Emission of terahertz radiation from SiC

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We report the emission of strong coherent broadband terahertz radiation from 6*H*-silicon-carbide (SiC) excited with optical pulses. The measured terahertz spectral signal-to-noise ratio is better than one thousand. We determine that the terahertz radiation is generated via second order optical nonlinearity (optical rectification). We present a measurement of the ratio of nonlinear susceptibility tensor elements $\chi^{(2)}_{zzz}/\chi^{(2)}_{zxx}$ and the complex index of refraction of silicon carbide at terahertz frequencies. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3194152]

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] Recently, SiC has also been explored for terahertz applications. Electrically pumped terahertz emitters based on electronic transitions between impurity states have been demonstrated.[3] SiC devices, such as IMPATT oscillators, are also being explored for high power applications in the low terahertz region.[4]

In this letter, we present results on the emission of coherent terahertz radiation from semi-insulating 6*H*-SiC excited with near-IR femtosecond optical pulses. Broadband terahertz generation and detection in semiconductors with femtosecond 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 terahertz 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*H*-SiC has a large spontaneous polarization and, therefore, a large permanent bulk electric field.[6] However, the large indirect bandgap of SiC [>3 eV (Refs. 7 and 8)] implies that free-carrier generation via direct interband absorption is not possible for ultrafast pump lasers operating around 800 nm. Free-carrier generation through two-photon or defect absorption is possible. A nonlinear mechanism, such as optical rectification,[5] can also be responsible for the generation of terahertz radiation. The nonlinear optical properties of various SiC polytypes have been previously studied, and 6*H*-SiC is known to have a large second-order nonlinear susceptibility[8,9] comparable to crystals such as lithium niobate ($\chi^{(2)}_{zzz}\sim-60$ pm/V)[10] and ZnTe ($\chi^{(2)}_{xyz}\sim 90$ pm/V).[11] In this paper, we study the terahertz 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 terahertz emission. Given its material hardness, high optical damage threshold, small optical losses, and high optical nonlinearity, SiC is promising for generating broadband high power terahertz radiation.

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 6*H*-SiC (0001) wafer pieces, 380 μm thick, were mounted vertically 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 terahertz 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 1 mm thick (110)-ZnTe crystal.[5,12] Strong terahertz pulses with maximum spectral power signal-to-noise ratios better than one thousand and detection-limited bandwidths wider than 3 THz were observed. We used a 1 s lock-in time constant. A representative electric field transient and its accompanying spectrum are shown in Fig. 1. The dip in the spectrum around 1.9 THz is discussed later.

Terahertz emission via the generation and subsequent motion of free-carriers generally has little 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 terahertz radiation at the crystal interfaces). In contrast, the emission of terahertz 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 nonlinear susceptibility tensor $\chi^{(2)}_{ijk}$.[5,10] 6*H*-SiC has 6*mm* hexagonal crystal symmetry and

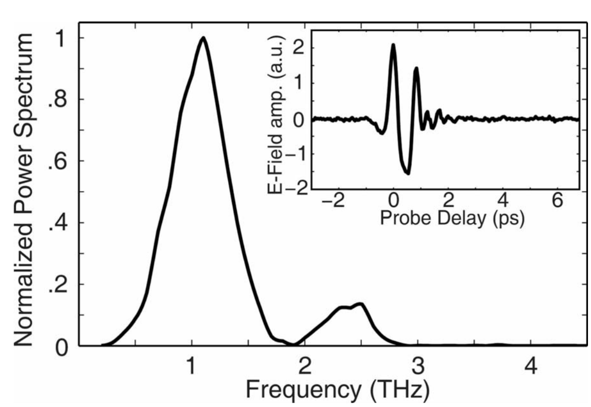

FIG. 1. Power spectrum of observed electric field. Inset: Observed transient electric field amplitude.

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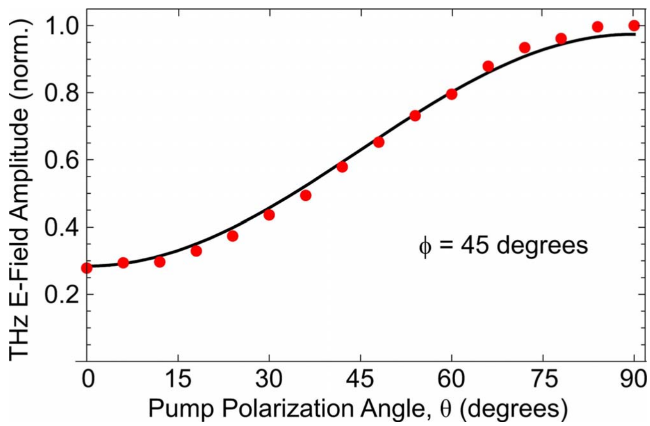

FIG. 2. (Color online) Normalized peak-to-peak electric field versus excitation polarization: 0 degrees corresponds to TE polarization, 90° to TM. Fit with $\chi^{(2)}_{zzz}/\chi^{(2)}_{zxx}=-3.0$.

