μ, the ratio of the intensity of successive subharmonics in the power spectrum studied by M. Feigenbaum [Phys. Lett. 74A, 375 (1979)], who found $\mu \simeq 4\alpha[2(1+\alpha^{-2})]^{1/2}$. The current best values of β and μ are, respectively, 6.618 and 6.557. That β and the "noise-free" parameter μ are related can be made plausible by requiring the ratio of the intensities of the noise-induced power spectra for chaotic transitions at r_n and r_{n+1} to coincide with the ratio of the spectral peaks at corresponding values of the control parameter, and performing a field-theoretic calculation which identifies the former ratio with the ratio of noise levels causing the transition.

Ion Beam Crystallography at the Si(100) Surface

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The geometric structures of the clean 2×1 reconstructed and the hydrogen-stabilized (1×1)2H Si(100) surface have been studied by means of ion channeling and blocking. The latter surface appears to be contracted by 0.08± 0.03 Å and has a surface Debye temperature of ~230 K. For the 2×1 surface it is shown that only the surface dimer models by Appelbaum and Hamann and by Chadi agree well with the backscattering data. The silicon atoms in the very surface are displaced more than 0.45 Å in the surface dimer direction, but those in deeper layers are displaced less than ~0.2 Å.

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A large number of investigations, both theoretical and experimental, have been dedicated to the problem of determining the structure of the clean Si(100) surface. These efforts have yielded structural models for this surface, but have not yet yielded a solution to the problem.[1] The models proposed can be divided into two main categories. First there are models that explain the observed low-energy electron-diffraction (LEED) patterns by assuming missing rows in the surface, leading to an increase in the surface unit cell. All other models attribute the reconstruction to displacements of surface atoms from bulk lattice positions. Recently subsurface displacements have been included in both types of models. The simplest surface-vacancy model has missing rows giving rise to a 2×1 LEED pattern. A more complicated model, proposed by Poppendieck, Gnoc, and Webb,[2] combines missing rows in the two outer layers of the crystal with displacements in the three outer layers. The LEED pattern belonging to this model is $c(4\times2)$, observed in several LEED experiments and with He diffraction.[3] The dimer model is an example of the second category. Surface atoms, having two dangling bonds, form a dimer to lower their energy. In the simplest case, only surface atoms are involved.[4] The conjugated-chain model[5] also features a pairing of rows in the outer crystal layer. Appelbaum and Hamann[6] showed that the energy can be lowered further by displacing subsurface atoms from their bulk positions when a symmetric dimer is formed between Si surface atoms. Chadi calculated that tilting the surface dimer is energetically even more favorable.[7] LEED studies show reasonable agreement with models from both classes, even when these models differ drastically. He diffraction results have confirmed the existence of the $c(4\times2)$ structure, along with $p(2\times2)$ and possibly $c(2\times2)$ regions.[3] Chadi presented arguments in favor of 2×2 reconstructions against a pure 2×1 reconstruction.[8] The 2×2 reconstructions are obtained by a rearrangement of the asymmetric surface dimers, still including subsurface distortions. Photoemission experiments, in which the dispersion of the intrinsic surface state was determined, gave evidence for an asymmetric-dimer model.[9]

Medium-energy ion scattering with the combined effects of channeling and blocking has been shown to be extremely sensitive to surface structural parameters such as atomic displacements and thermal vibrations.[10] It is a quantitative technique and the experimental results are, in general, easily interpreted. We have used this technique to determine the structure of the Si(100)-(1×1)2H surface,[11] to establish some essential structural parameters of the Si(100)-(2×1) reconstructed surface, and to investigate the merits of a variety of structural models proposed in the literature. Our experiments were performed in a stainless-steel UHV scattering chamber with a base pressure in the 10^{-11}-Torr region. The scattering chamber is coupled to a 20–200-keV ion accelerator via a differentially pumped beam line. Backscattered ions are analyzed in a cylindrical electrostatic analyzer with an energy resolution $\Delta E/E$ of 3.6×10^{-3} and an angular resolution of 0.75°. The ions are detected by a Bendix magnetic electron multiplier. The analyzer can be rotated around the target center in steps of 0.0375 deg. The silicon sample, mounted in a precision three-axis goniometer, was cleaned by direct-current heating to 1250 °C and cooling down to room temperature at a cooling rate of ~1 °C/sec. This procedure resulted in a clean and reproducible surface exhibiting a sharp 2×1 LEED pattern. The 1×1 pattern was obtained by exposing the surface to hydrogen at a pressure of 10^{-5} Torr for five minutes, in the vicinity of a hot filament. The ratio of the C to Si Auger peak height was less than 2×10^{-3}. On some samples ion backscattering spectra showed a very small amount of a heavy impurity, probably molybdenum. The amount of this impurity was always less than 0.01 monolayer. It did not influence the observed LEED patterns or the hydrogen adsorption.

