Optical Properties of 6H Silicon Carbide Around 10.6µm and 9.4µm Via Carbon Dioxide Laser Ablation

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Abstract

This research focuses on the measured reflection and transmission characteristics of the 6H polytype of silicon carbide through a range of wavelengths for the R-branch around 10.6 micrometers and the P-branch around 9.4 micrometers using a carbon dioxide laser. Quantum selection rules dictate the available output lines relating to the rotational energy transitions generating the obtainable output spectra. These lines delimit the span of accessible output power levels, each relating to a distinctive percentage of energy absorption and therefore a distinctive temperature of the silicon carbide wafer. The aim of this research is to find the optimal wavelength(s) for maximum heat flux absorbed by the wafer capable of generating a target temperature for carbon dioxide laser induced heteroepitaxial synthesis of graphene on silicon carbide.

1. Introduction

Carbon dioxide (CO₂) laser ablation of 6H silicon carbide (SiC) wafers in an argon gas environment has successfully produced monolayer graphene¹. When compared to graphene samples manufactured by other means, including the popular method of chemical vapor deposition (CVD), laser ablation merits several notable distinctions such as shortened production time, area manipulation, and growth precision¹. Due to the widespread use of silicon carbide in electrical circuit components such as transistors, increased comprehension of the production of graphene in this manner is an important consideration regarding its applications as a superconducting nanomaterial. To better understand and improve this method, it is imperative to analyze the intrinsic characteristics and output capabilities of the CO₂ laser system as well as the optical properties of 6H SiC at these infrared wavelengths.

For epitaxial growth to occur due to laser stimulation, the SiC wafer must absorb enough power to heat up significantly^{1,2}. Modeling heat transfer through the SiC is a highly complex issue involving the processes of conduction, convection, and radiation of energy specific to the experimental environment. This research focuses on the calculated percentage of output power absorbed by the SiC and its correlation to temperature rise. A carbon dioxide laser system with a blazed diffraction grating allows for access to a range of output lines of marginally different wavelengths and thus variable total output power and fractions of this power reflected, transmitted, and absorbed by the 6H silicon carbide^{3,4,5}. Due to the wide range of power levels and resulting optical characteristics of the output spectrum in relation to the SiC, it is possible to isolate several available lines that correlate not only with high power but high absorption as well. This ability to vary energy absorption and ultimately the resulting temperature of the ablated SiC by choosing optimal output lines plays a critical role in designing an environment for laser induced graphene growth.

2. Theory

Power reflection and transmission for electromagnetic waves can be expressed through the propagation of the parallel and perpendicular polarizations upon contact with an optical interface via the Fresnel equations. These relationships derive from the applications of Maxwell's equations for the continuity of the tangential components of both the electric and magnetic fields at a boundary.

For any electromagnetic wave, the associated electric and magnetic fields can be defined as

$$E_0 = vB_0 = \frac{c}{n}B_0 \tag{1}$$

where E_0 is the amplitude of the electric field, B_0 the amplitude of the coupled magnetic field, v is the wave speed, c is the speed of light in a vacuum, and n is the index of refraction for a given travel medium. The resulting Fresnel amplitude ratios of the reflected and transmitted components of the electric field in relation to the incident electric field are as follows for perpendicular polarization

$$r_{\perp} = \left(\frac{E_{0r}}{E_{0i}}\right)_{\perp} = \frac{n_i \cos \theta_i - n_t \cos \theta_t}{n_i \cos \theta_i + n_t \cos \theta_t} \tag{2}$$

$$t_{\perp} = \left(\frac{E_{0t}}{E_{0i}}\right)_{\perp} = \frac{2n_i \cos \theta_i}{n_i \cos \theta_i + n_t \cos \theta_t} \tag{3}$$

and for the parallel polarization

$$r_{\parallel} = \left(\frac{E_{0r}}{E_{0i}}\right)_{\parallel} = \frac{n_i \cos \theta_t - n_t \cos \theta_i}{n_i \cos \theta_t + n_t \cos \theta_i} \tag{4}$$

$$t_{\parallel} = \left(\frac{E_{0t}}{E_{0i}}\right)_{\parallel} = \frac{2n_i \cos \theta_i}{n_i \cos \theta_t + n_t \cos \theta_i} \tag{5}$$

These amplitude ratios can be used to produce the reflection and transmission coefficients, *R* and *T*, which define the fractions of an incident electromagnetic wave reflected by and transmitted through a given interface for both perpendicular and parallel polarizations.

