

Spectroscopic Studies of Optical Second-Harmonic Generation

from Si(001) Surfaces

by

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Thesis directed by Professor Steven T. Cundiff

I present a spectroscopic study of optical second-harmonic generation (SHG) from Si(001) surfaces and interfaces in the vicinity of the direct two-photon E_1 transition using tunable femtosecond lasers. The samples investigated are oxidized Si, hydrogen terminated Si, and Cr-SiO₂-Si structures. I first use a phenomenological theory and susceptibility tensors to predict the symmetry properties of several different SHG contributions and present methods for separating bulk and surface SHG contributions and uniquely determining susceptibility tensor elements. By measuring polarization selected rotational-anisotropy SHG (RA-SHG), I show that both bulk and surface SHG contributions display resonances and that interference between these contributions can shift the apparent resonance energy. The strength of bulk and surface SHG contributions varies with photon energy. Linear optics also plays a role in SHG spectroscopy. For certain photon energies, the peak locations of the RA-SHG signals from oxidized and hydrogen terminated Si(001) surfaces differ. This indicates phase shift between surface SHG fields. For appropriate polarizations, peaks of the RA-SHG signals from oxidized Si surfaces can be turned into valleys by varying the photon energy, and eightfold symmetric RA-SHG signals can be observed at certain photon energies. Comparison of RA-SHG signals from Cr-SiO₂-Si

structures and oxidized Si samples also shows a difference in the peak location at certain photon energies. Further experimental results show that an ultrathin Cr coating film on oxidized Si introduces additional sources of SHG, which modify the spectra and time-dependence of SHG. I also study the effect of thermal oxidation of Si(001) samples on SHG and show that SHG is sensitive to interface width. RA-SHG signals with eightfold symmetry are found for several different polarizations and the corresponding photon energies are sensitive to interface conditions. Thermal oxidation affects the time-dependence and spectroscopy of SHG. These results indicate that SHG spectroscopy is a powerful tool for characterizing Si surfaces or interfaces.

Dedication

To the memory of my grandfather.

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CONTENTS

Abstract, Acknowledgments, Contents, List of Tables, and List of Figures	i-xv
Chapter	
1 Introduction	1
1.1 Historical Overview of Second-Harmonic Generation (SHG)	1
1.2 This Work	4
1.3 Experimental Setup	8
2 Phenomenological Theory of SHG from Si Surfaces	14
2.1 Introduction	14
2.2 The Model	14
2.3 Rotational-Anisotropy SHG (RA-SHG)	18
2.3.1 Bulk and Surface Second-Harmonic (SH) Contributions	18
2.3.2 RA-SHG from Vicinal Si Faces	21
2.3.3 RA-SHG from the Si(001) Face	32
2.4 Separation of Bulk and Surface SH Contributions	35
2.5 Summary	38
3 Bulk and Surface Contributions to Resonant SHG	40
3.1 Introduction	40
3.2 Bulk and Surface SHG Contributions	42
3.3 Sample Preparation and Experimental Methods	43
3.4 Experimental Results	44
3.4.1 Spectroscopy of the Bulk Anisotropic Contribution	44
3.4.2 Spectroscopy of the Isotropic Contribution	47

3.5 Discussion	51
3.6 Summary	52
4 Phase Inversion in RA-SHG	54
4.1 Introduction	54
4.2 Experimental Conditions and Sample Preparation	55
4.3 Observed Phase Inversions	55
4.3.1 Phase Inversion due to Photon Energy Variation	55
4.3.2 Phase Inversion due to Surface Modification	57
4.4 Phase and Amplitude of Susceptibility Tensors	59
4.5 Discussion	63
4.6 Summary	66
5 SHG from Cr-SiO ₂ -Si(001) Structures	68
5.1 Introduction	68
5.2 Theoretical Background	71
5.3 Experimental Conditions and Sample Preparation	73
5.4 Experimental Results	73
5.4.1 Comparison of RA-SHG from Cr-SiO ₂ -Si and SiO ₂ -Si Surfaces	73
5.4.2 Spectroscopic Study of RA-SHG from Cr-SiO ₂ -Si	79
5.4.3 Comparison of TD-SHG from Cr-SiO ₂ -Si and SiO ₂ -Si Surfaces	81
5.5 Discussion and Further Experiments	83
5.5.1 Phase Inversion due to Additional SHG Sources	83
5.5.2 Further Experiments to Detect for SHG Sources	84
5.5.3 Insight into the Charge Trapping Process	88

5.6 Summary	91
6 SHG from Thermally Oxidized Si(001) Surfaces	93
6.1 Introduction	93
6.2 Experimental Conditions and Sample Preparation	96
6.3 Variation of SHG with Oxide Thickness	97
6.3.1 Isotropic and Anisotropic SHG Contributions	97
6.3.2 Phase Inversion Photon Energy	101
6.3.3 Time-dependent SHG	104
6.3.4 Effect of Thermal Oxidation on SHG Spectroscopy	105
6.4 Discussion	108
6.5 Summary	109
7 Conclusions	111
Bibliography	113
Appendix: SHG from Vicinal Si(001) Surfaces	121

LIST OF TABLES

Table

2.1 Angular functions $\Phi_m(\alpha)$	25
2.2 $b_{00,(g,h)}^{BQ}$ and $b_{m,(g,h)}^{BQ}$ as functions of Fresnel factors	26
2.3 $b_{00,(g,h)}^d$ and $b_{m,(g,h)}^d$ as functions of Fresnel factors	28
2.4 $s_{m,(g,h)}^{SD}$ as combinations of Fresnel factors and tensor elements	30

