Optical SHG and RAS of molecular adsorption at Si(001) step edges

Robert Ehlert, Jinhee Kwon and Michael C. Downer
Department of Physics, The University of Texas at Austin, Austin TX 78712, USA.


Outline

- Motivation
- SHG & RAS
- Results from oxidized vicinal Si(001)
- Clean and H-adsorbed vicinal Si(001)
- SBHM analysis
- Conclusion and Outlook
Optical sensors provide feedback control for self directed growth of nanostructures

Nanostructures offer possibility to further reduce size of micro/nano-electronics

- 1D quantum wires, molecular electronics, atomic-scale memory, quantum computers

- Atom by atom assembly very slow
- Self directed growth techniques desirable
- Silicon stepped surfaces can be used as templates for self directed growth of nanostructures
- Self directed growth of nanostructures is faster but also less controlled

Non-invasive in-situ sensors are needed

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1. McChesney, Nanotech. 13, 545 (02)
2. Kasemo, Surf. Sci. 500, 656 (02)
3. Bennewitz, Nanotech. 14, 499 (02)
4. Ladd, Phys. Rev. Lett. 89, 017901 (02)

Optical probes are non-invasive, non-destructive, and applicable to any transparent medium.

**SHG**

- High spatial and spectral resolutions
- Real-time mapping of fast surface dynamics

**RAS**

- Measure surface-induced optical anisotropy
- Obtain information concerning surface electronic structure and chemistry

Combination of SHG and RAS provides comprehensive database for bond level modeling of step surfaces.
Optical setups to investigate samples in UHV environment

- Coherent MIRA
  Ti:Sapphire oscillator
  710-900nm
- NOPA
  520nm-780nm
Results from previous studies of oxidized vicinal Si(001) surfaces


- Applied SBHM to vicinal Si/SiO$_2$ interface

**Summary of main results:**

- relative bond strength
- varying upper bond angle

**Basis for analysis:**

- mutually consistent SHG & RAS spectra
- Kramers-Kronig consistency
- 2 SHG polarization combinations (sP,pP)

**Recommended** but not done yet:

- extend to other polarization combinations (sS,pS)

Clean reconstructed vicinal Si(001) surface prepared in ultra high vacuum

Ab-initio DFT calculation of $D_B$ step structure on Si(001):6° at $T = 0$ K
(courtesy E. Pehlke)

Selective adsorption of H$_2$ on step-edges strongly influences SHG at $\lambda = 1064$ nm


Single wavelength SHG sensitive to H$_2$ adsorption

OSI VII, Jackson Hole, Wyoming, July 16th 2007
Spectroscopic SH and RAS study of H$_2$ adsorption on step-edges

+ H$_2$

SHG of reconstructed vicinal Si:6°

RAS

H$_2$

p-in/p-out

clean p-in/p-out

OSI VII, Jackson Hole, Wyoming, July 16th 2007
SBHM using ab-initio structure calculation fits SHG data with high fidelity

**Input**

- *ab initio* DFT calculation provides input structure for SBHM analysis
- complex hyperpolarizabilities $\beta$ of up to 7 classes of bonds

**Results**

- SBHM fits to RA-SHG data at selected wavelength

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Fitted hyperpolarizabilities yield linear bond polarizabilities that reproduce RAS data.
Hyperpolarizability spectra of individual surface bonds reveals charge transfer at identifiable molecular sites.
Conclusion

- application of SBHM to spectroscopic SHG and RAS data suggests general **ability to “see” charge transfer** from dangling bonds to back bonds.

- Preliminary results, limited ability to give reliable microscopic information

- expand data set: other polarization combinations*, broader spectral range

- better microscopic calculations are needed ➭ beyond SBHM


- Eventually move to other adsorbates i.e. organics molecules

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**Research group**

<table>
<thead>
<tr>
<th>Name</th>
<th>Title</th>
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<tbody>
<tr>
<td>Jinhee Kwon</td>
<td>PhD</td>
</tr>
<tr>
<td>Yong An</td>
<td>PhD</td>
</tr>
<tr>
<td>Adrian Wirth</td>
<td>MA</td>
</tr>
<tr>
<td>Junwei Wei</td>
<td>MSc</td>
</tr>
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