

## Step-Induced Optical Anisotropy of Vicinal Si(001)

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It is demonstrated, using reflectance difference spectroscopy, scanning tunneling microscopy, and low-energy electron diffraction, combined with deliberate straining of the surface, that the presence of atomic steps dramatically changes the optical anisotropy of the Si(001) surface. The step-induced reflectance difference signal originates predominately from rebonded steps and is comparable in magnitude to that of the terrace signal. [S0031-9007(98)08282-9]

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Recently considerable effort has been made to develop reflectance difference spectroscopy (RDS) and reflectance anisotropy spectroscopy (RAS) as a tool for real-time characterization of surface phenomena and growth processes in semiconductor surfaces [1–4]. RDS displays high surface and chemical sensitivity, is easily combined with other surface techniques, and has proven especially useful in high-pressure chemical vapor deposition (CVD) growth environments, where most conventional surface analysis techniques fail. RDS measures the anisotropy in the optical reflectance from a material [1]. In systems with an isotropic or nearly isotropic bulk, optical anisotropies are primarily induced by the surface. If a sample is illuminated with linearly polarized light, anisotropies in the reflectance from the principal symmetry directions of the surface will result in a rotation of the polarization vector and/or a phase delay between orthogonal polarization components. Because reconstructions on many semiconductor surfaces rotate from layer to layer, the probe becomes a means to investigate growth and layer completion.

Although early work has focused primarily on III-V surfaces, RDS is potentially a powerful diagnostic also for group IV film growth. To that end, a number of studies have been performed on the model group IV surface Si(001). Despite considerable effort, however, our understanding of the origins of particular features in RD spectra from Si(001) is still quite limited and a correlation between the optical signal and the atomistic surface structure has not been established. In particular, the role of steps in modifying the optical anisotropy of clean Si(001) has been investigated in several studies [5–7], but has not been clarified. For Si(001) with an adsorbed layer, an influence of atomic steps on the optical anisotropy has been demonstrated in several studies. The behavior of the RD signal from H-terminated and Si suboxide (Si-O-substrate) covered Si(001) as a function of substrate miscut suggests that steps are the dominant source of the optical anisotropy for the surfaces [5,6]. On the other hand, atomic steps appear to make no contribution to the RD signal from As-terminated Si(001) surfaces miscut  $4^\circ$  toward [110], for which the step

density is very high [7]. A recent study looking directly at clean, vicinal Si(001) also concluded that steps make a negligible contribution to the optical signal [6]. These last two studies have been used to support the premise that steps play no role in the optical anisotropy of the clean Si(001) surface [6–8], despite the fact that this conclusion seems physically implausible. In contrast, a recent RD study utilizing electromigration-induced step motion postulated the existence of a step-induced optical signal [9].

In this Letter we clarify this issue. We demonstrate a significant step-induced component to the RDS signal from Si(001). Our measurements are made on surfaces with well-defined morphology, which is varied by changing the sample miscut and by applying external strain. We use step morphological data from direct scanning tunneling microscopy (STM) measurements. We are able to extract the terrace and step spectra separately by quantifying the spectra self-consistently. Our result that atomic steps make a major contribution to the RD spectrum from vicinal Si(001) has important implications for the validity of first-principles theoretical calculations of the optical anisotropy of the Si(001) surface.

The Si(001) surface is cleaned following the conventional procedure [10]: high-temperature flashes up to  $1250^\circ\text{C}$  for approximately 20 sec followed by a slow anneal down to room temperature at a base pressure of  $\leq 1 \times 10^{-10}$  Torr. All measurements are made in a chamber containing an STM, a high-resolution low-energy electron diffractometer (HR-LEED), and an external RD spectrometer, allowing the characterization of both the microscopic surface structure and the net optical anisotropy. The chamber also contains a station that allows cantilevered samples to be strained precisely along the [110] or  $[-110]$  axes [11]. The STM can image the surface along the entire length of the sample, allowing position dependent measurements of the step morphology on the externally strained samples. The RDS apparatus is similar to ones described in the literature [12]. RD spectra from orthogonal sample orientations were subtracted and divided by 2 to remove systematic background. RDS is performed with the sample in the LEED position.

