emission via nonlinear optical rectification, no sharp features in the spectrum are expected. The dip in the spectrum observed around 1.9 THz (see Figure 2) is attributed to absorption by impurity states within the bandgap. These impurity states, due to the presence of Boron [10], Nitrogen [11], and Vanadium [12] in compensated 6H-SiC, are known to exist with energy level spacings in the THz frequency range [20]. Radiative transitions in these states have also been demonstrated in the THz range [21].

In conclusion, we have demonstrated broadband coherent THz emission from SiC. The dependence of the THz field amplitude on the pump polarization, the pump angle of incidence, and the pump power indicates optical rectification as the main THz emission mechanism. THz emission via optical rectification has been well studied in zinc blende crystals with cubic symmetries, such as GaAs and ZnTe [14, 22], as well as in crystals of trigonal ($LiNbO_3$ [23]) and D_{3h} hexagonal (GaSe [24]) symmetries. Among these alternatives, ZnTe stands out as being particularly well phase-matched for THz generation [23]. However, its surface is known to burn under strong optical excitation and it also suffers from two-photon absorption because of its small direct bandgap (∼2.25 eV). GaSe has a high optical damage threshold [25], but the crystal is soft and fragile and its direct bandgap is also small (∼2.12 eV). Given SiC's higher optical damage threshold [8], comparable second order nonlinear susceptibility, robust mechanical properties, and large direct bandgap (>5 eV for 6H-SiC), it could prove to be a useful source of broadband THz radiation. However, absorption by impurity states in SiC would need to be reduced or eliminated.

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