Differential Reflectivity of Si(111)2×1 Surface with Polarized Light: A Test for Surface Structure

P. Chiaradia, A. Cricenti, and S. Selci
Dipartimento di Fisica, Università di Roma "La Sapienza," Roma, Italy, and
Gruppo Nazionale di Struttura della Materia del Consiglio Nazionale delle Richerche, Roma, Italy

and

G. Chiarotti
Dipartimento di Fisica, II Università di Roma, Roma, Italy, and
Istituto di Struttura della Materia del Consiglio Nazionale delle Richerche, Roma, Italy
(Received 2 December 1983)

The polarization dependence of the differential reflectivity of the Si(111)2×1 single-domain surface has been studied experimentally. A marked anisotropy of the optical peak at 0.45 eV associated with dangling bonds is observed with maximum absorption along [0$\bar{1}$1] directions. The chain model is in good agreement with the experimental result.

PACS numbers: 78.20.Dj, 68.20.+t

The structure of the Si(111)2×1 surface has been the subject of an extensive investigation aimed at providing a model for the surface reconstruction. Until the end of the 1970's, there was general agreement that the "buckling model,"[1] with alternate rows of atoms along [0$\bar{1}$1] directions displaced up and down, was the most suited for explaining the various experimental results. The only controversial point was the inconsistency of surface bands, as observed by angle-integrated photoemission,[2] with the optical gap obtained in reflectivity experiments.[3] Angle-resolved photoemission,[4,5] while settling this controversy, showed that the dispersion of the filled dangling-bond band was considerably larger than predicted by the buckling model with a reasonable choice of surface parameters. For this reason, Pandey proposed a new structure[6] consisting of zig-zag chains of π-bonded atoms in [0$\bar{1}$1] directions with dangling bonds at nearest-neighbor positions, thus explaining the dispersion observed in photoemission. Moreover, adjacent chains are well separated (~ 6.7 Å) giving a marked anisotropy of the surface. In spite of the complexity of the rearrangement, the chain model is favored by a total-energy calculation[7] and agrees with other experimental data,[3,8] except dynamical low-energy electron-diffraction (LEED) analysis.[9]

The different symmetry properties of the buckling and the chain models suggest that measurements of surface reflectivity with polarized light can give unambiguous support to one of these models. Earlier experiments with unpolarized light and multidomain Si(111)2×1 surfaces both in multiple internal[3] and external[10] configurations have shown a narrow asymmetric peak at 0.45 eV associated with dangling-bond states. A detailed analysis by Del Sole and Selloni[11] shows in the case of the chain model a very strong anisotropy (ratio 1:0) for the 0.45-eV peak with a maximum of $\Delta R/R$ when the electric field is in the direction of the chains (y direction). On the contrary, the buckling model predicts[12] a maximum of $\Delta R/R$ for x polarization, the anisotropy being in the ratio 1:3. This result can be explained intuitively, since in the buckling model the charge transfer occurs between up and down atoms, i.e., along a direction at 30° with the x axis.

In this Letter we present experimental results that strongly favor the chain model or any model with a marked anisotropy in the y direction.

The experiment consists of shining a polarized beam of monochromatic light at normal incidence onto a single-domain surface of Si(111)2×1, cleaved under UHV conditions (2×10^{-10} Torr), along a [$\bar{2}$11] direction, by the double wedge–double notch technique. The optical apparatus was coupled to the UHV chamber by a CaF_2 window tested for optical isotropy. The details of the method and the apparatus will be published elsewhere. The reflectivity R for the clean and oxidized surfaces was measured and the value of $\Delta R/R$, essentially proportional to the imaginary part of the surface dielectric function,[13] recorded. The overall stability of $\Delta R/R$ was typically of the order of 10^{-3} over a few hours.

