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Electronic structure of Rb-adsorbed Si(100) surfaces studied with angle-resolved photoemission

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Angle-resolved photoemission has been used to study Rb adsorption on Si(100)2×1 at room temperature. The development of the valence-band spectra with increasing Rb coverage and the dispersions of the surface-state bands of the saturated Si(100)2×1-Rb surface are reported. Similar to the adsorption of other alkali-metal atoms on the Si(100) surface a strong surface-state peak appears at the Fermi level in the initial stage of Rb deposition. This surface-state peak is a result of partial occupation of a normally empty surface band. The onset of occupation leads to an abrupt upward shift of the Fermi-level pinning position. For the room-temperature saturation coverage, two surface states were observed in the valence-band spectra with energies of -0.55 and -1.55 eV relative to the Fermi level at normal emission. The dispersions of these two surface-state bands were determined along the [010] azimuthal direction of the two-domain 2×1 reconstructed surface. The surface electronic structures of the different Si(100)2×1-alkali metal (Na, K, Rb, and Cs) surfaces are also compared in the paper. [S0163-1829(97)00312-3]

I. INTRODUCTION

The adsorption of alkali-metal (AM) atoms on semiconductor surfaces has attracted a widespread interest in recent years.[1] The simple electronic structure and the chemically active nature make the knowledge of this adsorption system regarded as the basis for understanding fundamental physics and important technological applications. As a prototype system, AM adsorption on Si(100)2×1 surfaces has been studied extensively both experimentally and theoretically. Nevertheless, the general conclusions about the development of the atomic and electronic structure as a function of coverage is still controversial. Basic questions such as the room-temperature (RT) saturation coverage, the metallization of the substrate and/or the overlayer and the nature of the chemical bond between the AM atoms and the substrate are still under debate.

When the Si(100)2×1 surface is saturated by AM atoms at RT the 2×1 periodicity remains. In an early study of the Si(100)2×1-Cs system Levine[2] proposed a one-dimensional AM chain model. In this model Cs atoms form linear chains on top of the Si dimer rows which results in a 0.5-ML coverage [1 ML is defined as 6.78×10^{14} atoms per cm^2, i.e., the surface atomic density of a truncated Si(100)1×1 surface]. Half a monolayer coverage of alkali-metal atoms should result in a metallic surface with a half-filled π^* band since one valence-band electron is added to each 2×1 surface unit cell. Later on, Enta, Abukawa, Kono, and co-workers reported, from their angle-resolved photoemission[3,4] (ARPES) studies, that the surface was semiconducting and from x-ray photoelectron diffraction[5,6] studies on the Si(100)2×1-K and 2×1-Cs surfaces the so-called double-layer model was proposed. In this model the AM atoms are located above the third layer Si atoms. Half a monolayer of the AM atoms adsorbs above the dimer rows on the so-called ‘‘pedestal’’ site while the other half monolayer adsorbs between the dimer rows slightly lower than the pedestal site at what is called the ‘‘valley-bridge’’ site. The resulting total AM coverage is thus 1 ML. The double-layer model has been supported by several subsequent experimental and theoretical studies.[7–13] However, different views concerning, for instance, the RT saturation coverage and the surface metallization still exist in several papers on the AM-Si(100) system.[14–24]

In the past, extensive investigations have been performed of Na, K, and Cs adsorption on the Si(100)2×1 surface, while very little effort has been made to study the Rb/Si(100)2×1 system. Many similarities were found for the Si(100)2×1-Na and the 2×1-K surfaces. For example, two dominating surface-state bands were observed by angle-resolved photoemission.[25–27] This was in agreement with theoretical band-structure calculations[12,13] which report two completely filled surface state bands corresponding to two hybrid states formed by the Si dangling bonds and the AM valence electrons. Si 2p core-level spectroscopy shows one prominent surface shifted component for both Na and K which points to a similar atomic structure, i.e., the asymmetric dimers of the clean Si(100) surface become symmetric after the alkali-metal adsorption.[24,28] However, the photoemission results from the Si(100)2×1-Cs surface give a different picture compared to the 2×1-Na and 2×1-K surfaces, e.g., the surface band structure looks somewhat different;[25] a pair of surface components is found in the Si 2p spectra indicating the presence of asymmetric Si dimers;[29] a strong metallic character is observed as evidenced by a finite emission at the Fermi level and by the large singularity index

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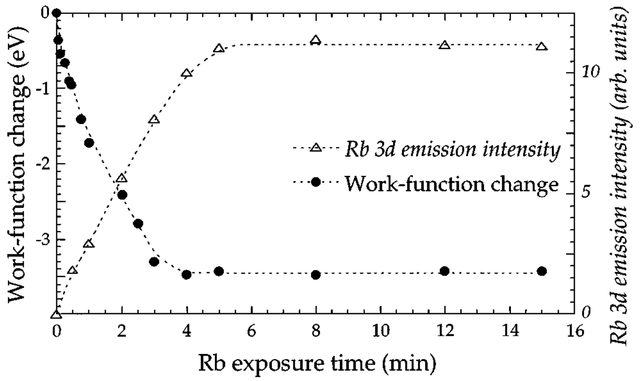

FIG. 1. Solid dots correspond to the measured shifts in the work function, ΔΦ, for the Si(100)2×1 surface as a function of the Rb exposure time. Open triangles represent the integrated emission intensities from the Rb $3d$ spectra.

