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A diffraction-based technique for determination of interband absorption coefficients in bulk 3C-, 4H-, and 6H-SiC crystals

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Abstract

Knowledge of an absorption coefficient values for wavelengths above the bandgap and the injected carrier density profile is an important issue for analysis of carrier dynamics in highly excited semiconductors, e.g. for evaluation of the carrier density in photoexcited layer, density-dependent recombination rate, and diffusivity. In this work we present a novel way for determining the interband absorption coefficient α for SiC crystals in a wide temperature range. The proposed method is based on recording of a transient free carrier grating in a bulk semiconductor by strongly absorbed light and measurements of a probe beam diffraction efficiencies on the grating for Bragg and symmetric anti-Bragg directions. The method was applied for 3C-, 6H-, 4H-SiC polytypes at 351 nm wavelength and revealed 3 to 10-fold increase of the interband absorption coefficients in 80-800 K temperature range. Increase of absorption coefficients with temperature was simulated by bandgap shrinkage and increase of phonon density. A good agreement of the determined α values with *a priori* known room-temperature data verified validation of this technique.

1. Introduction

Progress in epitaxial growth technologies of SiC has expanded its applications for power devices, such as high-frequency power controllers and bipolar switching devices, operating under varying high injection conditions. Therefore, an appropriate attention must be paid to the peculiarities of recombination and transport processes at low and high carrier densities.

A deeper understanding of these processes in high-density carrier plasma can be obtained by applying time-resolved optical techniques, which allow the injection of an excess carriers by a short laser pulse and subsequent monitoring of the recombination and diffusion processes [1-7]. Therefore knowledge of absorption coefficient at commonly used laser wavelengths is an important issue for more precise determination of injected carrier density profile and related plasma parameters in SiC, e.g. nonlinear and surface recombination rates, carrier diffusivity and band gap renormalization [1,2,8-10].

In this work we present a novel way for determination of absorption coefficient α in a wide temperature range. The proposed method is based on recording of a transient free carrier grating by strongly absorbed UV light ($h\nu > E_g$) and measurements of the probe beam diffraction efficiencies on the grating for two symmetric directions: the Bragg (BR) and anti-Bragg (ABR) one. The method was applied in 80 - 800 K range for three commonly used SiC polytypes which are commercially available as bulk wafers. The determined absorption coefficient values were found in good agreement with the known data at room temperature [11], thus confirming applicability of the proposed technique.

2. Samples and experimental techniques

2.1. The light induced transient grating technique

A standard experimental setup was used for recording light-induced transient gratings (LITG) in SiC [12]. The experimental setup is provided on figure 1. The spatially modulated structure of non-equilibrium carriers was generated in a semiconductor by two coherent ~5.6 ps duration picosecond pulses, using the third harmonic ($\lambda_3 = 351$ nm) of a Nd:Yttrium Lithium Fluoride (Nd:YLF) laser. The grating recording beam passed a diffractive optical element - a permanent diffraction grating with a fixed period, and the two first order diffracted beams were selected by a spatial mask. These beams passed an optical telescope

with two lenses of f_1 and f_2 focal lengths and created refractive index modulation in the sample, called a transient grating. The diffraction from this grating was monitored by an optically delayed probe beam at 1053 nm. The energies of excitation, transmitted and diffracted beams were measured by Si photodetectors. The excitation energy was varied using an attenuator. The reflection from a quartz plate provided the excitation energy calibration. For the background scattered light measurements at photodetectors, an electromechanical shutter was used. The diameters of excitation and probe beams were 650 μm and 180 μm , respectively.

The grating recording beams intersected at angle Θ in a sample plane, creating an interference pattern with a spatial period Λ :

$$\Lambda = \lambda_3 / (2 \sin(\Theta / 2)). \quad (1)$$

The spatially in-plane modulated light intensity was used to generate nonequilibrium carriers, as described by equation (2):

$$N(x) = \alpha I_0 (1 + \cos(2\pi x / \Lambda)) / h\nu = N_0 (1 + \cos(2\pi x / \Lambda)). \quad (2)$$