The insets in Figs. 1 and 2 show the (010) scattering plane used in the present experiments, perpendicular to the (100) surface. The proton beam can be aligned with a major crystallographic direction such as [102] or [103]. Under such channeling conditions the first atom in each row casts a shadow cone, reducing the hitting probability of atoms further along the row. If this shadowing is perfect the ion beam hits only one atom per row. If the first atom is not on a regular lattice position the second atom in the row can come outside the shadow cone and thus also be visible to the ion beam. By varying the proton-beam energy, the radius of the shadow cone at the position of the second atom in the row can be changed. Now, an ion scattered by an atom in the second layer in the scattering plane can leave the crystal in any direction, except the direction in which its outgoing trajectory is blocked by an atom in the first layer. In this direction there will be a minimum in the yield of ions backscattered from the surface: the surface blocking minimum. If the ion detector is aligned with the direction of the surface blocking minimum, we speak of double alignment. If the detector is looking in a direction in which no blocking occurs, we speak of single alignment. In single alignment we measure the number of atoms per row visible to the ion beam. This number can be obtained by comparing the area of the distinct peak in the energy spectrum of ions backscattered

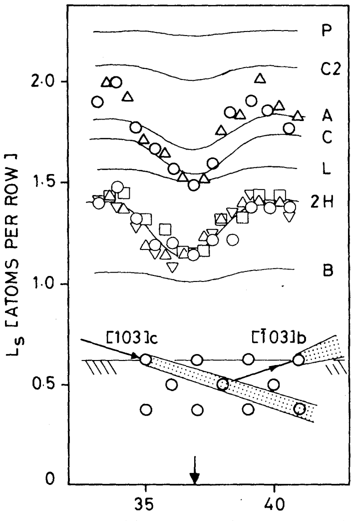

FIG. 1. Blocking minima for the Si(100)-(1× 1)2H surface (lower data) and the Si(100)-(2× 1) surface (upper data). The several full curves are explained in the text. The inset shows the scattering geometry in the (010) plane perpendicular to the surface.

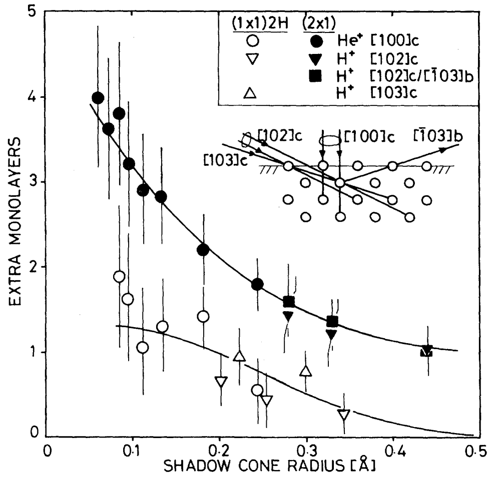

FIG. 2. Number of extra monolayers as a function of shadow-cone radius for the (1× 1)2H surface (open points) and the 2×1 surface (full points). The inset shows the scattering geometries in the (010) plane perpendicular to the surface. The circles are taken from Feldman *et al*. (Ref. 12).

from the surface under channeling conditions to the height of the energy spectrum under random incidence conditions.