(Doniach-Šunjić line shape) needed in order to fit the strong asymmetric tail in both the Si $2p$ and the Cs $4d$ core-level spectra.[29] In the present study we concentrate on the Rb/Si(100)2×1 system which is the ''missing link'' between the 2×1-K and 2×1-Cs surfaces. A series of normal-emission valence-band spectra for different Rb coverages are presented. The dispersions of the surface-state bands of the saturated Si(100)2×1-Rb surface are presented and they are compared with the surface band dispersions of the other 2×1-AM surfaces.

II. EXPERIMENTAL DETAILS

The experiment was performed at the toroidal grating monochromator beam line[30] at the MAX synchrotron radiation facility in Lund, Sweden. An angle-resolving hemispherical analyzer (VSW) was used for collecting and analyzing the photoelectrons. The base pressure of the ARPES chamber was less than 1×10^{-10} torr. An n-type, mirror polished, Si(100) single crystal (ρ=2 Ω cm, P) was preoxidized using an etching method[31] and cleaned *in situ* by stepwise heating up to ~900 °C to remove the oxide layers until no trace of contamination was present in the photoemission spectra and a sharp two-domain 2×1 low-energy-electron-diffraction (LEED) pattern was observed. Rubidium was evaporated from a well outgassed getter source (SAES Getters) onto the samples at room temperature. The pressure increased less than 3×10^{-10} Torr during the exposures. The Rb atoms were always deposited onto a fresh Si(100)2×1 surface obtained by annealing at ~900 °C for 30 sec. The Rb exposures were repeated several times with excellent reproducibility of the data.

III. RESULTS AND DISCUSSION

The solid dots in Fig. 1 correspond to the measured shifts in the work function, ΔΦ, as a function of the rubidium exposure time at room temperature. These data were derived by measuring the changes in the cutoff position of the photoemission spectra. The decrease in ΔΦ is quite linear up to an exposure time of 3 min. After an exposure time of 4 min or more ΔΦ has a value of ~−3.44 eV. A two-domain 2×3 LEED pattern was observed at ΔΦ~−2.41 eV but, otherwise, 2×1 LEED patterns were observed for the other coverages. The open triangles present the measured relation between the integrated Rb $3d$ emission intensities and the exposure time. As shown in the figure, a saturation coverage was obtained on the surface after ~5-min Rb exposure.

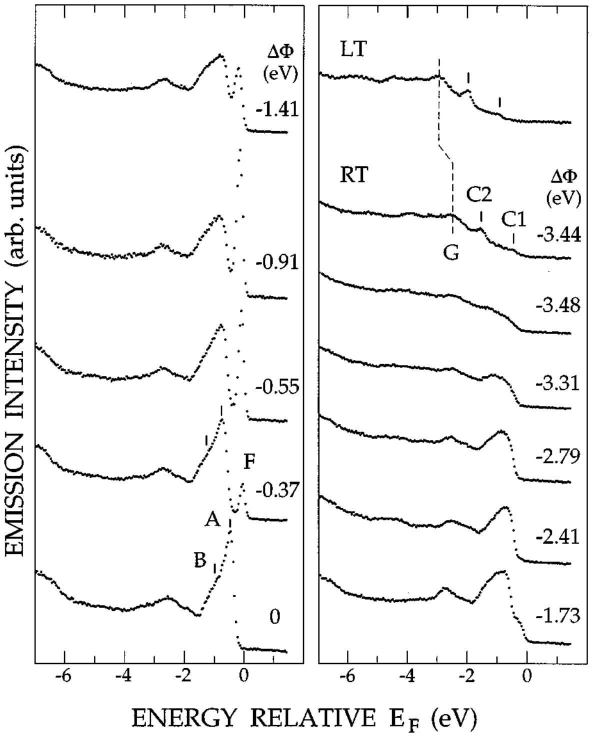

FIG. 2. Normal-emission valence-band spectra recorded from Si(100) for increasing Rb coverage. The coverage was controlled indirectly by measuring the work-function change. The uppermost spectrum in the right panel was recorded from the same surface as the one labeled ΔΦ=−3.44 eV but after the sample had been cooled down to ~120 K.