Here $I_0 = (1-R)I_{\text{inc}}$ is the total excitation fluence in the sample, α , R are the absorption and reflection coefficients at λ_3 with quantum energy $h\nu$, I_{inc} is the incident excitation fluence and $N_0 = \alpha I_0 / h\nu$ is the nonequilibrium electron-hole concentration. The generated carriers induced a complex refractive index change (temporary modulation of a refractive index and an absorption coefficient), which is proportional to the generated carrier density: $\Delta\tilde{n} = \Delta n_{FC} + i\Delta k \propto N_0$. The change of the real part of refractive index was dominant at the used probe wavelength λ_1 [13] and described by the Drude-Lorentz formula [14]:

$$\Delta n_{FC} = n_{eh} N_0, \quad n_{eh} = -\frac{e^2}{2n_1 \epsilon_0 m_{eh}^* \omega^2} \frac{\omega_T^2}{\omega_T^2 - \omega^2}, \quad \frac{1}{m_{eh}^*} = \left(\frac{1}{m_e^*} + \frac{1}{m_h^*} \right), \quad (3)$$

here $\omega = 2\pi/\lambda_1$ is the probe frequency, ω_T is the frequency of the effective bandgap (direct, $E_T = 7.3$ eV for 4H- and 6H-SiC [15]), n_{eh} is the refractive index change by one electron-hole pair, m_{eh}^* is the reduced electron-hole effective mass, and n_1 is the refractive index for probe wavelength.

Figure 1. Light induced transient grating setup.

The recorded free carrier grating was probed by an optically delayed probe beam from the same laser at wavelength $\lambda_1 = 1053$ nm. The diffraction efficiency η of the probe beam primarily depends on phase modulation Φ of the beam by the grating, $\eta \propto \Phi^2 = (2\pi \Delta n d^* / \lambda_1)^2$, and as well on a parameter $Q = 2\pi \lambda_1 d^* / (n_1 \Lambda^2)$, which describes the propagation and overlapping of the diffracted beams inside the photoexcited crystal. If the excited layer is thin ($d^* = 1/\alpha$) and the distance between the grating peaks Λ is large ($\Lambda \gg d^*$), then $Q \ll 1$ and multiple symmetric diffraction orders are observed in the far field of diffraction (so called diffraction on a „thin grating“ [13]). If the excited layer thickness increases and grating period Λ decreases, then the parameter Q may become high enough, $Q > 10$, that only one dominant diffraction order is observed. The latter case corresponds to probe beam diffraction on a “thick Bragg grating”. Light diffraction in the intermediate range of $1 < Q < 10$ corresponds to a transient regime between the thin and thick gratings, and will be further called as an “intermediate grating”. Probe beam diffraction efficiency on the Bragg grating is described by equation (4) [13]:

$$\eta_{i=0} = \frac{I_D}{I_T} = \tan^2 \left(\frac{\pi \int \Delta n(z) dz}{\lambda_1 \cos(\varphi)} \right) = \tan^2 \left(\frac{\pi n_{eh} I_0 (1 - \exp(-\alpha d))}{h\nu \lambda_1 \cos(\varphi)} \right). \quad (4)$$

Assuming strong interband absorption $\alpha d \gg 1$, the carrier injection depth is small and the probe beam, incident at angle φ , integrates the refractive index change over the depth z (as well it integrates the injected carrier density $N_0(z) \propto I_0(z)/h\nu$).

As we consider light diffraction on a transient free carrier grating, recorded in a relatively thin surface layer d^* , its parameters are changing with time. Firstly, the thickness d^* may increase due to carrier diffusion to the depth, therefore evolution of the grating in-depth profile $N(z)$ must be taken into account. In addition, grating modulation $\Delta n(x,t) \propto N(x,t)$ decays due to carrier recombination and in-plane diffusion. A solution of the continuity equation [6,13] provides the transient grating dynamics $N(x,z,t)$, seen in decay of its diffraction efficiency:

$$N(x, z, t) = N_0 \left(1 + \cos\left(\frac{2\pi x}{\Lambda}\right) \exp\left(-\frac{4\pi^2 D t}{\Lambda^2}\right) \right) \exp\left(-\frac{t}{\tau_R}\right) \exp(-\alpha z), \quad (5)$$

$$\eta(t) = \eta_{t=0} \exp(-2t/\tau_G), \quad \frac{1}{\tau_G} = \frac{1}{\tau_R} + \frac{1}{\tau_D}, \quad \frac{1}{\tau_D} = \frac{4\pi^2 D}{\Lambda^2}.$$

Here τ_G is the grating decay time, τ_R is the carrier recombination lifetime, τ_D is the grating diffusive erase time due to ambipolar diffusion with coefficient D . As the modulated carrier density N decays due to carrier recombination and diffusion, and the thickness d^* increases with time, the diffraction efficiency must be measured shortly after excitation pulse until the grating temporal and spatial profile retains the features of photoexcitation (i.e. at $t < \tau_R, \tau_D$; then $d = d^*$).

