Time-resolved observation of carrier and coherent phonon in 4H-SiC under off-resonant excitation

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1. Introduction
Silicon carbide (SiC) is an important material for the development of high-power and high-speed electronic devices with high breakdown voltage and superior thermal conductivity. Such devices often operate at high injections of carriers, whose excess energy is dissipated through phonon emission. Therefore, a deeper understanding of carrier and phonon dynamics at high carrier concentrations is essential for designing better and faster devices.

Time-resolved optical techniques are useful for monitoring carrier and phonon dynamics under high injection of excess carriers generated with short laser pulses. In this work, we studied carrier and phonon dynamics in 4H-SiC by time-resolved reflectivity measurement with sub-20fs time resolution. We found that the n-type doping induces the generation of excited carriers through the inter-conduction band transition. The frequency and dephasing time of coherent phonons in 4H-SiC exhibit the off-resonant nature, originating from the virtual transition even in the presence of excited carriers.

2. Experiment
The light source was a sub-10-fs Ti:sapphire oscillator with a center wavelength of 780 nm. The linearly polarized pump and probe pulses were focused by a 50-mm-focal-length mirror to a 17-µm spot on the sample. The intensity of the pump laser pulses was varied from 3.9×10¹⁰ to 1.3×10¹¹ W/cm². The excited-carrier density, estimated from the absorption coefficient [1], is 3.1×10¹⁶ – 1.0×10¹⁷ /cm³.

For the detection of carrier and phonon dynamics in 4H-SiC, we performed isotropic or anisotropic reflectivity measurements depending on the anisotropy (or symmetry) of carriers and phonons [2,3]. Details of both detection schemes are described in Ref. 1 and 2. In both measurements, we measured the reflectivity of the probe beam with and without the pump beam. Then, we took the difference between them (∆R) as a function of the pump-probe delay (∆t). The differential signal was amplified by a preamplifier and recorded with a digital oscilloscope. The signal was normalized to ∆R/R, where R is the reflectivity without the pump pulse. We also performed time-integrated Raman measurements, where the excitation wavelength was 532 nm and the spectral resolution was 2 cm⁻¹.

The samples investigated in this work were semi-insulated (SI) and n-type 4H-SiC. The surface orientation was (0001). The pump beam polarization is parallel to [1120], and probe beam polarization is tilted at 45° with respect to the pump beam polarization. The doped carrier concentration (n₀) estimated from the resistivity, is >10¹⁵ /cm³ for SI and 10¹⁸-10¹⁹ /cm³ for n-type 4H-SiC [4]. The n-type SiC has absorbance in the visible region due to inter-conduction band transition, whereas SI SiC does not [1,5].

3. Result
Time-resolved isotropic reflectivities for SI and n-type SiC are shown in Figs. 1(a) and (b), respectively. In both, a sharp and intense response is observed at ∆t=0 due to the polarization grating that is formed with an overlap between the pump and probe pulses [2]. In the n-type SiC, then, an exponential component appears and decays within 20 fs [arrow in Fig. 1(b)]. We attribute this doping-induced component as the decay of carriers generated through the inter-conduction band transition according to the absorption spectrum and band structure of n-type 4H-SiC [1,5].

After the carrier responses, periodic oscillatory signals, originating from coherent phonons, are observed in both the SI and n-type SiC. The oscillation in the n-type SiC decays more rapidly than that in SI SiC. To identify effects of the doping on the phonon dynamics, we performed a Fourier transform (FT) analysis. The FT power spectra of oscillatory signals are shown in Figs. 1(c) and (d). In SI SiC, the most intense peak at 29 THz is assigned to A₁-symmetry longitudinal optic (LO) phonon according to the Raman spectrum [6]. The weak peak at 23 THz and 18 THz are assigned to folded modes of E₁-symmetry transverse optic branches (FTO) and A₁-symmetry longitudinal acoustic branches (FLA), respectively. In the n-type SiC, the doping-induced shifts are observed only in the A₁-symmetry LO mode, which exhibits an asymmetric broadened shape due to the LO-phonon-plasmon-coupled (LOPC) mode [6].

The amplitude of all phonon modes and electronic responses exhibit linear power dependence, which indicates one-photon excitation occurred under the present experimental condition. Since the one-photon energy (1.6 eV) is less than the band gap in 4H-SiC (3.26 eV), coherent
phonons in 4H-SiC are generated through the virtual transition [2]. The off-resonant nature is also reflected in the power independence of the phonon frequency and dephasing rate.

The power independence of the phonon frequency and dephasing rate for the LOPC mode is noteworthy. Generally, the LO phonon, which couples with both doped and excited carriers, exhibits the power dependence [7]. The present result indicates the absence of the coupling between the LO and excited carriers in 4H-SiC. We consider that the decay time of excited carriers (20 fs) is too short, and the excited carriers could not couple with the LO mode. The short carrier decay time results in the power independence of the LOPC even in the presence of excited carriers.

4. Summary

We performed time-resolved reflectivity measurements to investigate ultrafast carrier and phonon dynamics in 4H-SiC. With n-type doping, excited carriers are generated through the inter-conduction band transition and decay within 20 fs. We observed the $A_1$-symmetry LO mode, which exhibits the coupling with the plasmon, in addition to folded phonon modes. The linear power dependence of the phonon amplitude for all the modes suggests coherent phonons in 4H-SiC are generated through the virtual transition under the present condition. The power independence of the phonon frequency and dephasing time reveals the coupling of the LO phonon mode in 4H-SiC occurs with the doped carriers, but not with excited carriers probably due to the short decay time of excited carriers.

References