Figure 1 shows experimental surface blocking minima obtained with 50-keV protons incident along the [103] channeling direction and exiting around the [$\bar{1}$03] blocking direction. The absolute number of atoms per row contributing to the backscattered yield is plotted versus scattering angle. The lower experimental data were measured on the Si(100)-(1 × 1)2H surface. This blocking minimum appears to be shifted to smaller scattering angles compared to the direction of the bulk blocking axis indicated by the arrow on the horizontal axis. This must be due to an inward relaxation of the Si(100)2H surface. The higher experimental data show the blocking minimum for the clean 2 × 1 surface. The increase in yield with respect to the (1 × 1)2H minimum must be due to atomic displacements in the reconstructed surface. With use of Monte Carlo techniques it is possible to calculate surface blocking minima for any structural model. In Fig. 1 the line labeled "*B*" is the calculated blocking minimum for a surface in which the atoms are not displaced and have a bulk thermal vibration amplitude {0.078 Å[one dimensional (1D), rms], based on a bulk Debye temperature of 543 K}. From the difference between the (1 × 1)2H blocking dip and curve "*B*" the thermal vibration amplitude of the atoms in this surface, or equivalently the surface Debye temperature, can be deduced. The solid line "2H" is a best fit for an inward relaxation of -0.08 ± 0.03 Å of the outer surface layer, and a 1D surface thermal vibration amplitude of 0.18 and 0.11 Å, respectively, in the first and second layer of the crystal, deeper layers being bulklike. This corresponds to a surface Debye temperature of 230 K. Measurements performed at 100 keV in this same geometry and with 50-, 100-, and 150-keV protons channeling in the [102] direction and blocked around the [$\bar{1}$03] outgoing direction are fully consistent with these results.[11] Also, recent measurements by Feldman, Silverman, and Stensgaard[12] can now be fully explained with these parameters. The large enhancement in surface thermal vibrations that we find agrees reasonably well with theoretical results by Ohkuma and Nakamura.[11, 13]

The data obtained for the 2 × 1 surface are compared with calculations for a number of structural models. Because the displacements of surface atoms are very large in these models and hence these atoms do not contribute to shadowing or blocking, their vibrational amplitude is not relevant to the calculations. For the subsurface atoms one expects thermal vibration amplitudes to be bulklike. Therefore all calculations for the 2 × 1 surface have been performed under the assumption of bulklike thermal vibrations. Blocking dip "L" is the result for the dimer model proposed by Levine,[4] in which only surface atoms are displaced to form a dimer. The conjugated chain model[5] would give the same blocking minimum. In these two models the surface atoms are displaced so much that they do not contribute to shadowing or blocking. Still, we find that the single alignment yield for these models is far lower than the measured value. This means that in these models displacements of subsurface atoms must be incorporated in order to find agreement with the data. Any model trying to explain the reconstruction by vacancies or displacements in the very surface only is not consistent with our data. Line "A" is the blocking dip obtained for the Appelbaum and Hamann model.[6] We see that the inclusion of subsurface distortions greatly improves the agreement with our data. This is also true for curve "C," the result for Chadi's asymmetric dimer, which also includes subsurface distortions.[7] Chadi's double-conjugated-chain model[7] (C2) is evidently not in agreement with our data, and neither is the model proposed by Poppendieck, Gnoc, and Webb, "P," which combines multilayer distortions with missing rows to form microfacets on the surface.[2] In this model the displacements in the first three layers are so large that the atoms do not contribute to shadowing or blocking and the effect is merely a large increase in the number of atoms per row. Thus we find that the models proposed by Appelbaum and Hamann and by Chadi agree best with our experimental results, and that all other models can be ruled out. Data obtained at 100 keV in the same scattering geometry, as well as in other geometries, fully confirm this picture. Additional calculations[11] show that the small disagreement between lines "A" and "C" and the data cannot be explained by an enhancement of surface thermal vibrations. On the other hand, we expect that a small increase in static displacements in deeper layers, keeping the models qualitatively the same, may lead to a better agreement with our results. Atoms which are displaced such that they do not fall inside the shadow cone cast by a previous atom in the row contribute to the backscattered yield.