Assuming different α values for SiC polytypes at used 351 nm wavelength ($\alpha = 290 \text{ cm}^{-1}$ for 4H-, 2260 cm^{-1} for 3C-, and 1060 cm^{-1} for 6H-SiC [11]), the light penetration depth d^* and the chosen period $\Lambda = 2.85 \text{ }\mu\text{m}$ resulted in $Q = 10.84, 1.39, 2.97$ values, correspondingly. Light diffraction features for these „thick“ or „intermediate“ surface gratings are given in part 2.3.

2.2. Samples

We investigated low doped 3C- ($n_0 \sim 10^{16} \text{ cm}^{-3}$) and high resistivity 4H- and 6H-SiC samples of thickness $d = 260 \text{ }\mu\text{m}, 360 \text{ }\mu\text{m}$ and $400 \text{ }\mu\text{m}$, respectively. At very low intrinsic carrier density, the spectral and temperature dependences of absorption coefficient can be described solely by phonon assisted light absorption processes. The samples were mechanically polished to optical quality on both sides in order to minimize the scattering losses of the probe beam and render high signal to noise ratio. The nitrogen cryostat was used for varying the sample temperature in the 80-800 K range.

The used experimental conditions ensured the grating decay time τ_G above 100 ps, i.e longer than the picosecond probe pulse duration. The τ_G values were estimated on basis of the measured carrier lifetimes τ_R in 0.5 ns – 1 μs range [2,16] and diffusive decay times $\tau_D > 230 \text{ ps}$ for used period $\Lambda = 2.85 \text{ }\mu\text{m}$ (the D values for 3C SiC varied in 1.5 – 9 cm^2/s interval with temperature [4], and similarly in the other polytypes). Therefore diffraction efficiency value if measured at the end of laser pulse ($t \approx 20 \text{ ps}$) was not affected by the grating decay and reflected the instantaneous values of Δn . Similarly, the diffusion did not increase the initial thickness of the photoexcited layer d^* . This allowed us to apply algorithms of grating diffraction efficiency on the intermediate gratings with fixed Q values for calculation of the relevant d^* and α values.

2.3. Light diffraction on intermediate gratings

Let us consider laser beam absorption in the media and deduce formulas for the first diffraction orders on the intermediate grating (i.e. for the Bragg and anti-Bragg beams). Approaching the thin grating regime, intensities of both diffraction orders become equal, while the anti-Bragg beam intensity vanishes at approaching the Bragg diffraction condition.

Figure 2. Experimental scheme (a) and formation of diffracted Bragg and anti-Bragg beams (b). The sample is positioned normally to the probe beam in order to get equal reflection coefficients for both diffracted beams.

The Bragg diffraction angle is given by $2\sin(\varphi) = \lambda_1 / \Lambda$ [13]. The angle 2φ is between the transmitted probe and the first order diffracted beams (in our case $\varphi = 3\Theta/2$ as $\lambda_1 = 3\lambda_3$). In this work diffraction on intermediate gratings will be considered, thus the first order Bragg beam I_{BR} is stronger than the symmetric anti-Bragg beam I_{ABR} (see figure 2(a)). Due to the Snell's law, the incident beams inside the crystal propagate with a lower angle $\sin(\varphi_1) = \sin(\varphi) / n_1$ (see figure 2(b)).

The Anti-Bragg beam electric vector can be obtained by integrating the electric vectors of diffracted beams on the depth elements [13]. The phase difference $\Delta\varphi(z)$ of the beams, diffracted at the interface ($z = 0$) and at the depth z varies with z , and is expressed by their path difference $\Delta l_{ABR}(z)$ (see details in figure 2(b)):

$$\Delta\varphi(z) = 2\pi n_1 \Delta l_{ABR} / \lambda_1, \quad (6)$$

$$\Delta l_{ABR}(z) = z(1 - \cos(2\varphi_1)) / \cos(3\varphi_1) \approx 2z\varphi_1^2 / (1 - 4\varphi_1^2)$$

