

Title	H-Si(111)ジャストおよびチルト表面の光和周波分光と顕微観察
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Optical sum frequency spectroscopy and microscopy of flat and vicinal H-Si(111) surfaces

Abstract

Silicon is one of the most important substrate for thin film growth which is employed in industrial applications such as biological sensor and solar cell. In order to grow a high quality thin film on a Si wafer, chemical vapor deposition (CVD) is a good method. Before loaded into CVD chamber, the Si substrate is terminated by hydrogen to protect the surface from oxidization. In the CVD process, the qualities of the grown thin film layer on Si surfaces depend on not only the radical reaction in the vapor phase but also an early reaction with the Si surface. Therefore, removal of the hydrogen layer from this Si surface is the central issue in the CVD growth. In this study, I will investigate the hydrogen desorption from H-Si(111) 1x1 surfaces by observing sum frequency generation (SFG) spectroscopy and microscopy. SFG is a sensitive method for surface analysis. Furthermore, in our previous work, hydrogen desorption on H-Si(111) surfaces irradiated by desorption-inducer IR light pulses was investigated by observing SFG intensity images. The interesting point which I stress here is the appearance of the dark boundary between the areas with and without the irradiation (corresponding to the areas with non-resonant and resonant SFG signals). This unidentified bonding state of H-Si bonds is still mysterious and not understood well. The dynamics of the electrons and phonons on the edges of the light pulse spot become complicated because the IR light pulse not only raises the surface temperature but also simultaneously excites the electron-hole (e-h) plasma. In order to clarify the dynamics at a particular part of this region, picoseconds-order snapshots of the hydride on a Si surface have to be taken after the pump light irradiation. Therefore, the pump-probe technique must be applied to the SFG microscope. SFG microscopy is forbidden for centrosymmetric bulk. Thus, it is considered as a sensitive method for monolayer of adsorbate on the surface.

In this study, I constructed a pump-probe SFG microscopy system for the first time. It contributes for surface science as a new analysis systems, and opens new opportunity for directly observing two dimension images of particular adsorbate species. By using this system, dynamics of H-Si species on the flat H-Si(111)1x1 surfaces were investigated by observing the time-resolved *ppp*-SFG spectra and microscopic images in a UHV chamber. The SFG intensity images showed the consistent results with the time resolved SFG spectroscopy. After visible pump light irradiation non-resonant SFG signal increased at probe delay time ~ 0 ps, and then decreased with the life time of ~ 1 ns. The original candidate of this change of the non-resonant SFG signal is suggested to be due to excitation and relaxation of e-h pairs after the pump light irradiation.

Also the reduction of hydrogen coverage of the H-Si(111)1x1 surface due to high surface temperatures was observed directly for the first time. I obtained isothermal desorption spectra of the H-Si(111) 1x1 surface at temperatures of ~ 711 , 732, 752 and 771 K by probing directly the vibrationally resonant optical SFG spectra. The desorption order of hydrogen was consistent with the second order scheme. As the first time of discovery, a theoretical peak shift (CPA calculation) reproduced the experiment quantitatively, and thus the peak shift was due to the modulation of average polarizability of the Si-H oscillators via dipole coupling.

On the other hand, I also investigated the hydrogen desorption from regular step H-Si(111) surfaces with 9.45° miscut toward $[\bar{1}\bar{1}2]$ direction. The hydrogen was terminated on the regular Si(111) surface by dosing hydrogen molecules in the UHV chamber. After hydrogen termination process on the vicinal Si surface, the upstairs SFG spectra with *ppp* and *ssp* polarization combination were taken. In both of cases, the terrace mode A (2082cm^{-1}), and step modes C_1 (2094cm^{-1}) peaks were clearly observed, while C_2 (2101cm^{-1}) and C_3 (2134cm^{-1}) were not. The A mode is readily attributed to the in-phase terrace vibration of the monohydride. The step C_2 , C_3 modes cannot be detected on the step bunched 9.45° cut surface because of the decrease in the step mode intensities (step bunches appeared when the Si surface was flashed at 1200°C). On the other hand, the downstairs SFG spectra with *ppp* and *ssp* polarization combination were also observed. The terrace mode A appeared in all spectra while the step C_1 mode appeared in *ssp* only.

Isothermal hydrogen desorption on the step Si(111) was investigated for the first time by observing the time-resolved SFG spectra. It reveals existence of a new configuration of step hydrogen orientation. Intensity transfers from the terrace peak to the step one and the step mode shifted to the lower frequency via dipole interaction effect. This study about hydrogen desorption on the step Si surface contributes to fundamental science and opens a new research topic.

Keywords: hydrogen desorption, sum frequency generation, vibrational mode, step surface, flat surface, heating.