Figure 2 shows the extra number of monolayers visible to the ion beam, i.e., extra with respect to the number calculated for a bulklike surface, versus the radius of the shadow cone at the position of the second atom in the row. The lower data set is for the (1×1)2H surface, the higher for the 2×1 surface. The circles in Fig. 2 are data taken from Feldman, Silverman, and Stensgaard,[12] obtained with 100–2000-keV He^+ channeling in the [100] direction. The other data were measured in the present experiment in the (010) scattering plane with proton energies of 50, 100, and 150 keV. The ion beam was aligned with either the [102] or the [103] direction (see inset in Fig. 2). In both the [100] and the [102] directions one atom per row corresponds to four monolayers. Therefore measurements on the reconstructed surface in these two directions can be compared. Since the [102] direction makes an angle of 50.77° with the [011] and $[0\bar{1}1]$ directions along which dimerization of the surface atoms should mainly occur, the sensitivity to displacements in the direction of the surface dimer is reduced by $\sin 50.77°$. Therefore the shadow-cone radii of the data obtained in the [102] channeling direction on the 2×1 surface are multiplied by $1/\sin 50.77°$ in order to correct for this effect. It is seen that all data fall on a smooth line. When the shadow-cone radius is small the number of extra monolayers visible to the ion beam is large, indicating the presence of subsurface distortions. At a shadow-cone radius of 0.45 Å, one extra monolayer of silicon atoms is still visible both in single and in double alignment. Furthermore, we see that the data level off at one monolayer. This means that one monolayer of silicon must be displaced over a distance of more than 0.45 Å from a bulk lattice position in the direction in which dimerization takes place. Since the displacements in deeper layers are smaller than ~0.2 Å, as can be seen from Fig. 2, we interpret this monolayer to be the outer layer of the Si(100)-(2×1) surface.

In conclusion, we find that the Si(100)-(1×1)2H surface shows an inward relaxation of 0.08 ± 0.03 Å, or $(-6 \pm 3)\%$ of the interplanar distance. The thermal vibration amplitude of this surface is greatly enhanced. The surface Debye temperature is around 230 K, to be compared with the bulk Debye temperature of 543 K. From a comparison of blocking minima calculated for a variety of reconstruction models with our experimental results for the reconstructed surface, we learn that only the models proposed by Appelbaum and Hamann and by Chadi give a satisfactory description of the reconstruction. All models which do not include subsurface distortions are ruled out; neither can Poppendieck's model of a microfacetted surface explain our data. Combining the arguments presented in this Letter and recent experimental results in photoemission,[9] we favor the asymmetric-dimer model. An arrangement of the dimers such that they form a 2×2 rather than a 2×1 symmetry[8] would not influence our calculations on Chadi's model as presented in Fig. 1. In the favored picture of surface dimers accompanied by subsurface distortions the surface atoms must be shifted from bulk lattice positions over a distance larger than 0.45 Å in the direction of the surface dimer, the displacements in deeper layers being less than ~0.2 Å.

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[1] For a recent review on the reconstruction of silicon surfaces, see D. E. Eastman, J. Vac. Sci. Technol. 17, 492 (1980).

[2] T. D. Poppendieck, T. C. Gnoc, and M. B. Webb, Surf. Sci. 75, 287 (1978).

[3] M. J. Cardillo and G. E. Becker, Phys. Rev. Lett. 40, 1148 (1978), and Phys. Rev. B 21, 1497 (1980).

[4] J. D. Levine, Surf. Sci. 34, 90 (1973).

[5] R. Seiwatz, Surf. Sci. 2, 473 (1964); F. Jona, H. D. Shih, A. Ignatiev, D. W. Jepsen, and P. M. Marcus, J. Phys. C 10, L67 (1977).

[6] J. A. Appelbaum and P. R. Hamann, Surf. Sci. 74, 21 (1978).

[7] D. J. Chadi, Phys. Rev. Lett. 43, 43 (1979), and J. Vac. Sci. Technol. 16, 1290 (1979).

[8] D. J. Chadi, to be published.

[9] F. J. Himpsel and D. E. Eastman, J. Vac. Sci. Technol. 16, 1297 (1979), and to be published.

[10] See, for instance, J. F. van der Veen, R. M. Tromp, R. G. Smeenk, and F. W. Saris, Surf. Sci. 82, 468 (1979).

[11] R. M. Tromp, R. G. Smeenk, and F. W. Saris, to be published.

[12] L. C. Feldman, D. J. Silverman, and I. Stensgaard, Nucl. Instrum. Methods 168, 589 (1980).

[13] K. Ohkuma and K. Nakamura, J. Phys. C 12, L835 (1979).