The maximum φ angles are deduced using the limit of internal reflection for the Bragg and anti-Bragg beams as $2\varphi = \pi/2 = \arcsin(\lambda_1/\Lambda)$, which leads to $\lambda_1 = \Lambda$. If using $\Lambda = \lambda_1 = 1053$ nm, we obtain $\varphi_1 = \pi/4n_1 \approx 0.3$ rad, thus approximation by (6) leads to a negligible error of about 0.1 %. Therefore, combining the phase difference from (6) and Bragg condition $\varphi_1 \approx \lambda_1 / 2n_1\Lambda$, we get

$$\Delta\varphi(z) = \pi\lambda_1 / (n_1\Lambda^2 [1 - 4\varphi_1^2]) z = qz/2; \quad q = 2\pi\lambda_1 / (n_1\Lambda^2 [1 - 4\varphi_1^2]). \quad (7)$$

Here q is the parameter of phase shift for the Anti-Bragg beam. The phase factor for complex electric vector becomes $\exp(i\Delta\varphi(z))$, and the vector can be expressed as:

$$\vec{E} = E_0 \exp(-\alpha z/2 - i\Delta\varphi(z)). \quad (8)$$

Complex electric vector after averaging over the thickness and neglecting the probe beam intensity losses with depth due to diffraction is expressed as:

$$\vec{E} = E_0 \frac{\int_0^d \exp(-(\alpha/2 + iq/2)z) dz}{\int_0^d \exp(-\alpha z/2) dz}. \quad (9)$$

This equation was integrated analytically. For Bragg beam, $q = 0$ (all electric vectors are phase matched as phase difference is multiple to wavelength, see figure 2(b)) and the electric vector is equal to E_0 . After calculations we got:

$$\frac{I_{ABR}}{I_{BR}} = \frac{\vec{E} \cdot \vec{E}^*}{E_0^2} = \frac{1}{1+Q^2} \frac{(1 - 2\cos(qd/2)\exp(-\alpha d/2) + \exp(-\alpha d))}{(1 - \exp(-\alpha d/2))^2}. \quad (10)$$

Here $Q=q/\alpha$. At very low absorption coefficients, equation (10) leads to $I_{ABR}/I_{BR}=16\sin^2(qd/4)/(q^2d^2)$. A case of relatively weak interband absorption of the pump beam can be approximated by condition $\alpha d < 8$, then oscillating function of $\cos(qd/2)$ can be eliminated by using a slightly slanted sample across the probe beam waist (slant of $\Delta d = 4\pi/q$). For strong absorption ($\alpha d > 8$), more simple formula from (10) is obtained, assuming that sample thickness d is infinite:

$$\frac{I_{ABR}}{I_{BR}} = \frac{\vec{E} \cdot \vec{E}^*}{E_0^2} = \frac{1}{1 + \left[\frac{2\pi\lambda_1}{n_1\Lambda^2\alpha[1-4\phi_1^2]} \right]^2} = \frac{1}{1+Q^2}. \quad (11)$$

The latter approximation gives an error $\leq 5\%$ for α determination at $\alpha d > 8$ condition. In our case the $\alpha d > 8$ condition is satisfied.

The Q value, determined experimentally, enables one to determine the absorption coefficient by equation (11), as the other parameters are given by experimental conditions. In figure 3 we present dependence of the calculated diffraction efficiency ratio I_{ABR}/I_{BR} versus absorption coefficient values for two grating periods. At very strong absorption, equation (11) is approximated by $I_{ABR}/I_{BR} = 1 - q^2/\alpha^2$. The upper limit for determination of absorption coefficient is $\alpha_{\max} = 3q(\Lambda)$ and it corresponds to $I_{ABR}/I_{BR} = 0.9$. In our case, using $\Lambda = 2.85 \mu\text{m}$, $\lambda_1 = 1053 \text{ nm}$, $\alpha_{\max}$ equals to 10^4 cm^{-1} , while for $\Lambda = 1.3 \mu\text{m}$ period the $\alpha_{\max}$ value is $5 \times 10^4 \text{ cm}^{-1}$. The lower limit for absorption coefficient is obtained from $\alpha d < 8$ condition, at which the $\alpha_{\min} = 8/d \sim 200 \text{ cm}^{-1}$ can be determined for $400 \mu\text{m}$ thick sample.