$$R_{\perp} = r_{\perp}^{2} = \left(\frac{n_{i}\cos\theta_{i} - n_{t}\cos\theta_{t}}{n_{i}\cos\theta_{i} + n_{t}\cos\theta_{t}}\right)^{2} = \left(\frac{n_{i}\cos\theta_{i} - n_{t}\sqrt{1 - \left(\frac{n_{i}}{n_{t}}\sin\theta_{i}\right)^{2}}}{n_{i}\cos\theta_{i} + n_{t}\sqrt{1 - \left(\frac{n_{i}}{n_{t}}\sin\theta_{i}\right)^{2}}}\right)^{2}$$
(6)

$$T_{\perp} = \left(\frac{n_{t} \cos \theta_{t}}{n_{i} \cos \theta_{i}}\right) t_{\perp}^{2} = \frac{4n_{i} n_{t} \cos \theta_{i} \cos \theta_{t}}{\left(n_{i} \cos \theta_{i} + n_{t} \cos \theta_{i}\right)^{2}} = \frac{4n_{i} n_{t} \cos \theta_{i} \sqrt{1 - \left(\frac{n_{i}}{n_{t}} \sin \theta_{i}\right)^{2}}}{\left(n_{i} \cos \theta_{i} + n_{t} \sqrt{1 - \left(\frac{n_{i}}{n_{t}} \sin \theta_{i}\right)^{2}}\right)^{2}}$$

$$(7)$$

And for the parallel polarization,

$$R_{\parallel} = r_{\parallel}^2 = \left(\frac{n_i \cos \theta_t - n_t \cos \theta_i}{n_i \cos \theta_t + n_t \cos \theta_i}\right)^2 = \left(\frac{n_i \sqrt{1 - \left(\frac{n_i}{n_t} \sin \theta_i\right)^2} - n_t \cos \theta_i}{n_i \sqrt{1 - \left(\frac{n_i}{n_t} \sin \theta_i\right)^2} + n_t \cos \theta_i}\right)^2$$
(8)

$$T_{\parallel} = \left(\frac{n_t \cos \theta_t}{n_i \cos \theta_i}\right) t_{\parallel}^2 = \frac{4n_i n_t \cos \theta_i \cos \theta_t}{\left(n_i \cos \theta_t + n_t \cos \theta_i\right)^2} = \frac{4n_i n_t \cos \theta_i \sqrt{1 - \left(\frac{n_i}{n_t} \sin \theta_i\right)^2}}{\left(\frac{n_i \sqrt{1 - \left(\frac{n_i}{n_t} \sin \theta_i\right)^2} + n_t \cos \theta_i}\right)^2}$$
(9)

Where we have that

$$R_{\perp} + T_{\perp} = 1 \tag{10}$$

and

$$R_{\parallel} + T_{\parallel} = 1 \tag{11}$$

for an arbitrary angle of incidence, θ_i .

Regarding the total energy reflected and transmitted, R and T, we get

$$R = \sqrt{R_{\perp}^2 + R_{\parallel}^2} \tag{12}$$

and

$$T = \sqrt{T_{\perp}^2 + T_{\parallel}^2} \tag{13}$$

Any energy from the electromagnetic wave that is not reflected by or transmitted through the material must be absorbed by it, so that

$$R + T + A = 1 \tag{14}$$

where A is the fraction absorbed.

It should be noted that, for any slab of material, the reflection coefficient must take into account the energy reflected upon first interaction with an optical interface as well as the reflection that occurs upon the second interface within the material. For completeness, it can be said that this process takes place within the material repeatedly until the remainder of the internally reflected energy is absorbed by the material. In this experiment, a power meter was used to measure overall reflected power. Both reflected beams exit the material with only a small difference in direction and can be measured effectively if the power meter is close enough to the interface to capture them simultaneously.

The respective fractions of power absorbed by the silicon carbide wafer correspond directly to a rise in its temperature. It has been determined from previous research that the epitaxial growth of graphene occurs on the SiC wafer when temperatures are high enough to cause the sublimation of the silicon atoms, which triggers a change in the bonds between the carbon atoms on the surface and the formation of a graphene sheet. The exact temperature range(s) where this process occurs are not yet published in detail; however,

sources have recorded temperatures around 1500° C¹. In this way, maximizing both the power of the laser used for ablation as well as the absorption by the material is crucial to the progression of this method of graphene production. This research aims to isolate several choice CO_2 laser output lines that correlate to maximum output power and maximum power absorption by silicon carbide.

3. Experimental Procedure

The CO₂ beam was split upon exiting the cavity into a power meter as well as an IR spectrum analyzer to monitor the particular output line for each set of measurements. Each available line was located and maximized to its highest recorded power level using X-Y axes adjustments for both the blazed diffraction grating and the output coupler to maximize the optical cavity alignment, along with adjustments to the cavity length via a piezoelectric transducer (PZT). This power level was recorded as a reference of a line's total power for each sample of measured power both reflected by and transmitted through the wafer.