Angle-resolved normal-emission spectra of the Si(100) valence band recorded with a photon energy of 21.2 eV are shown in Fig. 2 for increasing Rb depositions. The lowermost spectrum was recorded on the clean Si(100)2×1 surface. The spectra obtained for the initial stages of Rb adsorption are similar to the corresponding spectra for the other AM adsorbates. A clear peak, F, appeared at the Fermi level for a very small Rb exposure, and it was accompanied by an abrupt energy shift of the spectrum by ~0.27 eV toward higher binding energies. The intensity of peak F reached its maximum for ΔΦ~−0.91 eV. A spectrum recorded from that surface at an emission angle of 33° along the [010] direction, which corresponds to the J' point for the F state, is shown in Fig. 3. In addition to the F state, the surface-state features of the clean surface, i.e., two surface structures A and B corresponding to the dangling-bond states and a surface resonance D corresponding to a back-bond state, were also present in the spectrum at this small Rb coverage. The appearance of the F peak has been interpreted as due to a partial occupation of an empty surface band by the outermost s electron of the AM atoms.[24] The initial population of the empty band leads to the abrupt shift of the Fermi-level pinning position. A similar phenomenon has been observed on clean, highly n-doped, samples where the F state is popu-

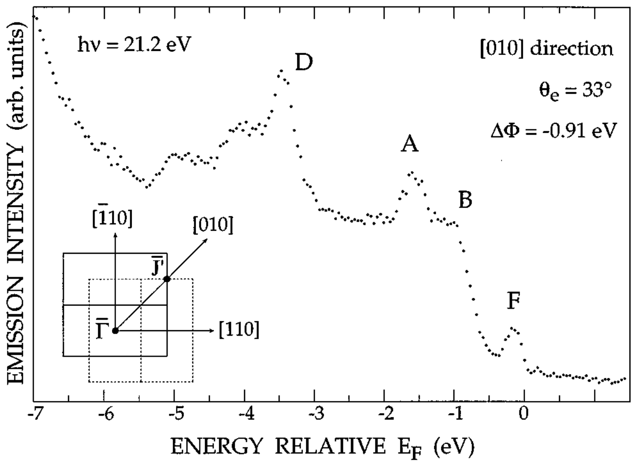

FIG. 3. Spectrum recorded on the surface for $\Delta\Phi=-0.91$ eV at an emission angle of 33° along the [010] direction which corresponds to the J' point for the F state.

lated by the extra doping electrons.[32] Surface band-structure calculations by Northrup[33] confirmed the existence of an empty surface-state band with minima at $k_{\|}$ points corresponding to the Γ and J' points of a 2×1 surface Brillouin zone which is in agreement with the experimental finding for the clean Si(100) surface and for the Si(100) surface with a small amount of AM atoms. The empty band on the clean Si(100)2×1 surface has also been observed in an earlier inverse photoemission study.[34] Recent studies of the alkali promotion effect of O_2 sticking on semiconductor surfaces[35] as well as low-energy ion scattering studies[36–38] have reported an ionic bonding for K and Cs atoms adsorbed on Si(100) and Ge(100) at small coverages. The observations of ionic bonding are consistent with the presence of the F state in the valence-band spectra and the explanation of F as resulting from a donation of the outermost s electron of the AM atoms to an empty Si-derived dangling-bond band.

Further adsorption of Rb results in a lower intensity of the F peak. A mixed two-domain 2×3 and 2×1 LEED pattern was observed for $\Delta\Phi$ around -2.4 eV. As the Rb coverage increased the spectra became broader and it was difficult to discern the different structures. Close to RT saturation coverage, two Rb-induced states $C1$ and $C2$ started to develop and became clearly resolved at RT saturation coverage with energies of -0.55 and -1.55 eV relative to the Fermi level, respectively. Structure G at an energy of ~-2.55 eV could be identified as a bulk transition by using different photon energies. A good two-domain 2×1 LEED pattern was observed from the saturated surface. The uppermost spectrum in the right panel of Fig. 2 was recorded on the RT-saturated Si(100)2×1-Rb surface after cooling the sample to a low temperature (LT, $\sim$120 K). The spectrum became a little bit sharper after cooling and there is an overall downward shift of the spectrum by $\sim$0.45 eV due to the surface photovoltage effect (SPV) but the overall shape of the LT spectrum is essentially the same as that of the RT spectrum.

