

Atomic steps on Si(100) surfaces

J. A. Martin,^{a)} C. E. Aumann, D. E. Savage, M. C. Tringides, and M. G. Lagally^{b)}
*Department of Metallurgical Engineering and Materials Science Center, University of Wisconsin-Madison,
 Madison, Wisconsin 53706*

W. Moritz and F. Kretschmar
Institut für Kristallographie und Mineralogie, Universität München, München, West Germany

(Received 18 November 1986; accepted 15 December 1986)

It is demonstrated that surface-sensitive electron diffraction measurements of steps on singular, but rough Si(100) surfaces are subject to a multiple-scattering effect that can be misinterpreted as double-atomic-height steps. It is shown that only single-height steps are present. This phenomenon can occur in any surface in which adjacent terraces have different terrace structure factors. Determination of terrace sizes can be influenced; however, they can be accurately made if the diffraction geometry is chosen appropriately. Possible terrace shape effects may also be inherent in the diffraction data.

Steps on surfaces represent a continuing area of fascination because of the variety of chemical, transport, and growth processes that are associated with them. A quantitative determination of step density, average terrace size, terrace size distribution, and edge roughness can be of considerable benefit in specifying the fundamental mechanisms associated with such processes. High-resolution surface-sensitive diffraction is the ideal tool for an evaluation of these quantities. The basic methodology is well developed^{1,2} and involves measuring the diffracted-beam profile as a function of diffraction parameters. Steps on a large number of surfaces have been at least crudely investigated,¹⁻³ with the usual parameters that are determined being the step height on singular, but rough, surfaces and the step height and mean terrace size on vicinal surfaces. It is generally found that surfaces form single-atomic-layer-height steps, with several exceptions.

Si(100) is such an exception. With the diamond structure, the repeat unit in the $\langle 100 \rangle$ direction is actually four atomic layers. Thus, a Si(100) surface can have four different terrace types: adjacent ones are related by a 90° rotation and alternate ones by a translation (screw axis). A schematic view of a vicinal near-(100) Si surface illustrating these features is shown in Fig. 1.

Diffraction from such surfaces can be much more complicated than the simple analysis that is applicable to surfaces in which all terraces are equivalent. Measurements on the vicinal near-(100) Si surface have been published elsewhere.³ We discuss here briefly results on the singular, but rough, Si(100) surface. Such surfaces have a random up-down arrangement of terraces of varying sizes. We make the following observations and draw the consequent conclusions:

(1) Step heights are routinely determined from the periodicity in S_{\perp} of the oscillation in width of angular profiles, where S_{\perp} is the component of the momentum transfer vector normal to the surface. We observe a dependence of this oscillation on the azimuth of the incoming k vector. Apparent double-atomic-height steps are observed in all azimuths except those corresponding to mirror planes for adjacent terraces in the crystal (the $\langle 010 \rangle$ directions). In these azimuths single-atomic-height periodicity is observed, for widely dif-

ferent step densities and by groups in different laboratories, as shown in Fig. 2. The measurements in Fig. 2(a) were made with a high-resolution diffractometer in a configuration where the incident angle was quite small, on a surface on which the average terrace size is of the order of 300 Å. Measurements in Fig. 2(b) were made in a more typical low-energy electron diffraction (LEED) geometry on a surface with a considerably smaller terrace size, of the order of 65 Å.

(2) A dependence of the width of an angular profile on the azimuth of the incident wave vector can be explained only on the basis of effective structure factors of adjacent terraces