The SiC wafer was secured in a holder and was stepped into and out of the beam using a stepper motor that allowed for exact and accurate placement of the SiC wafer for each power measurement. It was observed that the entirety of the beam was contained by the wafer to minimize the chances of transmitted power readings including areas of the beam not in contact with the SiC. The wafer was oriented close to normal incidence, however, angled slightly as to direct the reflected beam into a second power meter behind the beam splitter for the reflected power reading. For each output line, a total power measurement was recorded. The wafer was then stepped into the path of the beam and data for power reflected and transmitted was documented (Fig. 1).

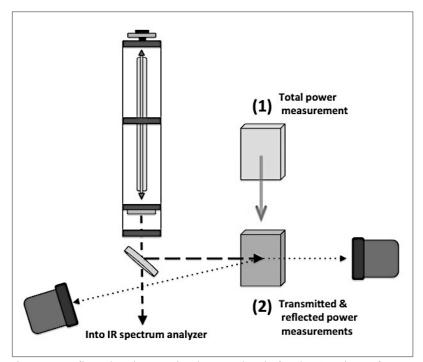


Figure 1. Reflected and transmitted power levels for the 6H SiC wafer are measured per output line. (1) Total power measurements are recorded with the wafer outside of the beam path. (2) Reflected and transmitted measurements are recorded after the wafer is stepped into the beam.

After all sets of measurements were made for both the R and P branches around 10.6µm and 9.4µm respectively, recorded values of the total, reflected, and transmitted power levels were used to estimate the percentage of power absorbed by the SiC in each instance. Using this data, the highest power lines with the largest absorption in the available output spectrum could be identified.

4. Data and Results

Total, reflected, and transmitted power data was collected for each line accessible by adjustments to the present diffraction grating. The fraction of power absorbed by the SiC was calculated as

Fraction absorbed,
$$A = 1 - (T + R)$$
 (14)

where T and R represent the measured fractions of power transmitted through and reflected by the SiC wafer, respectively. Listed associated wavelengths for these output lines were recorded from the CRC Handbook of Laser Science and Technology⁶.

Table 1. Measured optical properties of 6H silicon carbide at wavelengths associated with the R-Branch around 10.6 microns.

R-Branch Output Line	Associated Wavelength (micrometers)	Total Power (Watts, measured)	Reflected Power (W, measured)	Transmitted Power (W, measured)	Fraction Reflected R	Fraction Transmitted T	Fraction Absorbed A
R-6	10.34928	0.43	0.22	0.02	0.51	0.04	0.44
R-8	10.3337	0.79	0.29	0.04	0.37	0.05	0.58
R-10	10.31843	1.24	0.33	0.04	0.27	0.03	0.70
R-12	10.30347	0.39	0.14	0.05	0.36	0.13	0.51
R-14	10.2888	1.09	0.23	0.06	0.21	0.05	0.74
R-16	10.27455	1.33	0.27	0.04	0.20	0.03	0.76
R-18	10.26039	0.58	0.15	0.05	0.25	0.09	0.66
R-20	10.24663	0.79	0.11	0.06	0.13	0.08	0.79
R-22	10.23317	1.38	0.14	0.06	0.10	0.05	0.85
R-24	10.22001	1.00	0.11	0.02	0.11	0.02	0.87
R-26	10.20715	0.56	0.07	0.02	0.13	0.03	0.84
R-28	10.19458	0.29	0.04	0.03	0.13	0.09	0.78
R-30	10.18231	0.16	0.02	0.01	0.09	0.06	0.84
R-32	10.17033	0.38	0.02	0.00	0.05	0.00	0.95

Table 2. Measured optical properties of 6H silicon carbide at wavelengths associated with the P-Branch around 9.4 microns.

P-Branch Output	Associated Wavelength	Total Power (Watts,	Reflected Power (W,	Transmitted Power (W,	Fraction Reflected	Fraction Transmitted	Fraction Absorbed
Line	(micrometers)	measured)	measured)	measured)	R	T	A
P-6	9.44333	0.15	0.10	0.01	0.64	0.08	0.28
P-8	9.45805	0.48	0.34	0.01	0.71	0.02	0.27
P-10	9.47306	0.31	0.27	0.03	0.88	0.10	0.02
P-12	9.48835	0.79	0.60	0.02	0.75	0.03	0.22
P-14	9.50394	0.81	0.55	0.03	0.67	0.03	0.30
P-16	9.51981	0.38	0.32	0.07	0.83	0.17	0.00
P-18	9.53597	1.01	0.63	0.05	0.62	0.05	0.33
P-20	9.55243	0.81	0.44	0.04	0.54	0.05	0.42
P-22	9.56918	0.47	0.38	0.04	0.80	0.08	0.12
P-24	9.58623	0.45	0.27	0.05	0.58	0.11	0.31
P-26	9.60357	0.97	0.57	0.04	0.59	0.04	0.37
P-28	9.62122	1.01	0.57	0.03	0.56	0.03	0.41