Figure 3. Dependence of diffraction efficiency ratio I_{ABR}/I_{BR} on absorption coefficient for two grating periods: calculations according equation (10) – dotted lines, and with averaged oscillating function – solid lines. The dashed lines designate limits for determination of the absorption coefficient.

The calculations have shown that the proposed method allows determination of high absorption coefficient using very thick wafers ($\alpha d > 8$), when its value can not be determined by transmission measurements or ellipsometric techniques [15]. On the other hand, lower absorption coefficient ($\alpha d < 8$) can be extracted either by the proposed method or by conventional UV-VIS spectrometry [15], as well by scanning the carrier density in-depth profile [17]. The proposed method allows determination of the temperature-dependent absorption coefficient, fitted by the approximations, given in section 2.4.

2.4. Approximations of absorption coefficient and refractive index temperature and quantum energy dependences

Absorption coefficient temperature and quantum energy dependence in SiC can be approximated using temperature dependent energy gap $E_g(T)$, absorption strength A , and phonon energy E_{ph} [18]:

$$\alpha(T) = A \left[\frac{(h\nu - E_g(T) - E_{ph})^2}{1 - \exp(-E_{ph}/kT)} + \frac{(h\nu - E_g(T) + E_{ph})^2}{\exp(E_{ph}/kT) - 1} \right]. \quad (12)$$

The bandgap temperature dependence can be described by Passler formula [17]:

$$E_{gP}(T) = E_g(0) - (\varepsilon \Theta_p / 2) \left(\sqrt[p]{1 + (2T/\Theta_p)^p} - 1 \right). \quad (13)$$

Here $\varepsilon = 3 \cdot 10^{-4}$ eV/K, $\Theta_p = 450$ K and $p = 2.9$ for both 4H- and 6H-SiC. The formula is valid in 77-450 K range. The same temperature dependence of bandgap can be used for all polytypes. The temperature dependence of SiC bandgap can be also precisely fitted by vibrational energy function $E_{gD}(T)$ [19]:

$$E_{gD}(T) = E_g(0) - a_D f_D(\Theta_D/T), \quad f_D(\Theta_D/T) = 18k_B T^4 / \Theta_D^3 \int_0^{\Theta_D/T} (\exp(x) - 1)^{-1} x^3 dx. \quad (14)$$

Here a_D accounts for atomic oscillator interaction (if $a_D=1$ there is no interaction), $\Theta_D = 1000 \pm 50$ K is the Debye temperature and $E_g(0)$ is the bandgap at zero temperature, the same as in Passler formula [17].

The spectral dependence of absorption coefficient is given by [17]:

$$\alpha(T) = \frac{A_1}{\hbar\omega} [\hbar\omega - E_g - E_p]^2 + \frac{A_2}{\hbar\omega} [\hbar\omega - E_g + E_p]^2. \quad (15)$$

The given above formulas were used for approximations of bandgap and absorption coefficient temperature dependences. The $E_g(T)$ dependences were taken from literature and were approximated by E_{gP} and E_{gD} functions (13,14). The fitting curves are shown in figure 4. The Choyke [20] data seem to be deviating from Grivickas data [17] in 4H- and 6H-SiC. In 3C the deviation is 60 meV, which is the phonon energy, as the bandgap was approximated disregarding phonon absorption. In 4H and 6H the gaps reduce much faster with temperature which may be the fact of high doping [18]. Grivickas [17] data, approximated with E_{gP} , may give more consistent results as it was obtained in low doped sample.

Figure 4. The bandgap temperature dependences of SiC polytypes [20], approximated with E_{gD} (14) function. The fitting parameters are shown on the plot. The fitting by E_{gP} (13) function is shown for comparison, using $E_g(0$ K) values, shifted accordingly to the determined $E_g(300$ K) values (table 1) for relevant polytypes. Note very similar E_{gP} and E_{gD} dependencies in 3C polytype, which can be a result of lower 3C doping.

Sample absorption coefficient in the vicinity of E_g (figure 5) was calculated using relationships from [21] with photon energy dependent refractive index n and reflection coefficient R . Ordinary refractive index was used for hexagonal polytypes, as the absorption edge measurements were performed in the $E \perp c$ geometry (the c axis is almost perpendicular to the surface of the wafers). In wide spectral range it can be described as: $n^2 = 1 + 5.49 / (1 - (E(\text{eV})/E_\Gamma)^2)$ according to [15], here $E_\Gamma = 7.3$ eV is the energy of the effective direct gap. Refractive index of 3C has very similar value and spectral dependence [22]. Therefore ordinary refractive index is essentially the same for all polytypes [22] and weakly depends on temperature: $dn/dT = 3.6 \times 10^{-5} \text{ K}^{-1}$ at RT [19].