The Si(001) surface reconstructs to form rows of dimers, and the dimer orientation rotates by  $90^\circ$  from one atomic level to the next, giving alternating  $2 \times 1$  and  $1 \times 2$  terraces. These terraces are separated (depending on the sample miscut) either by single-atomic-height steps, labeled  $S_A$  ( $S_B$ ), with the dimer rows at the upper terrace oriented parallel (perpendicular) to the step, or by double-atomic-height steps (separating  $2 \times 1$  terraces), labeled  $D_B$ , with the dimer rows on the upper terrace oriented perpendicular to the step [13]. For surfaces miscut toward [110], the relative abundance of  $2 \times 1$  terraces and  $D_B$  steps increases with miscut angle [14]. The surface dimerization is nearly unchanged by an  $S_A$  step, while the rebonding of the  $S_B$  and  $D_B$  steps results in significantly different atomic bonding not only for the actual step atoms but also for the nearest dimer both above and below the step. The rebonded dimer on the lower terrace at an  $S_B$  step has one of its dangling bonds occupied as a back bond while the back bond of the nearest dimer on the upper terrace is being strained by the rebonding. The bonding in the step region is the same for  $S_B$  and  $D_B$  steps [13]. The region directly affected by the rebonding has a width of  $9.6 \text{ \AA}$ .

The predominant sources of structural anisotropy on the surface are terrace dimers and steps, and one can imagine that they both contribute to the net optical anisotropy of the surface. Distinctly different RD signatures from terrace and rebonded-step dimers can be expected because the charge distribution is completely altered at the step. Nevertheless, the conventional wisdom states that atomic steps make no significant contribution to the RD signal from clean Si(001) [6,7]. As stated earlier, this conclusion seems physically implausible: For a  $4^\circ$  miscut surface, 25% of the surface atoms reside at steps (i.e., are not terrace atoms), while for a  $6^\circ$  miscut, 37% of the atoms do. It would be surprising if these atoms, which are in a highly anisotropic environment, would produce no optical signal.

To determine the step contribution to the optical anisotropy, we applied external stress to nearly perfectly (001)-oriented Si samples to manipulate the relative abundance of the  $2 \times 1$  terrace [11,15]. On a clean equilibrium surface miscut  $\leq 0.3^\circ$  from [001], the step density is very low ( $< 1$  step every  $250 \text{ \AA}$ ) and the areas of the  $2 \times 1$  and  $1 \times 2$  terraces are nearly equal [14]. Consequently, nearly no net optical anisotropy is observed from such a surface. We strain the surface along the [110] or  $[-110]$  directions at elevated temperatures, lowering the free energy (and thus increasing the area) of one terrace type with respect to the other [15]. We then quench the sample temperature before removing the applied stress, producing surfaces with the same low step density as the equilibrium surface but with high population asymmetries between the  $2 \times 1$  and  $1 \times 2$  terraces. The cantilevered straining method [11] causes the population asymmetry to vary along the length of

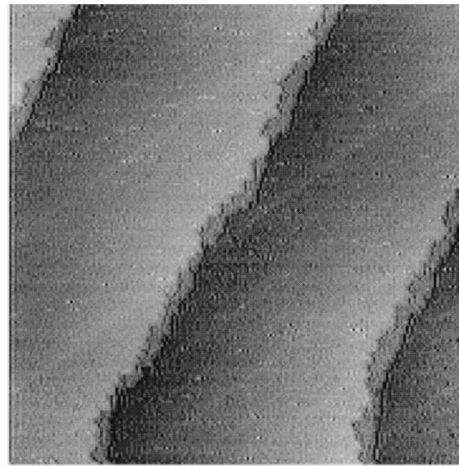


FIG. 1. Scanning tunneling microscopy image ( $300 \times 300 \text{ nm}^2$ ) of Si(001) miscut  $0.03^\circ$  towards [100], showing the population asymmetry induced by externally applied stress. A quantitative analysis of STM images yields an asymmetry of 92:8 for  $2 \times 1 : 1 \times 2$  reconstructions.

the sample [15]. Figure 1 shows an STM image of the high-strain end of a sample miscut  $0.03^\circ$  toward [100]. The surface has been driven to approximately 92%  $2 \times 1$  terrace and 8%  $1 \times 2$  terrace while the step area is  $< 1\%$  of the total surface area. The distance between step pairs is approximately  $1500 \text{ \AA}$ . For a miscut toward [100], all steps are composed of equal amounts of  $S_A$  and  $S_B$  step segments. The RD contributions of adjacent steps cancel overall because the  $S_A$  and  $S_B$  segments are rotated by  $90^\circ$  from step to step, and hence an influence of steps on the RD spectral shape can be neglected even if their density were high. The RD spectra of surfaces strained to predominantly  $2 \times 1$  or  $1 \times 2$  terraces (by reversing the strain axis) differ only in sign. We designate the RD spectrum of the strained sample miscut  $0.03^\circ$  toward [100] [Fig. 2(a)] as our pure terrace spectrum (see below). The same RD spectrum is produced by electromigration-induced single-domain surfaces of  $0.03^\circ$  miscut Si(001) [9].

