Spectroscopic Studies of Optical Second-Harmonic Generation

from Si(001) Surfaces

by

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A thesis submitted to the

Faculty of the Graduate School of the

University of Colorado in partial fulfillment

of the requirements for the degree of

Doctor of Philosophy

Department of Physics

2003

This thesis entitled:
Spectroscopic Studies of Optical Second-Harmonic Generation from Si(001) Surfaces
written by Yong Qiang An
has been approved for the Department of Physics

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The final copy of this thesis has been examined by the signatories, and we find that both the content and the form meet acceptable presentation standards of scholarly work in the above mentioned discipline.

An, Yong Qiang (Ph. D., Physics)

Spectroscopic Studies of Optical Second-Harmonic Generation from Si(001) Surfaces

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-SiO_2-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-SiO_2-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.

Acknowledgments

There are many people who have made this thesis and this work possible.

I wish first to thank my advisor, Steven Cundiff, for his support and guidance during my graduate studies. His ability to instruct clear physics and to explain it concisely has set a standard to which I will always aspire. His willingness to promote me professionally through teaching me English and sending me to conferences has been most generous. He has always been available to talk to me. Through his interest and enthusiasm for my work, he has driven me to do my personal best in the lab. An advisor can do little more for a student.

I would like to thank my committee members: Charles Rogers, Daniel Dessau, Henry Kapteyn, and Kelvin Wagner, for many helpful comments and questions on my thesis.

I am grateful for the help of many incredibly talented scientists, postdocs, and students at JILA during my graduate studies. I thank Prof. Stephen Leone for help in my Comps III exam. I thank Prof. David Nesbitt for critical reading of my manuscripts. JILA is a wonderful place to do research because of the wide array of talented, knowledgeable, and helpful support staff. I am thankful for the electronics shop, the computing, the instrument shop, and the administration staff for their help. I thank David Alchenberger for help in preparing experiment samples, and Barbara Tennis and Marilee DeGeode for helpful English classes.

I would like to thank the whole Cundiff group for making the lab an enjoyable place to work and to live. These members are Justin Shacklette, Amy Spivey, Tara Fortier, Kelda Furbush, Virginia Lorenz, Zhigang Chen, Qudsia Quraishi, Tianhao

Zhang, David Jones, Camelia Borca, Rudolf Bratschitsch, Pete Roos, Martin Griebel, Soobong Choi, and Jessi Ames. I enjoyed and would remember our group skiing trips to Eldora and Keystone.

Finally, I would like to thank all my family members for their constant love and support. Especially, I thank my wife, Chuanxia, for all of her kind support, encouragement, and unconditional love, and my son, Rocky, for the love and joy he brings.

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