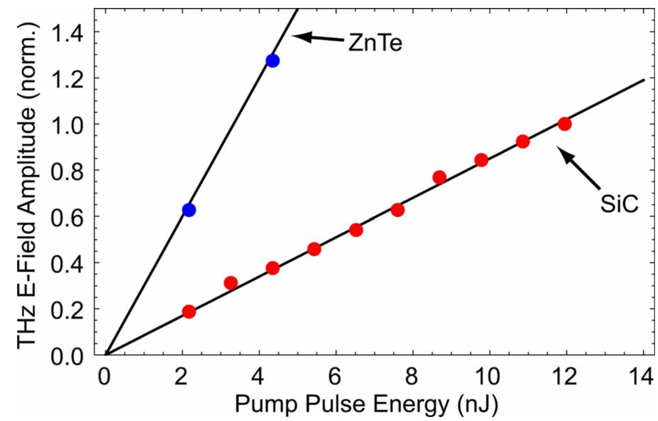

FIG. 4. (Color online) Normalized peak-to-peak electric field versus excitation pulse energy (at 780 nm) for SiC (ϕ=45°) and (110)-ZnTe (normal incidence).

therefore, under Kleinman's condition, there are only two independent components of $\chi^{(2)}_{ijk}$, written[10]

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

Figure 2 shows experimental results for the dependence of the terahertz E-field amplitude on the pump polarization while the pump angle of incidence is 45 degrees. The corresponding theoretical curve is also shown, calculated following the analysis of Chen *et al.*,[12] including the dependence of the detection on the terahertz polarization. The E-field amplitude shows a strong dependence on the pump polarization and there is 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 −3.0 was used to obtain the fit in Fig. 2.

To further confirm the optical rectification process, we study the terahertz E-field amplitude dependence on the pump angle of incidence. The experimental results for TE and TM pump polarizations are shown in Fig. 3 along with the theoretical curves. The theory fits the data well. As expected from the form of $\chi^{(2)}_{ijk}$, E_{THz} approaches zero at small ϕ. Discrepancies at extreme angles of incidence are attributed to non-ideal phase matching and reduced collection efficiencies (discussed later).

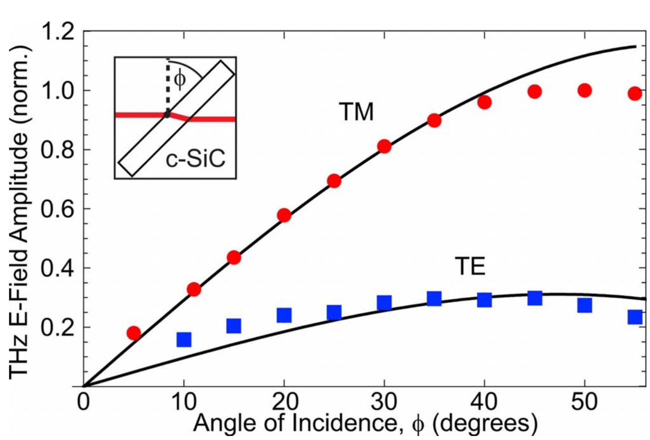

FIG. 3. (Color online) Normalized peak-to-peak electric field vs angle of incidence for TE (filled square) and TM (filled circle) pump polarizations.

For a second order nonlinear emission process, the terahertz field amplitude is expected to be proportional to the pump power. In Fig. 4, we plot the maximum terahertz pulse amplitude versus the pump pulse energy. The observed linear dependence agrees with terahertz generation via optical rectification. For comparison, we plot the amplitude of terahertz emission from a 1 mm thick (110) ZnTe crystal at normal incidence, adjusted for crystal length.

The results presented here indicate that the mechanism responsible for terahertz emission in SiC is nonlinear optical rectification. The ratio of $\chi^{(2)}_{zzz}/\chi^{(2)}_{zxx}$, the fitting parameter used in Figs. 2 and 3, has been the subject of several investigations (see Table I). The theoretically predicted value of this ratio is exactly −2 for cubic 3C-SiC and varies between −0.5 and −2.0 for *n*H-SiC (with $2\leq n\leq\infty$). To date, the measured values of this ratio have been found to be much larger than the predicted values. Our measurements of the TM/TE ratio at ϕ=40° yielded a value of $\chi^{(2)}_{zzz}/\chi^{(2)}_{zxx}$ equal to −3.0 with a 95% confidence interval of ±2.6. Table. I shows that our measured value of $\chi^{(2)}_{zzz}/\chi^{(2)}_{zxx}$ is the closest to the theoretical values among reported results for 6*H*-SiC.