To look for a potential step-induced component in the optical anisotropy of Si(001), we measured the RD spectra from equilibrium surfaces with miscuts ranging from  $1^\circ$  to  $6^\circ$  toward [110] [Fig. 2(b)]. The spectra show two main features, at 3 and 3.7 eV, which change their relative strength with vicinality.

The total optical anisotropy can be written as

$$\Delta \mathbf{r}/\mathbf{r} = f_t(\Delta \mathbf{r}/\mathbf{r})_t + f_s(\Delta \mathbf{r}/\mathbf{r})_s, \quad (1)$$

where  $\Delta \mathbf{r} = r_{110} - r_{1-10}$ ,  $r_{\alpha\beta\gamma}$  is the real component of the complex reflectance  $r$  along the  $[\alpha\beta\gamma]$  crystallographic axis,  $(\Delta \mathbf{r}/\mathbf{r})_t$  is the signal from a pure  $2 \times 1$  terrace,  $(\Delta \mathbf{r}/\mathbf{r})_s$  denotes the RD spectrum from steps, and  $f_t$  ( $f_s$ ) is the fraction of the surface generating the terrace-induced (step-induced) signal. Because the

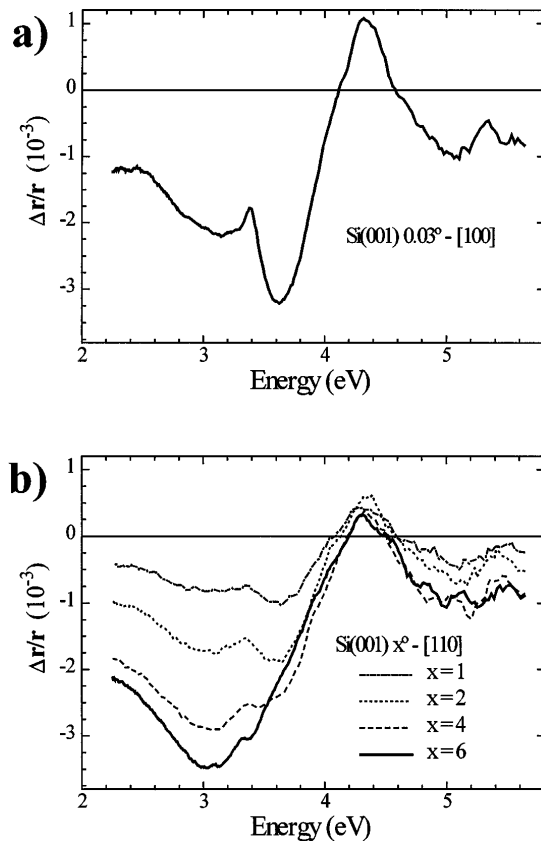


FIG. 2. Reflectance difference spectra for a pure Si(001) terrace and surfaces containing steps. (a) Pure terrace spectrum, from the surface shown in Fig. 1. (b) Spectra of vicinal Si(001) surfaces. The samples were miscut towards [110] by 1°, 2°, 4°, and 6°, respectively. All measurements were performed at room temperature immediately after cleaning.

contributions of equal  $2 \times 1$  and  $1 \times 2$  domains cancel each other,  $f_t$  is the difference between the surface fractions of  $2 \times 1$  and  $1 \times 2$  terraces. Clearly  $f_s$  increases with increasing miscut, but  $f_t$  does as well, because the terrace occupation anisotropy increases with the miscut. A comparison of the spectra in Fig. 2(b) with the terrace spectrum [Fig. 2(a)] scaled by the appropriate  $f_t$  determined from the STM analysis shows that the 3.7 eV feature closely tracks the terrace signal while the 3 eV feature increasingly deviates from the terrace signal with increasing miscut. Figure 3 shows the difference, for each vicinality, between the measured spectra in Fig. 2(b) and the scaled terrace spectra. The resulting curves display a reflectance difference that increases monotonically with miscut, with all of the spectra similar in shape, producing a broad peak at about 3 eV and a very sharp feature at 3.4 eV.