The dependence of $\Delta R/R$ for x (filled circles) and y (open circles) polarizations is shown in Fig. 1 as a function of the energy of the photons in the range 0.3 to 0.7 eV for a single-domain surface. A sketch of the LEED pattern with the relevant crys-

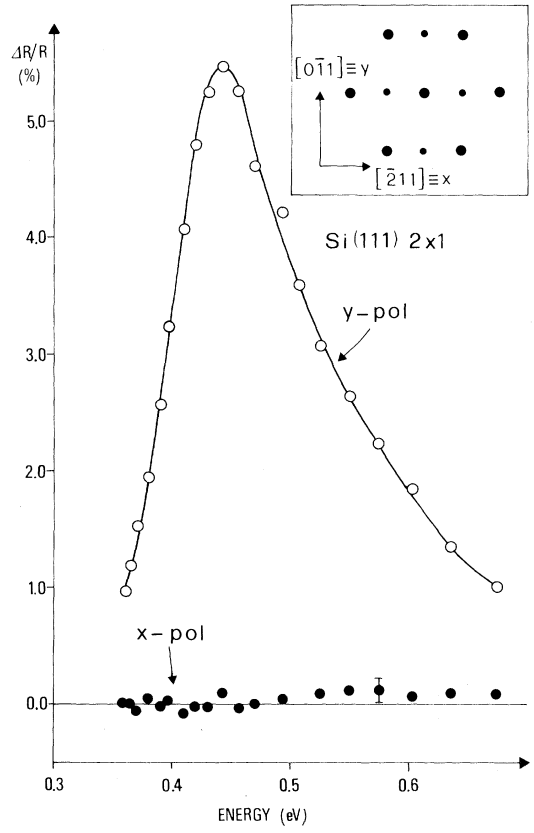

FIG. 1. Differential reflectivity spectra of a Si(111)2×1 single domain surface, for light polarizations along the [$\bar{2}$11] and [0$\bar{1}$1] directions (curves labeled x and y, respectively). The error bar of $\pm 1\times 10^{-3}$ is also shown. The inset represents a sketch of the LEED pattern (with integer and half-order spots) and the main crystallographic directions in the (111) plane.

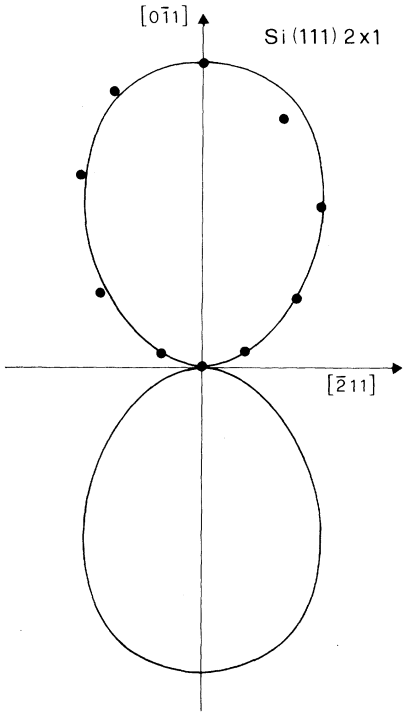

FIG. 2. Polar plot of $\Delta R/R$ at maximum (0.45 eV) vs polarization direction in the (111) plane. The solid curve is a $\cos^2$ fitting to the data points.

tallographic directions is shown in the inset of Fig. 1. A 2×1 single domain was present over the whole sample, whose dimensions were approximately 12×4 mm. Only at the edges were traces of the two other domains visible. Figure 1 shows a dramatic dependence of $\Delta R/R$ on the polarization direction, indicating a strong optical anisotropy in the energy range where dangling bonds contribute to the optical transitions. Measurements with intermediate polarizations show that the $\Delta R/R$ curves continuously change retaining the same shape. Previously reported results with "unpolarized" light[10] are close to the average value of $\Delta R/R$ for x and y polarizations.

