# Inter-conduction band electron relaxation dynamics in 6H−SiC

T. Tomita,[a] S. Saito, and T. Suemoto
*Institute for Solid State Physics, University of Tokyo, 5-1-5 Kashiwanoha, Kashiwa, Chiba 277-8581, Japan*

H. Harima
*Department of Applied Physics, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan*

S. Nakashima
*Department of Electrical and Electronic Engineering, Miyazaki University, 1-1 Gakuenkibanadai-nishi, Miyazaki 889-2192, Japan*

The ultrafast inter-conduction band carrier dynamics in 6H–SiC was observed by using pump and probe transient absorption technique. Probe wavelength dependence of the bleaching was compared with the steady-state absorption profiles for polarizations parallel and perpendicular to the $c$ axis, and these bleachings were ascribed to the decrease of electron populations in the lowest conduction band. The relaxation time from the higher to the lowest conduction band due to the inter-conduction band electron–phonon scattering is 1.25 ps. © *2001 American Institute of Physics.*
[DOI: 10.1063/1.1399304]

Recently, silicon carbide (SiC), belonging to wideband-gap semiconductors, has been receiving more and more attention, because of its possible application to high-speed and high-power devices, which can exceed the performance of today's silicon devices. The experiment in femtosecond regime gives the information about electron–phonon and electron–electron interactions which are indispensable for developing high-speed and high-mobility devices.

For GaN, which is often compared with SiC, experiments on the ultrafast electron[1,2] and hole[3] dynamics have been reported. As far as the authors know, however, the ultrafast carrier dynamics in SiC has been hardly studied. The major difference between them arises from the band structures. Observation of ultrafast carrier dynamics in indirect wideband-gap materials, such as SiC, is more difficult than that in direct ones, because the carriers cannot be produced with sufficient concentration in the lowest conduction band by photoexcitation. To obtain the information about ultrafast carrier dynamics in SiC, we propose to use the inter-conduction band transitions. Although the band-gap energy lies in the ultraviolet region, the $n$-type SiC has appreciable absorption in the visible region as reported by Biedermann.[4] These absorption bands are called "Biedermann bands," and assigned to the inter-conduction band transitions from the lowest conduction band based on a comparison with a theoretical calculation.[5]

The light source used in this experiment is a 1 kHz regenerative amplifier (Spectra Physics, Spitfire) which produces 120 fs pulses at 800 nm (1.55 eV). A part of the beam was chopped by an optical chopper, and used as a pump beam (the excitation density was about 10 mJ/cm$^2$). The other part was focused into a sapphire plate (5 mm thick) to produce the white probe beam. These two beams were directed into the sample, and the transmitted probe beam was dispersed by a single monochromator (Nikon G250) and de-

tected by a photomultiplier (Hamamatu R928) with a lock-in amplifier. To measure the polarization dependence of absorption, the polarization of the transmitted probe beam was analyzed by a polarizer. The time resolution of this system was below 300 fs, and the detection limit of transmission change was less than $10^{-3}$. The sample used in this experiment was a 6H–SiC single crystal with an electron doping level of about $1.2 \times 10^{18}$ cm$^{-3}$. The sample thickness was about 1 mm, and the face was parallel to the $c$ axis.

Measured transmission changes for various probe wavelengths are shown in Fig. 1. The polarization of the pump beam and that of the probe beam was parallel to the $c$ axis ($E_{\text{pr}} \| c$), where $E_{\text{pr}}$ is the polarization of the probe beam. The pump beam excites the electrons in the lowest conduction band to the higher conduction bands. The negative transmission peak around 0 ps delay is due to the spectral artifacts, probably due to the two photon absorption. Since these artifacts do not reflect carrier dynamics, they will not be discussed further.[6] After the disappearance of these artifacts, we observed positive transmission changes (bleaching). Except for the very early stage, the decay of this bleaching is fitted well by a single exponential curve. The obtained decay times did not have any appreciable wavelength dependence. The decay time of 1.25 ps was obtained by averaging the time constants in a whole wavelength region. The mean deviation of the time constants from the average was below 0.05 ps. We also measured the transmission changes for $E_{\text{pr}} \perp c$. The observed bleachings have almost the same decay time as for $E_{\text{pr}} \| c$ configuration, while their magnitudes are small and the wavelength dependence is totally different. The observed magnitudes of bleaching for various probe wavelengths were plotted by filled symbols in Fig. 2 for $E_{\text{pr}} \| c$ [Fig. 2(a)], and for $E_{\text{pr}} \perp c$ [Fig. 2(b)]. The absorption change depends on three quantities, matrix elements ($|M|^2$), initial electron distribution [$f(E_i)$], and the density of unoccupied final state [$D(E_f)\{1 - f(E_f)\}$]. In the following discussion, we assume that the changes of $|M|^2$ and $D(E_f)$ are small.

