

Terrace and step contributions to the surface optical anisotropy of Si(001)

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Abstract The contributions of flat terraces as well as of S_A , S_B and D_B steps to the optical anisotropy of Si(001) surfaces have been calculated using a real-space multigrid method. We find a distinct influence of the dimer arrangement on the optical spectra. The signal measured for atomically smooth terraces is well described by the calculated spectrum for the Si(001) $c(2\times 4)$ surface. Surface steps are responsible for the significant optical anisotropy around 3 eV observed for vicinal surfaces. The influence of sub-surface atomic relaxations on the anisotropy signal is found to be surprisingly small.

1 Introduction

Reflectance anisotropy/difference spectroscopy (RAS/RDS) is an extremely versatile tool of surface analysis. Thanks to the development of both computational techniques and resources it is now possible to calculate anisotropy spectra from *first principles* in quantitative agreement with experiment [1]. Such calculations are very involved and have by now been restricted to ideal surfaces. The complete understanding of RAS spectra, however, requires to consider realistic surfaces, containing defects such as surface steps. We calculate the RAS of flat Si(001) surfaces forming (1×2) , $p(2\times 2)$ and $c(2\times 4)$ reconstructions and of S_A , S_B and D_B surface steps from *first-principles*. Particular attention is paid to the origin of bulk-related features in the optical spectra.

2 Method

Our massively parallel, real-space finite-difference calculations [2] are based on density-functional theory in the local-density approximation and use a multigrid technique for convergence acceleration. The surface is modeled by periodic supercells containing asymmetric slabs of 11 atomic Si layers separated by 12 Å of vacuum. The dangling bonds at the bottom layer are saturated with hydrogen. S_A and S_B/D_B steps, respectively, are modeled by missing dimer structures in (2×6) and (2×7) surface unit cells. To calculate the dielectric function we use a sampling of about 500 – 1000 points in the full (1×1) surface Brillouin zone. The slab polarizability calculated in the independent-particle approximation is used to calculate the RAS spectra. Further details can be found in our study on stepped Si(111):H surfaces. [3]

3 Results and Discussion

3.1 Flat surface

The RAS spectra calculated for (1×2) , $p(2\times 2)$, and $c(2\times 4)$ reconstructed Si(001) are shown in Fig. 1. The

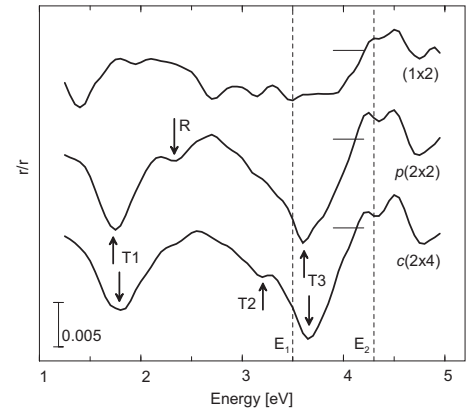


Fig. 1 RAS spectra $[Re\{(r_{[\bar{1}10]} - r_{[110]}) / \langle r \rangle\}]$ calculated for the Si(001) (1×2) , $p(2\times 2)$ and $c(2\times 4)$ surfaces. Dashed lines mark the positions of bulk critical point energies.

spectrum for (1×2) shows a pronounced minimum at 1.4 eV, a rather broad negative anisotropy between 2.5 and 4.0 eV and a maximum at 4.5 eV, consistent with previous *ab initio* results [4,5]. The alternating arrangement of the asymmetric dimers in a $p(2\times 2)$ rather than a (1×2) structure leads to pronounced spectral changes. Two distinct minima, T1 and T3, appear. The dimer-dimer interaction extends even beyond the dimer rows: differences in the RAS also appear in the spectra calculated for $p(2\times 2)$ and $c(2\times 4)$ surfaces. T1 shifts from 1.7 to 1.8 eV and the local minimum R evolves into a shoulder. A new feature, T2, appears at 3.2 eV.

Comparing the RAS calculated for (1×2) , $p(2\times 2)$ and $c(2\times 4)$ reconstructed surfaces with experiment, we find that the calculated anisotropy for $c(2\times 4)$ surfaces agrees well with the anisotropy measured for highly oriented samples [6, 7].