The dependence of the peak intensity upon the various polarization directions in the (111) plane is plotted in Fig. 2. Data relative to samples with more than one domain are consistent with the results reported here.[14]

The results of Figs. 1 and 2 are apparently at variance with those reported by Assmann and Mönch[15] from surface photoconductivity and photovoltage, showing a different polarization dependence. It should be noted, however, that the above results were obtained in the extreme wings of the reported photoconductivity and photovoltage peaks, namely, at 0.33 eV (maximum effect for x polarization) and 0.60 eV (isotropy). Moreover the portion of the curves around 0.60 eV does not depend upon oxidation so that it is probably unrelated to surface effects. Further measurements of the full spectrum as well as an accurate analysis of the multidomain structure are necessary to make a comparison with present results. We believe that conclusions drawn previously from the polarization results of Ref. 15 concerning models for surface reconstruction were premature.

Comparison of present results with angle-resolved photoemission data yields a consistent picture of the surface band structure in the two-dimensional Brillouin zone, based solely on experimental results and symmetry considerations. Angle-resolved photoemission shows[4,5] that the

surface band has a large dispersion along ΓJ and a negligible one along the $\Gamma J'$ and JK directions. The initial states of the optical transition at 0.45 eV should then occur along JK; otherwise the final states could not be above the Fermi level. On the other hand, dynamical LEED analysis has shown[16] the existence of a mirror plane perpendicular to the surface along the x direction. Wave functions for the initial and final states should then have opposite parity in order to explain the observed polarization dependence.[11] The only model that encompasses all such features is at present Pandey's chain model which, being almost one-dimensional, accounts for the observed marked anisotropy.

Finally we should mention that a possible source of strong anisotropy could in principle be a system of steps perpendicular to the direction of cleavage. However, given the small density of surface atoms in step position, the y polarization curve reported in Fig. 1 could only be explained by an unreasonably large optical cross section, of the order of 10^{-14} cm^2.

The authors wish to acknowledge useful conversations with R. Del Sole and A. Selloni.

[1]D. Haneman, Phys. Rev. 121, 1093 (1961).

[2]D. E. Eastman and W. D. Grobman, Phys. Rev. Lett. 28, 1378 (1972); L. F. Wagner and W. E. Spicer, Phys. Rev. Lett. 28, 1381 (1972).

[3]G. Chiarotti, S. Nannarone, R. Pastore, and P. Chiaradia, Phys. Rev. B 4, 3398 (1971).

[4]F. J. Himpsel, P. Heimann, and D. E. Eastman, Phys. Rev. B 24, 2003 (1981).

[5]R. I. G. Uhrberg, G. V. Hansson, J. M. Nicholls, and S. A. Flödstrom, Phys. Rev. Lett. 48, 1032 (1982).

[6]K. C. Pandey, Phys. Rev. Lett. 47, 1913 (1981).

[7]K. C. Pandey, Phys. Rev. Lett. 49, 223 (1982); J. Northrup and Marvin L. Cohen, Phys. Rev. Lett. 49, 1349 (1982).

[8]R. M. Tromp, L. Smit, and J. F. Van der Veen, Phys. Rev. Lett. 51, 1672 (1983).

[9]R. Feder, Solid State Commun. 45, 51 (1983); F. Jona and P. M. Marcus, Bull. Am. Phys. Soc. 28, 416 (1983).

[10]P. Chiaradia, G. Chiarotti, S. Nannarone, and P. Sassaroli, Solid State Commun. 26, 813 (1978).

[11]R. Del Sole and A. Selloni, to be published.

[12]D. J. Chadi and R. Del Sole, J. Vac. Sci. Technol. 21, 319 (1982).

[13]J. D. E. McIntyre and D. E. Aspnes, Surf. Sci. 24, 471 (1971).

[14]A careful analysis of the domain structure over the entire illuminated region is necessary in order to obtain unambiguous results. For example, when two domains oriented at 60° with respect to the one shown in the inset of Fig. 1 are present with equal weight a maximum of $\Delta R/R$ for x polarization is found with a ratio 3:1, as predicted by straightforward geometrical considerations.

[15]J. Assmann and W. Mönch, Surf. Sci. 99, 34 (1980).

[16]R. Feder, W. Mönch, and P. P. Auer, J. Phys. C 12, L179 (1979).