To study the efficacy of terahertz emission, we present measurements of the complex index of refraction of SiC at terahertz frequencies (Fig. 5). These measurements were obtained by transmitting broadband terahertz pulses generated with a photoconductive antenna through our samples. We see that SiC is nearly dispersionless in the low terahertz range,

TABLE I. Summary of calculated (Refs. 15–18) and measured (Refs. 8 and 9) values of the ratio $\chi^{(2)}_{zzz}/\chi^{(2)}_{zxx}$. Tensor elements $\chi^{(2)}_{zzz}$ and $\chi^{(2)}_{zxx}$ (in units of pm/V) are included for reference. The measured value from this work is in close agreement with theoretical values to date.

6*H*-SiC		$\chi^{(2)}_{zzz}/\chi^{(2)}_{zxx}$	$\chi^{(2)}_{zzz}$	$\chi^{(2)}_{zxx}$
Theory	Wu *et al.*[a]	−1.89	38.6	−20.4
	Adolph *et al.*[b]	−1.85	18.1	−9.8
	Rashkeev *et al.*[c]	−1.84	17.8	−9.7
	Chen *et al.*[d]	−1.84	27.6	−15.0
Exp.	Lundquist *et al.*[e]	−10	86	−8.6
	Niedermeier *et al.*[f]	−6	24 ± 10	−4 ± 2
	This work	−3.0 ± 2.6		

[a]Reference 15.
[b]Reference 16.
[c]Reference 17.
[d]Reference 18.
[e]Reference 8.
[f]Reference 9.

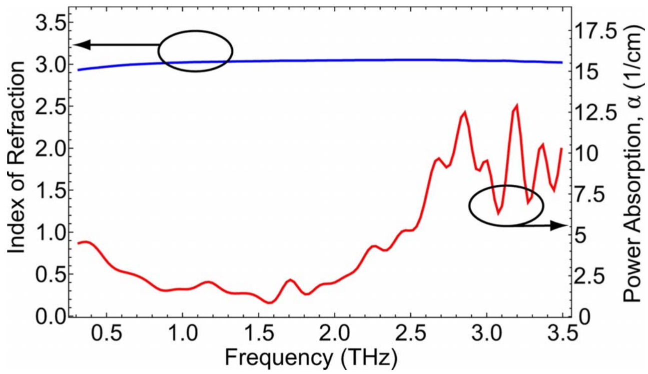

FIG. 5. (Color online) Index of refraction and power absorption of SiC vs frequency.

with an index of refraction $n=3.0$. Also, power absorption, α, is small. Since the group index of SiC at 780 nm is $n_{opt}^{gr} \approx 2.72$,[13] there is nonideal phase matching between the pump pulse and terahertz waves. The coherence length is $l_c = \pi/\Delta k \approx 0.54$ [mm THz]/f [THz] in the measured range, where $\Delta k = \omega(n_{THz} - n_{opt}^{gr})/c$ and f is the frequency.[14] The nonideal phase matching could explain the discrepancy between theory and experiment at extreme angles in Fig. 3. At large ϕ angles, total internal reflection of the emitted terahertz radiation cone becomes significant. Furthermore, since the terahertz radiation is not strictly collimated with the pump beam, a small amount of terahertz radiation is detectable even at angles near zero. The spectrum for difference frequency generation under non-ideal phase matching conditions is known to depend on the frequency.[10] Specifically, $E_{THz}(\omega) \propto \sin(\Delta k L/2)/(\Delta k L/2)$. According to this relation, the first zero in the spectrum should occur around 2 THz. Figure 1 shows that the measured terahertz spectrum is in good agreement with the predictions.

In conclusion, we have demonstrated broadband coherent terahertz emission from SiC by optical rectification. terahertz emission via optical rectification has been well studied in zinc blende crystals with cubic symmetries, such as GaAs and ZnTe,[12,19] as well as in crystals of trigonal (lithium niobate[20]) and $\bar{6}m2$ hexagonal [GaSe (Ref. 21)] symmetries. Among these alternatives, ZnTe stands out as being particularly well phase-matched for terahertz generation.[14] Given SiC's higher optical damage threshold,[7] comparable second order nonlinear susceptibility, robust mechanical properties, and large direct bandgap (>5 eV for 6*H*-SiC), it could prove to be a useful source of broadband terahertz radiation. Phase matching for high-efficiency terahertz emission has been demonstrated with a tilted optical pulse front of ~64° in lithium niobate.[20] Due to its low terahertz absorption and dispersionless index of refraction in the 0.5–3.5 THz range, SiC may be well-suited for this application. Since n_{THz} and n_{opt}^{gr} are relatively close in SiC, phase matching can be achieved with the comparatively small tilted pulse front of ~25° for a 780 nm pump.[20]

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