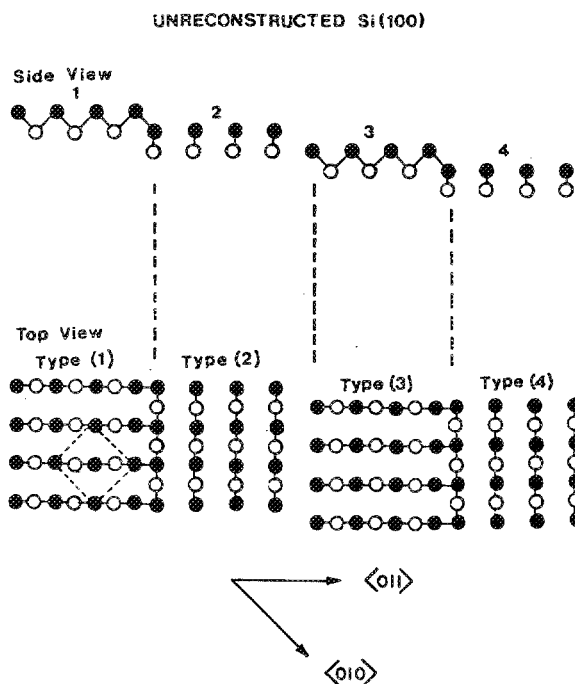


FIG. 1. Schematic illustration of the four possible terminations of Si(100). The surface is shown unreconstructed for ease of observation. The number of terrace types does not change if the surface is allowed to reconstruct. ● Surface atoms; ○ second-layer atoms; --- bonds. The dashed square is a unit mesh.

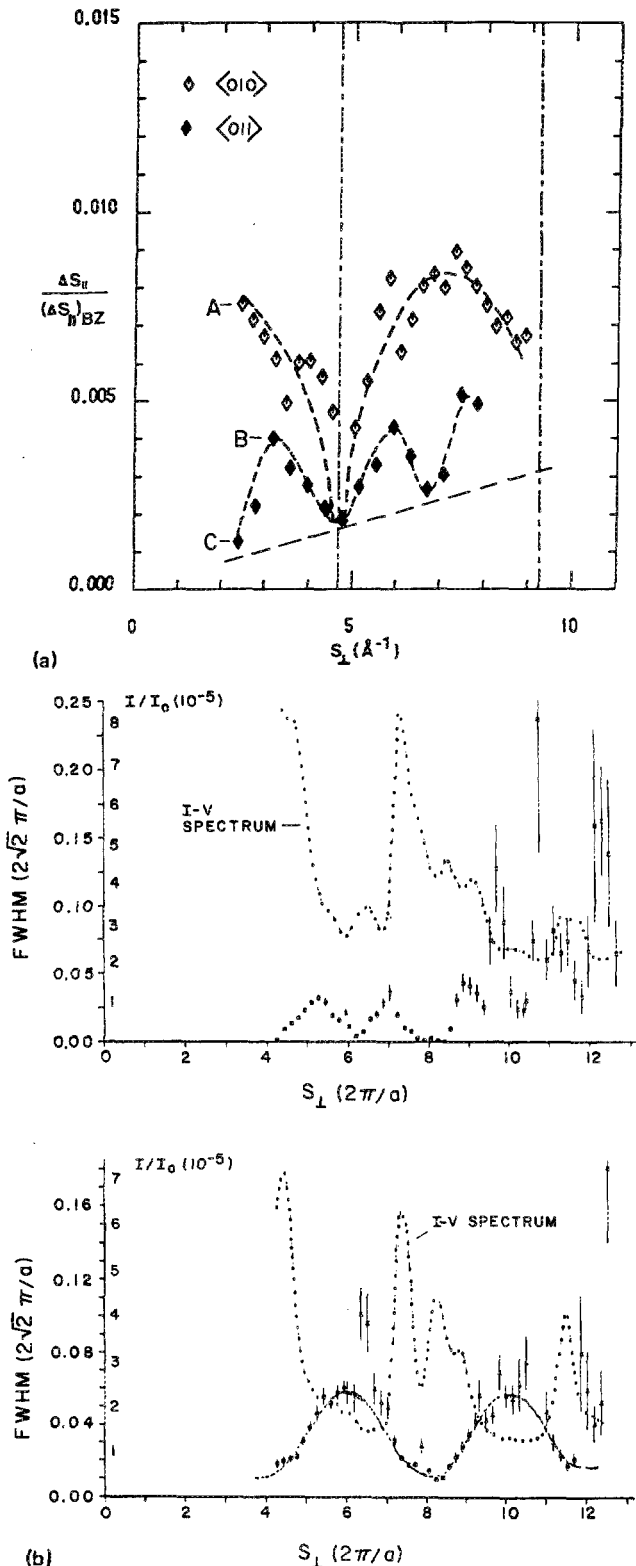


FIG. 2. I.EED measurements from singular, but rough, Si(100) surfaces at different azimuths. (a) Full width at half-maximum of the (00) beam vs perpendicular component of the momentum transfer for a surface with average terrace width of ~ 300 Å, with incident beam along two azimuths. Points A, B, and C refer to profiles in Fig. 3. Vertical dash-dot lines are in-phase conditions for a surface having single-atomic-height steps. Dashed line is the instrument response broadening. (b) Data of F. Kretschmar on the (00) beam for a surface with average terrace width of ~ 65 Å. Upper: measurement in the $\langle h, 0 \rangle$ scattering plane showing double-step-height oscillations. Lower: measurement in $\langle h, h \rangle$ plane, showing single-step-height oscillations. I - V profiles for the two situations are also shown.

