

Title	光和周波および第二高調波分光によるSi(111)表面の水素脱離研究
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Desorption of hydrogen on the flat Si (111) surface studied by optical sum frequency and second harmonic spectroscopies

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Abstract

The hydrogen adsorption and desorption from a single crystal Si (111) surface have been studied actively with interest for a long time, since hydrogen desorption is the central rule of CVD growth of Si devices. However, some unanswered problems about the desorption order are still remaining, especially at low hydrogen coverage. In literature, isothermal hydrogen desorption was investigated from a H-Si (111)1x1 surface prepared by using chemical etching method. They used sum frequency generation (SFG) to detect the hydrogen coverage. That study showed that the hydrogen desorbed homogeneously but could not decide the desorption order at the hydrogen coverage around 0.2 ML, because at coverage lower than 0.2 ML the resonant SFG signal was indistinguishable from non-resonant background. Referring to this work, I intend to analyze the order of desorption from a flat Si (111) surfaces at high and low hydrogen coverage by using optical SFG and SHG spectroscopies.

In this study, I have investigated the hydrogen desorption mechanism from a flat H-Si (111)1x1 surface at 711 K by using both SFG and SHG spectroscopies. I have used SFG spectroscopy for investigating the hydrogen desorption at higher hydrogen coverage, and the SHG for lower hydrogen coverage on the hydrogenated Si (111) surfaces.

The H- Si (111)1x1 surface was prepared in a UHV chamber by dosing hydrogen molecules with ~3.5 Torr on the Si (111)7x7 surface at 600 °C. The SFG signal was obtained as a function of the IR light wavenumber. SFG spectra of each experiment were taken from 2060 cm⁻¹ to 2110 cm⁻¹ with a scanning step of 1 cm⁻¹. Each measurement was conducted in the polarization combinations

as *ppp* (SFG in *p*-polarization, visible in *p*-polarization and IR light in *p*-polarization). Before heating, the sharp peak at 2083.7 cm^{-1} is attributed to the stretching vibration of monohydride on the Si surface. After heating for each 10 s at 711 K, the sample was cooled down to RT, and the SFG spectrum was taken. This procedure was repeated for 20 s, 30 s, 40 s... and up to 230 s and the series of SFG spectra as a function of heating time were taken. From these resonant SFG spectra, the coverage of hydrogen was calculated. The calculated hydrogen coverage decreased from 1 ML to ~ 0.18 ML in ~ 230 s and was best fitted to the second order desorption.

As the other investigation, I have applied SHG spectroscopy for the hydrogen desorption order at low hydrogen coverage below ~ 0.18 ML. When the SFG signal became comparable to the background due to the lower hydrogen coverage, I switched to SHG measurement and detected the Si dangling bonds. The fundamental light wavelength 1064 nm with power of $380\ \mu\text{J/pulse}$ was used as the excitation light. In this experiment, I used the polarization configurations $P_{\text{in}} P_{\text{out}}$. The sample was heated for each 50 s at 711 K and then cooled down to RT, and the SHG signal was taken. This procedure was repeated for 230 s, 280 s, 330 s, 380...up to 3880 s. Then I heated the sample in different interval of times and measured SHG signal up to 18330 s. The heating time dependent SHG intensity curve showed that the intensity initially increased rapidly as a function of heating time and then gradually saturated when the number of dangling bonds were saturated. The initial coverage of SHG measurement was 0.18 ML. The hydrogen coverages during the isothermal desorption was tried to be analyzed to the first (1st), intermediate (1.5th) and second (2nd) order theoretical curves. The reduction of hydrogen coverage showed that the first order was the best fitted.

In addition, investigation of the hydrogen desorption from the H-Si (111)1x1 surface has been done at various high surface temperatures. The heating temperatures were of ~ 711 , 730, 750 and 770 K. By SFG, I detected Si-H vibration and investigated hydrogen desorption at the high hydrogen coverage from 1 ML to 0.44 ML since SFG signal was close to background at low hydrogen coverage. After SFG measurement I switched to SHG measurement and detected Si dangling bonds

and monitored the hydrogen coverage when it was lower than ~ 0.44 ML. This investigation showed that the hydrogen desorption was assigned as second order in the coverage range 1 ML to 0.44 ML for all of the heating temperatures. The low coverage hydrogen desorption was assigned as first order in the range below 0.44 ML to 0.0 ML for all heating temperatures. Combining the SFG and SHG analyses, the desorption order and also desorption activation energy were clarified on the whole hydrogen coverage from 1 ML to 0 ML. From the series of SFG and SHG spectra observed at several high temperatures, I have calculated the desorption activation energy for high and low coverage of hydrogen. For second order desorption, the activation energy was 1.96 ± 0.49 eV and for first order desorption the activation energy was 1.41 ± 0.35 eV. The value of activation energy was low for lower coverages and high for higher coverage.

Keywords: Sum frequency generation (SFG); Second harmonic generation (SHG); Si (111)1x1surface; desorption order; desorption activation energy; hydrogen coverage.