The invariance of the shape of this step-generated RD signal with increasing vicinality is not necessarily expected; while  $S_A$  and  $S_B$  steps form almost exclusively on Si(001) miscut 1° towards [110], for miscuts higher than 3.5° more than 50% of all steps are  $D_B$ -type

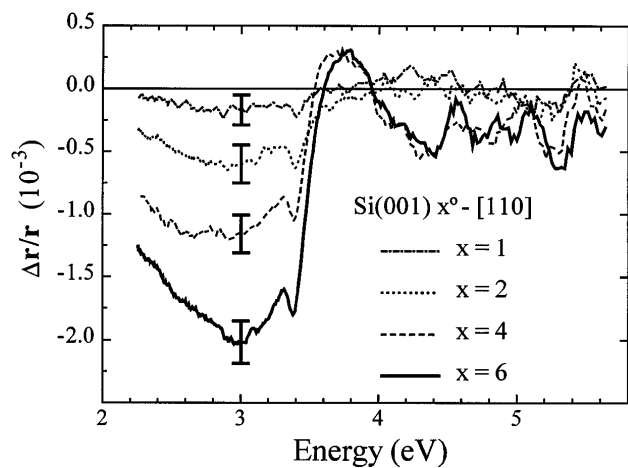


FIG. 3. Step-induced optical anisotropy of vicinal Si(001) shown for different miscut angles towards [110]. The spectra were obtained by subtracting the pure terrace spectrum from Fig. 2(a) multiplied by the measured terrace fraction  $f_t$  (see text) from the measured RD signal for a given vicinality. The error bars are based on the uncertainties in the measured value of the terrace fraction,  $f_t$ .

[14]. However, it is reasonable to assume that only the rebonded  $S_B$  and  $D_B$  steps contribute to the RD signal and that they induce similar RD features [16] because  $S_A$  steps do not disturb the bonding of adjacent terrace dimers and  $S_B$  and  $D_B$  steps rebond equivalently. The linear increase of the combined  $S_B$  and  $D_B$  step densities with vicinality is reflected by the almost linear scaling of the strength of the step-generated spectrum with vicinality (see Fig. 3). The elimination of the areal scaling factors  $f_t$  and  $f_s$  from the spectra in Fig. 2 reveals that the amplitudes per unit area of the step-generated RD signal  $(\Delta r/r)_s$  and of the terrace spectrum  $(\Delta r/r)_t$  are the same within a factor of 2.

These results contradict the common assumption that steps play no role in the optical anisotropy of Si(001), an assumption based upon extrapolation from two previous experiments [6,7]. In one case, the negligible importance of atomic steps on the RD spectrum of As-terminated Si(001) is postulated to be true also for the clean silicon surface despite evidence that arsenic termination dramatically changes the surface electronic structure, eliminating dangling bonds and perhaps altering the rebonding at steps [7]. In the second experiment [6], a negligible step contribution to the RD spectrum is inferred from the small optical anisotropy of Si(113), despite its high step density, and from the similar RD spectra of Si(001) miscut 4° to 10° toward [110]. The Si(113) surface has a different reconstruction [17,18] and is therefore not simply a Si(001) surface with a high step density. Efforts to conclude anything about the effect of steps from 4° to 10° miscut surfaces are meaningless because such surfaces are all already  $D_B$ -step dominated so that the shapes of their RD spectra will not

differ greatly. One must look at the difference between step-dominated and terrace-dominated surfaces to observe more clearly the contribution of steps.

Available calculations of RD spectra, such as recent results for Si(001) with buckled dimers [7], provide, at best, only an approximate fit to our measured spectra, and cannot fit at all the relative heights of the 3 and 3.7 eV features of our terrace spectrum. None of the calculations includes steps or rebonding at steps. In one recent work [9] the authors tentatively attribute the evolution of the RD spectrum with vicinality to step-induced pinning of dimer buckling. Steps pin  $2 \times 2$  or  $c(4 \times 2)$  buckling [19,20] and consequently, according to Ref. [7], would be expected to enhance the 3.7 eV feature with respect to the 3 eV feature. Instead, as the step abundance increases, the opposite occurs. In addition, the perturbation of the local surface structure, charge distribution, and surface energy caused by step rebonding is much greater than that caused by dimer buckling. We conclude that the effect of rebonding at  $S_B$  and  $D_B$  steps must be included in calculations of the optical anisotropy to improve their reliability.

In conclusion, we have unequivocally identified and quantified the step-induced contribution to the optical anisotropy of vicinal Si(001) surfaces. Atomic steps generate a broad RD feature at 3.0 eV. The relative RD signal strengths per unit area of the terrace and step contributions are of the same order of magnitude. We identify the step-induced modification of the optical anisotropy with the rebonding that occurs at  $S_B$  and  $D_B$  steps.

These results confirm what would seem intuitively clear: That atoms at steps, which find themselves in a highly anisotropic environment, contribute to the anisotropy of the optical signal from the surface.

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