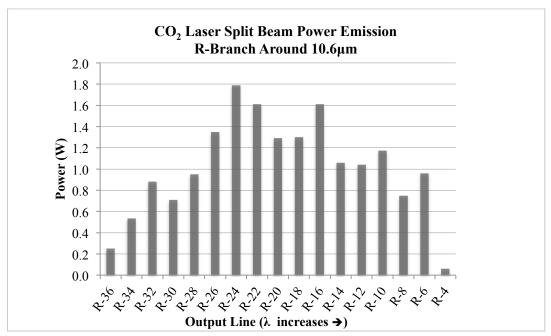


Figure 2. Total split beam power readings for each available output line of the R-branch.

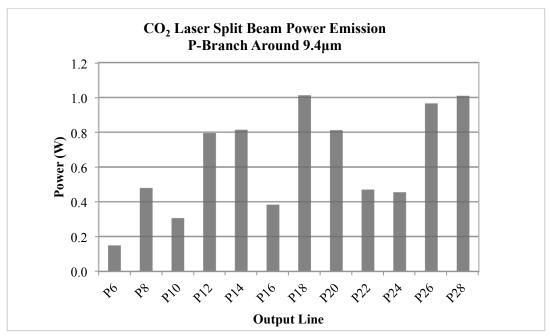


Figure 3. Total split beam power readings for each available output line of the P-branch.

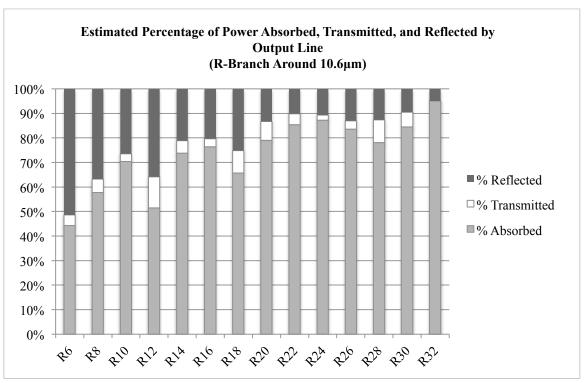


Figure 4. Estimated power absorption percentages for each available output line of the R-branch were calculated based on the reflection and transmission values.

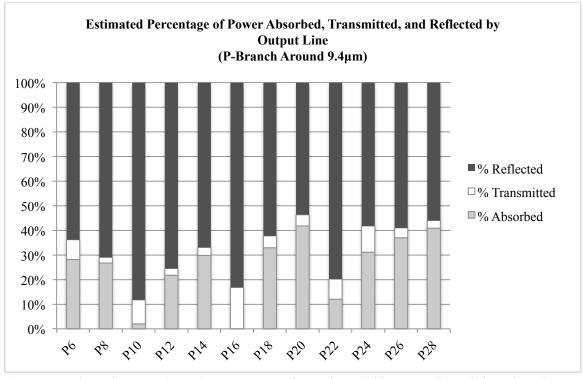


Figure 5. Estimated power absorption percentages for each available output line of the P-branch were calculated based on the reflection and transmission values.

5. Data Analysis

The collected data on the optical properties of 6H silicon carbide for the available wavelengths in the spectrum establishes that there are in fact preferable wavelengths within the output spectrum for use in epitaxial graphene growth through ablation. The R-Branch around 10.6 microns demonstrated both larger total output power levels per line as well as higher absorption percentages overall when compared to the P-Branch about 9.4 microns (Fig. 1, 2, 4, 5). Within the R-Branch, it was determined that the particular wavelengths associated with lines R-16, R-22, and R-24 had the greatest combinations of total power and fraction of power absorbed by the SiC. These are the lines that display the utmost potential to induce high temperatures within the material and that will be used for our ongoing research regarding the production of graphene.

6. Conclusion and Future Research

We have determined that R-16, R-22, and R-24 in the CO_2 laser output spectrum around 10.6 micrometers are the preferable choices for stimulating the epitaxial growth of graphene on 6H silicon carbide. These lines represent the maximum power output wavelengths of the laser system with high power absorption in the SiC needed to raise the temperature to appropriate levels. We have proceeded to rebuild the laser with this same diffraction grating to boost the overall output power and are currently researching the possibility of an ideal ablation temperature range for monolayer epitaxial graphene growth. The system is now set to lase on R-16, the current maximum power line that we understand to have high absorption rates for silicon carbide.

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8. References

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