that differ for most azimuths but become the same for mirror plane azimuths. If the structure factor of adjacent terraces is different, an apparent double-atomic-step-height oscillation will be observed even if only single-atomic-height steps exist. Such a behavior of terrace structure factors can readily be explained on the basis of multiple scattering. In the mirror plane directions, the structure factors for the specular beam become identical for symmetry reasons. A complete exposition of this phenomenon is presented elsewhere.⁴

(3) As a general rule, terrace size distributions can be accurately obtained by fitting the angular profile of diffracted beams at their out-of-phase conditions,² taking account of the instrument response function. The assumption in such fits is that the structure factors of all terraces are the same. If the terrace structure factors differ, errors are introduced in the terrace size determination. This can be seen qualitatively from Fig. 2, which shows that the angular profiles measured in azimuths where the structure factors are the same and not the same attain different maximum widths. If oscillations corresponding to double-height steps are observed, one would expect, on the basis of Fig. 2, to deduce a terrace size that is greater than the correct size. Fits to angular profiles, shown in Fig. 3, provide a more quantitative demonstration. The "correct" terrace size determination is shown in Fig. 3(a); Figs. 3(b) and 3(c) show situations where multiple scattering affects the terrace structure factor. It should be noted that in all cases a smooth profile is obtained, i.e., the multiple scattering does not destroy the smooth shape.⁴

(4) Based on our measurements, we can determine an average terrace size of 320 Å on the Si(100) surface shown in Fig. 2(a), with a geometric distribution of sizes.⁵ A geometric size distribution has its highest probability at small sizes. We picture this singular, but rough, Si(100) surface as consisting of a random arrangement of terraces with an average size of 320 Å, separated by single-height steps.

(5) Although we cannot prove from these results that double-height steps could not also exist on singular Si(100) surfaces under some sample preparation conditions, we do not observe them in two independent studies and can, in fact, put an upper limit of $\sim 20\%$ of all steps on the concentration of double steps for surfaces that have been sputter-etched and subsequently annealed at temperatures from 700 to 1100 °C, for 10 min each, resulting in average terrace sizes of ~ 100 to ~ 1000 Å. Our conclusions are in disagreement with those of Henzler and Clabes,⁶ who observed double-height steps at anneal temperatures above 700 °C. We have demonstrated here how oscillations suggesting double-height steps will occur at most azimuths. We also cannot exclude some difference in total area of terraces of different type (e.g., types 1 and 2 in Fig. 1). If the areas are different, the terrace structure factors will be different and a double periodicity will result that is suggestive of double-height steps. However, we cannot explain the azimuthal dependence in this way.

(6) Finally, there is an additional, suggestive aspect to the width oscillation curves in Fig. 2. The behavior of the widths of the curves in two different azimuths implies that the terraces may have preferentially square shapes with $\langle 110 \rangle$ step edge orientations. Such a terrace shape anisotropy would