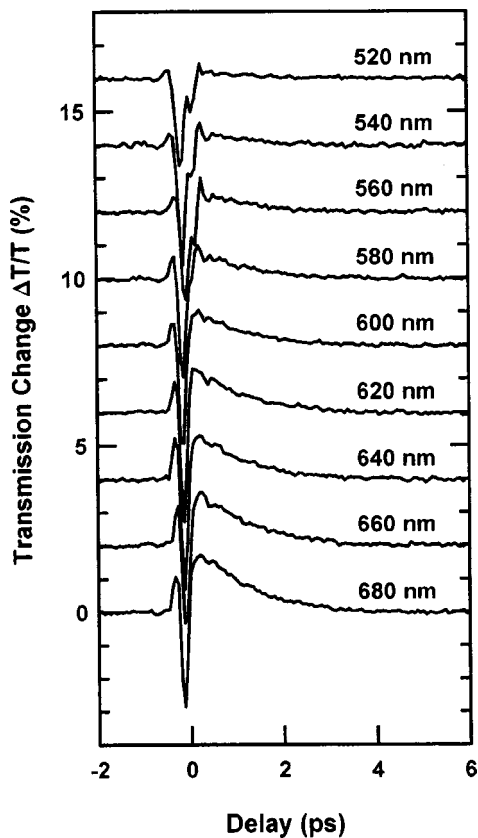[a]Electronic mail: tomita@issp.u-tokyo.ac.jp

FIG. 1. Measured transmission changes in 6H–SiC for various probe wavelengths from 520 nm (2.38 eV) to 680 nm (1.82 eV). The polarizations of pump and probe beams are parallel to the $c$ axis. The consecutive spectra are shifted by 2.0 for each along the ordinate for a convenient display.
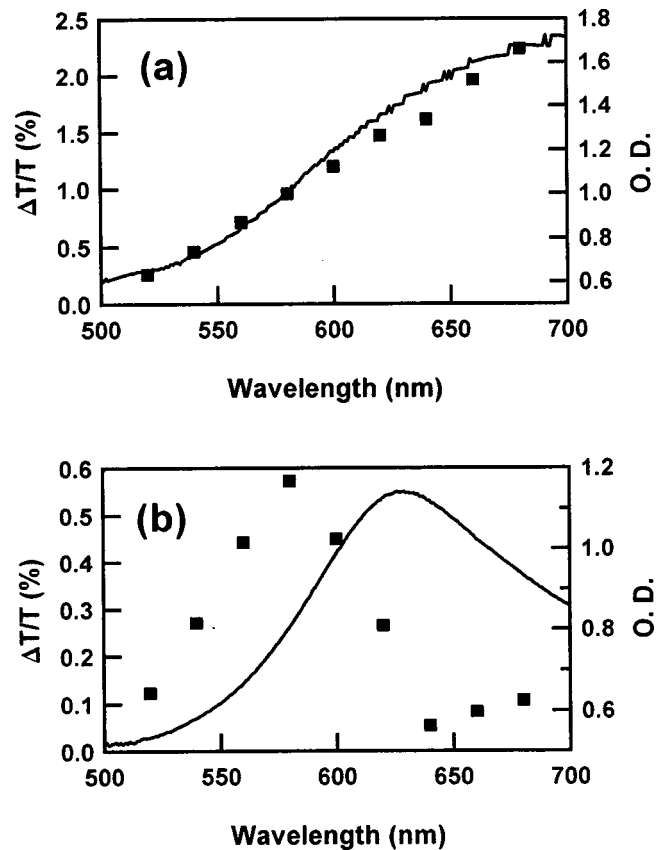


FIG. 2. The comparison between the intensity of transient transmission change (filled symbols, left scale) and the steady-state absorption band (solid line, right scale). The polarization of probe beam was parallel to the $c$ axis in (a) and perpendicular in (b).