3. Experimental results and discussion

3.1. UV-VIS absorption edges in SiC at RT

The absorption edges for the polytypes were measured by UV-VIS spectrometer in the E_⊥c geometry. The data are shown in figure 5. The largest α values determined from these measurements are of about 300 cm⁻¹. It is clear that it is not possible to measure much higher α values at 351 nm in 6H- or 3C-SiC due to large sample thickness, neither determine their temperature dependences.

The spectral measurements provided values of bandgaps E_g , phonon energies E_{ph} and absorption strengths A , which were used for approximation of $\alpha(T)$ dependence (see Appendix I). The obtained absorption edge fitting parameters are given in Table 1.

Figure 5. Absorption edges (E_⊥c) of the three SiC polytypes at 293 K, determined by conventional UV-VIS spectrometer. Solid lines – experimental data, dashed lines – linear fits (15). The A_1 and A_2 are the phonon-emission and phonon-absorption governed slopes of the absorption spectra. The arrows indicate determined bandgaps at 293 K.

Table 1. Absorption coefficient approximation parameters at 293 K.

Polytype	$E_{g\text{ abs}},$ eV	$A_{1\text{ abs}},$ cm ⁻¹ /eV	$A_{2\text{ abs}},$ cm ⁻¹ /eV	$E_{ph\text{ abs}},$ meV	$A(3.53\text{ eV})$ abs, cm ⁻¹ /eV ²	A_1/A_2 abs	A_1/A_2 calc	$A_1(3.53\text{ eV})$ ref, cm ⁻¹ /eV
3C-SiC	2.30	4275	330	63	1100	13.0	10	4800
4H-SiC	3.20	12056	441	63	3100	27.3	11	12500
6H-SiC	2.94	16640	1043	60	4300	16.0	11	10700

„abs“, „calc“, „ref“ subscripts indicate values deduced from absorption edges, calculations according (12,14) and reference [11] respectively. Subscriptus nukelt

The phonon assisted optical transitions involve the lowest conduction bands of the polytypes in valleys 3C-: X, 4H-: M, 6H-: M-L [22]. Average phonon energy is in 65-70 meV range for 4H- [17,18] and 63 meV for 6H-SiC [23]. These values coincide quite well with our approximations (in table 1). It was suggested [18] that LA phonons of 76-79 meV are determining the absorption. However, it was concluded in [17] that both the acoustic and optical phonons participate in transitions, so the fitted (table 1) phonon energies stand for the average of those participating in the absorption. Our measurements provided phonon energies lower than LA, thus indicating a need to account contribution of low energy TA phonons (besides LA and optical ones). Assuming that only acoustic phonons participate in absorption in equal strength, the average phonon energy is obtained as $E_{av}=(E_{TA1}+E_{TA2}+E_{LA})/3$ and leads to the E_{av} values of 57 meV, 61 meV and 59 meV for 3C-, 4H- and 6H-SiC, respectively, using data from [22].

The shape of $\alpha(h\nu)$ dependence in different spectral ranges can be described by phonon-emission related slope A_1 (see table 1 and Appendix I), which has different value in the vicinity of the bandgap ($A_{1\text{ abs}}$, see figure 5) and highly above E_g ($A_{1\text{ ref}}$, which stands for excitation wavelength λ_3 with $h\nu = 3.53$ eV) due to multiphonon absorption, excitonic effects [17] and band nonparabolicities [24]. This feature

was strongly pronounced in 6H (table 1) and could be explained by strong conduction band nonparabolicity in 6H [24]. The determined A_1/A_2 ratio in 4H is few times larger than that calculated according to estimated phonon energy (table 1). This is probably caused by two different conduction bands with close energies (separated by ~ 100 meV) [11,24]. This may lead to A_1 value two times larger at higher energies than at lower ones. Close bandgaps (including few conduction and valence bands) are also present in 6H- (100-200 meV), while in 3C-SiC there are close in energy valence bands (separated by 10 meV $\ll E_{ph}$), which leads to one effective bandgap. Thus in 3C polytype the fitting of experimental data was found the best, showing only excitonic absorption impact near the bandgap. Under these assumptions, the approximation of absorption coefficient by equations (12,15) reasonably well described the absorption edges for all polytypes.

