Chapter 7

Conclusions

The experiments described here have helped improve the understanding the physics of SHG at Si surfaces or interfaces. The experimental results show that the SH spectroscopy from Si surfaces is closely related to the band structure of the bulk Si, but surface modification can shift the apparent resonance through interference among or between different SH contributions. The major advantage of SHG is the extreme surface sensitivity. As shown for oxidized Si, hydrogen terminated Si, and ultrathin Cr coated SiO_2-Si surfaces, the measured RA-SHG signals for appropriate polarizations at a certain photon energy are apparently different between samples. This sensitivity can be greatly enhanced by combination with the phase information of the surface SH field. The observed peak-to-valley transition of RA-SHG by varying photon energy or by surface modification clearly demonstrates the phase variation of the surface SH field. With this information, SHG from Cr-SiO_2-Si structures and thermally oxidized Si surfaces have been comprehensively studied and new results of SHG have been obtained with respect to the sources of SHG.

A major weakness of the SHG technique is the lack of specificity for a certain given interfacial property. The observed variation of SHG with surface condition is possibly explained by alternative effects. Many conclusions drawn from SHG studies are not straightforward. It is also clear that SHG from SiO_2-Si interfaces is considerably time-dependent, especially for ultrathin oxide surfaces. Measured SH signals depend strongly on experimental conditions, thus quantitative characterization

is difficult. Another problem with the SHG probe is the possibility of damaging the surface with the high field intensity required to generate an observable response. For Si surfaces, our SHG studies so far have given adequate signals well below the threshold for optical damage. Further understanding of the interfacial electronic structure and a feasible microscopic theory are needed to explain microscopically the phase variation of SHG and the SH spectroscopy.

However, the high sensitivity of the SHG probe to interface properties can be put to the best use by comparing SH signals from different surfaces, as I demonstrated in this thesis. The use of SHG to monitor the time-evolution of the surface or interface and the carrier dynamics at the surface or interface is also very exciting. I have performed a new spectroscopic study of SHG from technologically important Si(001) surfaces. The SH spectroscopy with the phase information of the SH field is far more sensitive to interface properties than that with the SH intensity only.