For the $E_{pr}\|c$ configuration [Fig. 2(a)], the magnitudes of these bleachings become larger as the probe wavelength approaches longer wavelength limit. On the other hand, for $E_{pr}\perp c$ [Fig. 2(b)], the magnitude of the bleaching is several times smaller and the peak appears around 580 nm. For a comparison, the steady-state absorption spectra, corresponding to Biedermann bands, for the same sample are plotted in Fig. 2 by solid lines for each polarization configuration. The spectral shape of bleaching for $E_{pr}\|c$ is in good agreement with the steady-state absorption spectrum. From this fact, the transmission changes in the $E_{pr}\|c$ configurations are ascribed to the bleaching of the $c_1 - c_3$ absorption bands. Here, $c_1 - c_4$ denote the lower lying conduction bands as shown in Fig. 3.

On the other hand, the spectral shape of bleaching does not directly agree with the Biedermann bands for $E_{pr}\perp c$ configuration. This spectral shape is fitted well by the overlap of Biedermann bands and its derivative. This means that Biedermann bands are bleached and shifted to lower energy. These two effects are explained by the decrease of electron population in the $c_1$ band along with the anisotropy of the band shape around the lowest point in the $c_1$ band ($U$ point). In the $U-L$ direction, the slope of the $c_1$ band is steeper than that of the $U-M$ direction. Therefore, the decrease of electron population in the $c_1$ band by optical pumping leads to a movement of the center of gravity of electron population toward the $L$ point. Consequently, the absorption profile corresponding to the $c_1 - c_4$ transition shifts to lower energy, because of the negative steep slope of the $c_4$ band toward the $L$ point. In addition, the decrease of electron population

causes the bleaching of $c_1 - c_4$ absorption band. Then, the shift and bleaching is observed for the $E_{pr}\perp c$ configuration.

The fact that the observed decay time of bleaching is the same for both polarizations suggests that the relaxation behavior is dominated by a single mechanism. Thus, it is concluded that this bleaching corresponds to the decrease of
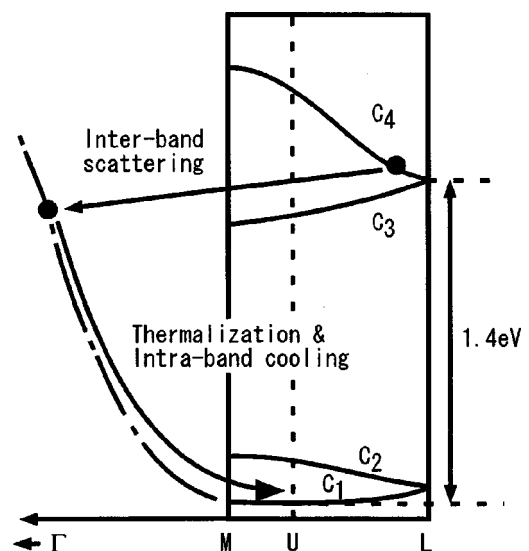


FIG. 3. Schematic band structure of 6H–SiC. In the $M-L$ direction, the band structure is plotted based on the calculated results of Ref. 5. In the $\Gamma-M$ direction, the $c_1$ band shown by a dash–dot curve is only qualitative. The decay process of excited electrons is shown by arrows.

electron population in the $c_1$ band rather than the filling of the $c_3$ and $c_4$ band.