3.2. Temperature dependences of interband absorption coefficients at 351 nm

The dependencies of Bragg diffraction efficiencies on $\Lambda = 2.85$ μm grating were measured at various excitation fluences, as shown in figure 6. The quadratic increase of diffraction efficiency with excitation was used for extraction of n_{eh} values for the SiC polytypes, which are given on the plot (n_{eh} error is about $0.2 \times 10^{-22} \text{ cm}^3$). The experimental values were found very similar to calculated ones by equation (3) ($n_{eh} = (8.4; 8.8; 7.3) \times 10^{-22} \text{ cm}^3$ for 3C, 4H and 6H respectively, using reduced optical masses $m_{eh} = 0.24; 0.23; 0.27$ [24]).

Figure 6. Dependence of Bragg diffraction efficiency on excitation fluence. γ are the slopes of the curves in the log-log plot. The $\gamma \sim 2.0$ values verify theoretical approximation by formula (4).

The measured ratios of I_{ABR}/I_{BR} diffraction efficiencies for different polytypes were used for extraction of absorption coefficient values and their temperature dependences (figure 7). The used excitation energy density for α determination were in the $0.1\text{--}2 \text{ mJ/cm}^2$ range (it corresponds to $\Delta N = 5 \times 10^{16} - 7 \times 10^{18} \text{ cm}^{-3}$ at 300 K), thus decrease of the probe beam intensity due to its diffraction was negligible (below 1%, see figure 6), and α values were calculated according to equation (11). Due to lower absorption coefficient in 4H, the α increase with temperature was found the largest, spanning over one order of magnitude. We note that the excitation induced sample heating, ΔT (due to injected carrier thermalisation) in the used injection range was negligible (it peaked up to $\Delta T < 10$ K at 80 K and < 1 K at 300 K in 3C-SiC and was much smaller for other polytypes).

The fitting of $\alpha(T)$ dependences was performed under two different simulation models, based on $E_g(T)$ dependencies (see Appendix I and table 2). Using Passler approximation (solid lines in figure 7(b)), we used phonon energies and slope A values as given in table 2. This is consistent with the average acoustic phonon energies and slope A values, obtained from the fitting of absorption edges. Using Debye approximation, higher phonon energies ($\sim 70\text{--}80$ meV) and slope A values (table 2) were used (dashed lines) with respect to those determined from the absorption edges. Thus the phonon energy of 60 meV for all studied SiC polytypes gives a reasonably good fit of the absorption edges and temperature dependences of interband absorption coefficients.

Figure 7. Temperature dependences of I_{ABR}/I_{BR} diffraction efficiency ratio (a) and the determined absorption coefficients (E_{lc}) (b). The fitting parameters are given in Table 2. Fitting by Passler and Debye approximations for $E_g(T)$ are denoted by solid and dashed curves, respectively. Comparison with data at 351 nm [11] and at 355 nm [18] are given.

Table 2. SiC absorption coefficient approximation parameters at 3.53 eV.

SiC polytype	$E_g(0)$, eV	A_{ref} , $\text{cm}^{-1}/\text{eV}^2$	A_{abs} , $\text{cm}^{-1}/\text{eV}^2$	Passler $E_g(T)$		Debye $E_g(T)$	
				A , $\text{cm}^{-1}/\text{eV}^2$	E_{ph} , meV	A , $\text{cm}^{-1}/\text{eV}^2$	E_{ph} , meV
3C-SiC	2.335	1200	1100	1250	63	1250	70
4H-SiC	3.232	3200	3100	3400	59	4300	82
6H-SiC	2.973	2700	4300	3100	63	3400	78

4. Conclusions

A new method, based on the light diffraction on transient free carrier grating, was used for the determination of interband absorption coefficient at wavelengths well above the bandgap. The absorption coefficient values in the range from 200 to 5000 cm^{-1} were measured for three bulk SiC polytypes in the 80-800 K range. The increase of indirect absorption coefficient by order of magnitude in 4H and by 3-5 times in 3C and 6H was approximated using ~ 60 meV phonon energy and the temperature dependent decrease of bandgaps. The temperature dependent interband absorption coefficients will ensure more correct analysis of the temperature and density dependent carrier recombination rates and diffusivity.

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