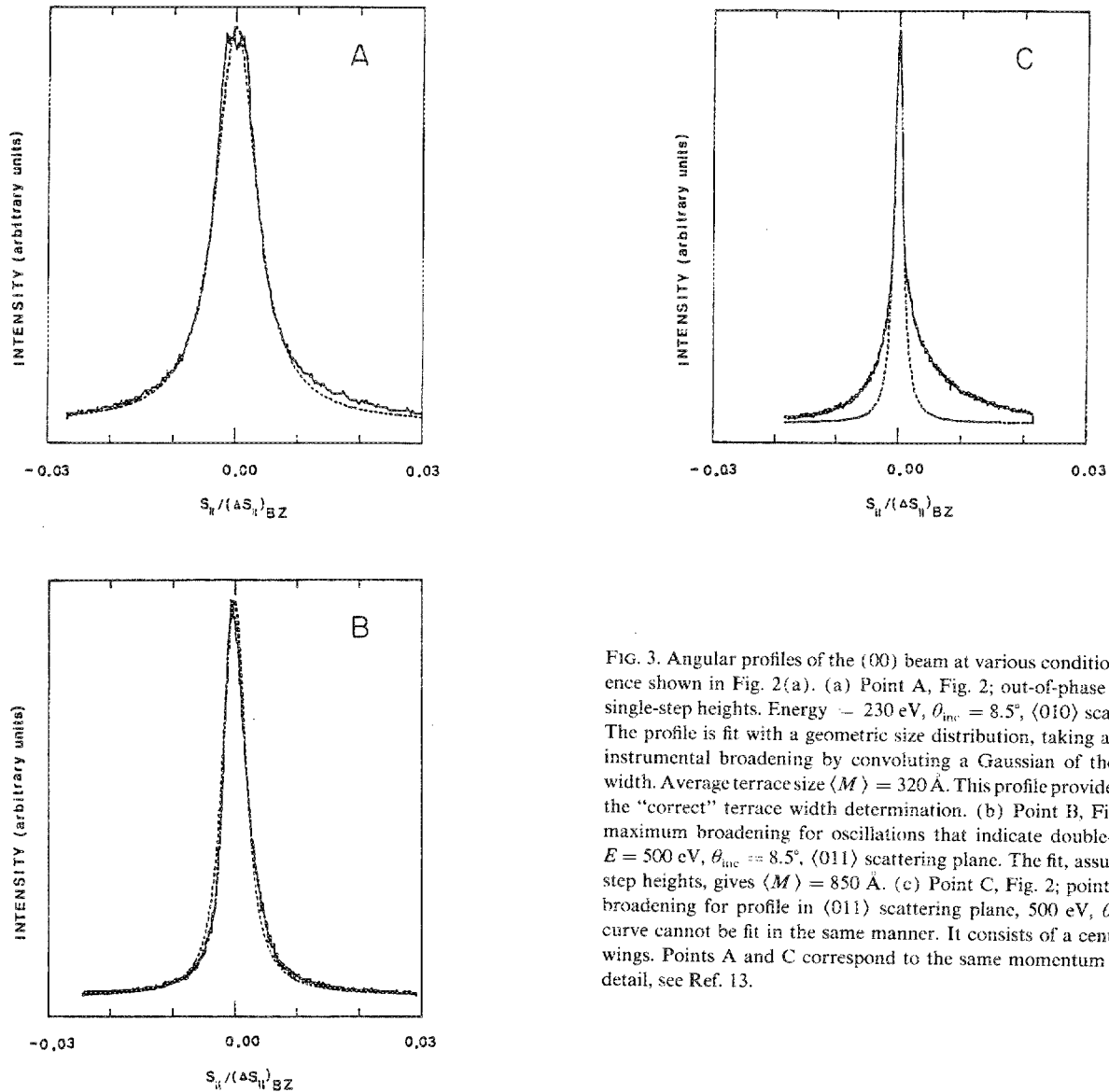


FIG. 3. Angular profiles of the (00) beam at various conditions of interference shown in Fig. 2(a). (a) Point A, Fig. 2; out-of-phase condition for single-step heights. Energy = 230 eV, $\theta_{\text{inc}} = 8.5^\circ$, $\langle 010 \rangle$ scattering plane. The profile is fit with a geometric size distribution, taking account of the instrumental broadening by convoluting a Gaussian of the appropriate width. Average terrace size $\langle M \rangle = 320 \text{ \AA}$. This profile provides the basis for the "correct" terrace width determination. (b) Point B, Fig. 2; point of maximum broadening for oscillations that indicate double-step heights. $E = 500 \text{ eV}$, $\theta_{\text{inc}} = 8.5^\circ$, $\langle 011 \rangle$ scattering plane. The fit, assuming double-step heights, gives $\langle M \rangle = 850 \text{ \AA}$. (c) Point C, Fig. 2; point of minimum broadening for profile in $\langle 011 \rangle$ scattering plane, 500 eV, $\theta_{\text{inc}} = 6^\circ$. The curve cannot be fit in the same manner. It consists of a central peak and wings. Points A and C correspond to the same momentum transfer. For detail, see Ref. 13.

require that step edges with unfavorable and favorable bond arrangements⁷ must be equally prevalent. The shape of the terrace will affect part of the intensity distribution (the diffuse wings) and hence the "width" one measures for the complete profile. Details will be presented elsewhere.⁴