In the following, we discuss the dynamics of electron excitation and relaxation. According to the calculations, there is no band lying around 1.55 eV (corresponding to the excitation photon energy) above the $c_1$ band at the $U$ point. The steady-state absorption, however, has appreciable absorption around 1.55 eV. This fact is understood by the breakdown of the symmetry and momentum selection rule due to the impurity effect.[5] Thus, the pump beam excites the electrons from the $c_1$ band to the $c_4$ band, which is located 1.4–2.0 eV above the $c_1$ band. The observed decay time is much faster than that of the radiative decay of the electrons from the $c_4$ band to the $c_1$ band. The transition of electrons by emitting zone center phonons also seems to be impossible, because the energy separation between the $c_4$ band and $c_1$ band is far larger than the typical phonon energies (about 0.1 eV).[7] The most probable scenario of the relaxation process is the inter-band scattering of the electrons in the $c_4$ band by emitting a large momentum phonon, followed by thermalization due to the electron–electron scattering and cooling due to the intra-band electron–phonon scattering. The excited electrons in the $c_4$ band will be scattered to the $c_1$ and $c_2$ bands in the $\Gamma$–$M$ region by emitting phonons (inter-band scattering) as shown in Fig. 3. The scattered electrons in the high energy region of the $c_1$ and $c_2$ bands will be thermalized by electron–electron interaction and then relax to the $U$ point in the $c_1$ band by emitting phonons (intra-band cooling). As the thermalization by electron–electron interaction at a carrier concentration of $10^{18}$ cm$^{-3}$ is expected to be very fast, it cannot correspond to the observed time constant 1.25 ps. If the observed decay of bleaching reflects the intra-band cooling processes, the induced absorption should be observed at the probe energies which correspond to the transitions from the $c_1$ band above Fermi level. Furthermore, the observed low energy shift of the peak in the $E_{pr} \perp c$ configuration cannot be explained, because the raise of the temperature will

shift the center of gravity of the electron population in the $c_1$ band toward $L$ point, resulting in high energy shift of the absorption peak. Thus, we assign the recovery of bleachings with a time constant of 1.25 ps to the inter-conduction band scattering time by emitting large momentum phonons. This time constant does not include the hot phonon effect, because we did not observe any appreciable change in the decay time for five times different excitation fluence.

This process can be compared with the inter-valley scattering process, which also involves the emission of large momentum phonons. The reported scattering time from $X_7$ to $\Gamma$ valley was 0.60 ps for GaAs[8] and that for $X_7$ to $X_6$ valley scattering via $\Gamma$ valley was 2 ps for GaP.[9] For ZeSe, the effective $L$–$\Gamma$ scattering time was 1.8 ps.[10] Comparing with these time constants, the observed 1.25 ps recovery time from higher conduction band to the lowest conduction band seems to be reasonable.

In conclusion, ultrafast inter-conduction band carrier dynamics in SiC was studied by using a femtosecond transient absorption spectroscopy. The polarization dependence was investigated for various probe wavelengths. The inter-conduction band relaxation time due to the inter-band scattering process in 6H–SiC, which was determined from this experiment, is $1.25 \pm 0.05$ ps.

[1] H. Ye, G. W. Wicks, and P. M. Fauchet, Appl. Phys. Lett. **74**, 711 (1999).
[2] C.-K. Sun, Y.-L. Huang, S. Keller, U. K. Mishra, and S. P. DenBarrs, Phys. Rev. B **59**, 13535 (1999).
[3] H. Ye, G. W. Wicks, and P. M. Fauchet, Appl. Phys. Lett. **77**, 1185 (2000).
[4] E. Biedermann, Solid State Commun. **3**, 343 (1965).
[5] S. Limpijumnong, W. R. L. Lambrecht, S. N. Rashkeev, and B. Segal, Phys. Rev. B **59**, 12890 (1999).
[6] J. Mørk, A. Mecozzi, and C. Hultgren, Appl. Phys. Lett. **68**, 449 (1996).
[7] T. Tomita, S. Saito, M. Baba, M. Hundhausen, T. Suemoto, and S. Nakashima, Phys. Rev. B **62**, 12896 (2000).
[8] M. A. Cavicchia and R. R. Alfano, Phys. Rev. B **48**, 5696 (1993).
[9] M. A. Cavicchia and R. R. Alfano, Phys. Rev. B **51**, 9629 (1995).
[10] D. J. Dougherty, S. B. Fleischer, E. L. Warlick, J. L. House, G. S. Petrich, L. A. Kolodziejski, and E. P. Ippen, Appl. Phys. Lett. **71**, 3144 (1997).