LIST OF FIGURES

Figure	
1.1 Experimental setup for surface SHG	9
2.1 Geometry and unit vectors for the propagating fundamental and SH fields	15
2.2 Diagram of the beam frame and the vicinal surface with respect to the cubic crystal axes	22
2.3 Definition of the linear polarization configuration with the incident fundamental beam (a) and the reflected SH beam (b)	33
3.1 Spectra of the anisotropic SHG contribution $a_{4,(p,s)}$, the linear optics coefficient $L_{(p,s)}$, and the magnitude (ζ) of the anisotropic element ζ	46
3.2 RA-SHG intensities from modified Si(001) surfaces (NO-Si, TO-Si and H-Si) at a two-photon energy of 3.26 eV for different polarization configurations: (p, p) , top panel and (p, s) , bottom panel	48
3.3 Spectra of the isotropic amplitudes $a_{0,r}$ for NO-Si and TO-Si (top panel) and the interfering and non-interfering isotropic amplitudes $a_{0,r}$ and $a_{0,i}$ for H-Si (bottom panel)	50
4.1 RA-SHG signals from both TO-Si and NO-Si samples for the polarization (s, p) at several two-photon energies	56

4.2 RA-SHG intensities from modified Si(001) surfaces (NO-Si, TO-Si, and H-Si) at a two-photon energy of 3.26 eV for different polarizations: (s, p), top panel, and (q, s), bottom panel	58
4.3 Spectra of the amplitudes and relative phases of the tensor elements ∂_{31} (upper panel) and ∂_{15} (lower panel) for all three samples: TO-Si, NO-Si and H-Si	62
5.1 RA-SHG signals from Cr-NO-Si and Cr-TO-Si samples for the (p, p), (s, p), and (q, s) polarizations at a two-photon energy of 3.40 eV	75
5.2 RA-SHG signals from both NO-Si and TO-Si samples for the (p, p), (s, p), and (q, s) polarizations at a two-photon energy of 3.40 eV	76
5.3 Comparison of the (p, s) polarized RA-SHG signals from TO-Si and Cr-TO-Si samples at a two-photon energy of 3.40 eV	78
5.4 (p, p) polarized RA-SHG intensities from the Cr-TO-Si sample at two-photon energies. Note that the RA-SHG with eightfold symmetry appears at the two-photon energy of 3.35 eV	80
5.5 Comparison of the TD-SHG signals for the (p, p) polarization at a two-photon energy of 3.40 eV for different samples: NO-Si and TO-Si (upper panel); Cr-NO-Si and Cr-TO-Si (lower panel)	82
5.6 Isotropic SHG signals variation with two-photon energy from the surfaces of bulk Cr, thin Cr coated silica, and silica covered Cr	86
6.1 Ratio of the isotropic to anisotropic SHG components for the (p, p) polarization (upper panel) and anisotropic SHG components for the (p, s)	

- polarization (lower panel) as a function of oxide thickness at several two-photon energies: 3.44 eV, 3.40 eV, and 3.26 eV 100
- 6.2 Upper panel: oxide thickness dependence of the two-photon energy at which the RA-SHG signal shows eightfold symmetry for (p, p) , (q, s) , and (s, p) polarizations. Lower panel: RA-SHG signals from a thermally oxidized Si(001) sample with 57.9 nm thick oxide for the (s, p) polarization, showing either eightfold or fourfold symmetry 102
- 6.3 Time-dependent SHG signal for the (p, p) polarization at a two-photon energy of 3.26 eV for several thermally oxidized Si(001) surfaces with different thicknesses of the oxide layer 104
- 6.4 SH spectra from a thermally oxidized Si(001) sample with 57.9 nm thick oxide at a fixed azimuthal angle of 22.5° for different polarization configurations: (p, p) , (s, p) , and (q, s) 107
- A.1 (p, p) polarized RA-SHG signals at a two-photon energy of 3.22 eV from natively oxidized vicinal Si(001) surfaces (NO-Si V) (upper panel) and thermally oxidized vicinal Si(001) surfaces (TO-Si V) (lower panel) with different vicinal angles of 0° , 1° , 2° , 3° , 4° , and 5° 123
- A.2 (p, p) polarized RA-SHG signals from a natively oxidized Si sample with vicinal angle of 5° (NO-Si V5) (upper panel) and a thermally oxidized Si sample with vicinal angle of 5° (TO-Si V5) (lower panel) at two-photon energies 4.34 eV, 4.41 eV, and 4.48 eV 124
- A.3 RA-SHG signals from a natively oxidized Si sample with vicinal angle of 5° (NO-Si V5) at several two-photon energies of 3.10 eV, 3.26 eV, 3.40 eV,

and 3.49 eV for different polarizations: (p, p) , (s, p) , (p, s) , and (s, s) 125

A.4 RA-SHG signal from a thermally oxidized Si sample with vicinal angle of 5° (TO-Si V5) at several two-photon energies of 3.10 eV, 3.26 eV, 3.40 eV, and 3.49 eV for different polarizations: (p, p) , (s, p) , (p, s) , and (s, s) 126

A.5 RA-SHG signal from a hydrogen terminated Si sample with vicinal angle of 5° (H-Si V5) at several two-photon energies of 3.10 eV, 3.26 eV, 3.40 eV, and 3.49 eV for different polarizations: (p, p) , (s, p) , (p, s) , and (s, s) 127