To determine the origin of the spectral features we separate the contributions to the RAS from electronic transitions within the uppermost four atomic layers and from the bulk-like layers underneath. Fig. 2 shows that surface-state related transitions are mainly responsible for the optical anisotropies below the E_1 energy, T1 and T2. T3 and the two maxima below and above the E_2 en-

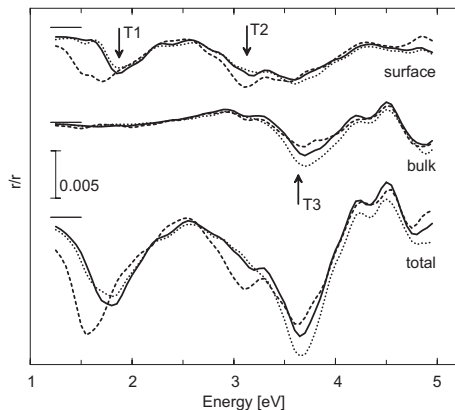


Fig. 2 RAS for the Si(001) $c(2\times 4)$ surface, calculated for the fully relaxed configuration (solid lines) in comparison to calculations for a geometry where the relaxation was restricted to the uppermost two atomic layers (dashed lines). Spectra calculated for an anisotropically strained slab (see text) are shown by dotted lines. Shown are contributions to the RAS from electronic transitions within the uppermost four atomic layers, from the layers underneath, and the total signal.

ergy originate from bulk layers. They depend only little on the surface symmetry and appear for (1×2) , $p(2\times 2)$, and $c(2\times 4)$ periodicities.

The appearance of optical anisotropies *not* related to transitions between surface states was suggested to be related to subsurface stress induced by the dimerization of the surface atoms [8]. We investigated this hypothesis by additional calculations. First a geometry was studied where only the uppermost two atomic layers were allowed to relax, while the remaining atoms occupied ideal bulk positions. Dashed lines in Fig. 2 show the resulting RAS. The surface-related features are distinctly changed. Only small modifications, in particular around the T3 feature, however, are observed for the RAS arising from bulk layers.

The dimerized surface is stretched along the dimer bonds and compressed across the dimer rows. As a second test we therefore relax the Si(001) $c(2\times 4)$ surface in a unit cell whose lateral dimensions are shrunk/enlarged by 0.5 % along $[110]/[\bar{1}10]$. The spectra calculated for this configuration are shown by dotted lines in Fig. 2. Again, the optical anisotropies arising from subsurface transitions are only slightly affected. The effect is strongest for the T3 feature, which may thus be partially attributed to reconstruction-induced stress beneath the surface. The salient RAS features originating from bulk layers, however, are neither affected by stress nor relaxation. This indicates that they are caused by the anisotropic surface potential.

3.2 Surface steps

In the present study we focus on the three most commonly observed step configurations: S_A steps and rebonded steps of type S_B and D_B . The calculated step-generated RAS signal is compared with the experimental

findings [7] in Fig. 3. Given the limitations of our study, in particular with respect to the slab size, the comparison is gratifying. In agreement with experiment we find that surface steps on Si(001) give rise to a broad negative anisotropy below the E_1 energy, with a minimum at around 3 eV. Experimentally, a positive anisotropy between 3.5 and 4 eV is observed for miscut angles larger than 4° . A similar feature appears in the calculation for D_B steps. As biatomic steps form for larger miscuts only, the observed changes of the measured line shape with vicinity finds a natural explanation.

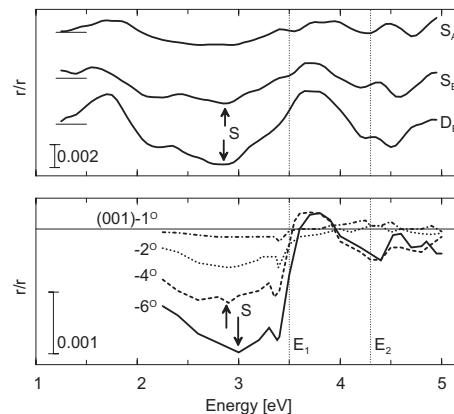


Fig. 3 Upper panel: Step-induced optical anisotropy calculated for S_A , S_B and D_B steps. Lower panel: Step-induced optical anisotropy measured for different miscut angles [7].

Combining our results for flat, $c(2\times 4)$ reconstructed terraces and for the step related anisotropies, the peculiar RAS lineshape observed for vicinal samples [4, 9] can be understood as superposition of terrace- and step-generated optical anisotropies.

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