Several other results in the literature appear to be related to the phenomenon we have described. It has recently been observed⁸ that two periods are obtained in reflection high-energy electron diffraction (RHEED) intensity oscillations during molecular-beam epitaxy (MBE) of Si(100), depending on the azimuth. This result is directly explained by our study, the growth oscillation period corresponding to our step periodicity for the two azimuths $\langle 010 \rangle$ and $\langle 011 \rangle$, without invoking the explanation of anisotropic oblong islands suggested by the authors. The only requirement is that the beam size in this study be small (of the order of an average terrace width). This is, in fact, the case,⁸ and is verified by a second study by the same authors,⁹ which shows that as the angle of incidence is made very grazing, with the incident beam not along $\langle 010 \rangle$ the growth oscillation period goes

from that expected for double to that for single-height steps. This can be explained by recognizing that at very grazing angles the ellipse formed by the beam on the surface gets quite large and can then cover more than one terrace, resulting in an intensity oscillation period suggesting single-height steps, while at less grazing angles the beam covers only of the order of one terrace, and the period of the intensity oscillation corresponds to double-height steps. In the $\langle 010 \rangle$ azimuth, it makes no difference what the angle of incidence is, in accordance with our suggestion.¹⁰ The same argument can be applied to the study of Aarts *et al.*¹¹ Their beam diameter is considerably larger¹² than that of Sakamoto,⁸ so that under all conditions more than one terrace is illuminated. We thus expect that a growth oscillation period corresponding to single-atomic-height steps will be observed at all conditions.

We conclude that in surfaces in which adjacent terraces have different symmetry, diffraction results on growth, finite-size effects, and perhaps even equilibrium atomic positions must be interpreted carefully,⁴ because the structure

factors of these terraces can be quite different due to multiple scattering. However, once this is taken into account, accurate information on finite-size effects and crystal growth can be readily extracted.¹³

ACKNOWLEDGMENTS

This research was supported by NSF, Solid State Chemistry Program, Grant No. DMR 83-18601, and in part by U.S.-Spain Joint Committee for Scientific and Technological Cooperation, U.S. Department of State. W. Moritz was a visiting professor at the University of Wisconsin during the performance of much of this work.

^{a)} Present address: Los Alamos National Laboratory, Los Alamos, NM.

^{b)} To whom correspondence should be addressed.

¹M. Henzler, in *Electron Spectroscopy for Surface Analysis*, edited by H. Ibach (Springer, Berlin, 1977).

²M. G. Lagally, in *Chemistry and Physics of Solid Surfaces*, edited by R. Vanselow (Springer, Heidelberg, 1982), Vol. IV; in *Methods of Experi-*

mental Physics: Surfaces, edited by R. L. Park and M. G. Lagally (Academic, Orlando, 1985).

³D. Saloner, J. A. Martin, M. C. Tringides, D. E. Savage, C. E. Aumann, and M. G. Lagally, *J. Appl. Phys.* **61**, 2884 (1987).

⁴W. Moritz, F. Kretschmar, J. A. Martin, and M. G. Lagally (in preparation). A summary was presented at the meeting of the German Physical Society, March, 1986.

⁵T. M. Lu and M. G. Lagally, *Surf. Sci.* **120**, 47 (1982).

⁶M. Henzler and J. Clabes, in *Proceedings of the 2nd International Conference on Solid Surfaces*, 1974; *Jpn. J. Appl. Phys. Suppl. 2, Part 2*, 389 (1984).

⁷D. E. Aspnes and J. Ihm, *Phys. Rev. Lett.* **57**, 3054 (1986).

⁸T. Sakamoto, N. J. Kawai, T. Nakagawa, K. Ohta, and T. Kojima, *Appl. Phys. Lett.* **47**, 617 (1985).

⁹T. Sakamoto, T. Kawamura, and G. Hashiguchi, *Appl. Phys. Lett.* **48**, 1612 (1986).

¹⁰Greater detail is provided in J. A. Martin, Ph.D. dissertation, University of Wisconsin-Madison, 1986.

¹¹J. Aarts, W. M. Gerits, and P. K. Larsen, *Appl. Phys. Lett.* **48**, 931 (1986).

¹²P. K. Larsen (personal communication).

¹³M. C. Tringides, C. E. Aumann, D. E. Savage, and M. G. Lagally (in preparation).