Figure 4 shows spectra recorded at increasing emission angles in the [010] azimuthal direction from a cold Si(100)$2\times$-Rb surface. Rb atoms, corresponding to an amount of 1 θ_s, were deposited onto a cold ($\sim$120 K) Si(100)$c(4\times2)$ surface. An overall energy shift of ~-0.45

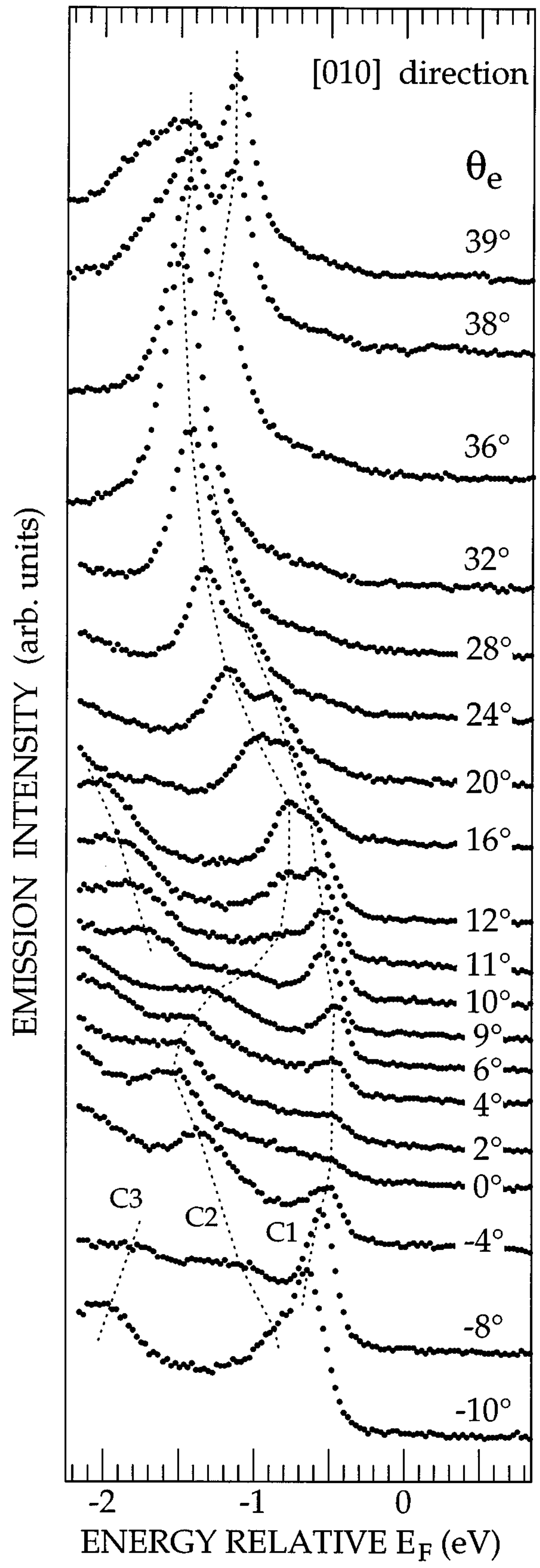

FIG. 4. Angle-resolved valence-band spectra recorded from the Si(100)2×1-Rb surface with a photon energy of 21.2 eV at a sample temperature of $\sim$120 K. The various emission angles, θ_e, along the [010] azimuthal direction label the different spectra.

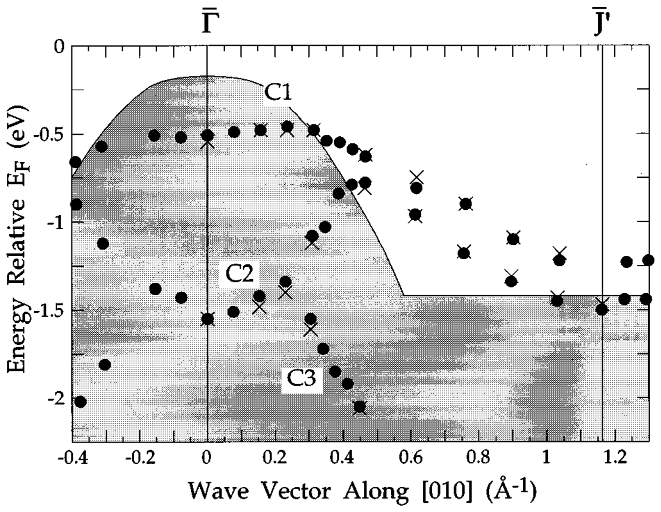

FIG. 5. Experimental two-dimensional band structure for the Si(100)2×1-Rb surface along the [010] direction. The solid dots were obtained from a surface prepared by Rb deposition directly onto a cold Si(100)$c(4\times2)$ surface. The crosses were obtained from a surface that was cooled after a RT preparation. Both surfaces were exposed to a Rb amount equal to 1 θ_s. The shaded region is the projected bulk band structure from Ref. 39. A value of 0.17 eV was used for $E_F - E_V$ which was determined from Si $2p$ core-level spectra (not included in this paper).

eV due to the SPV effect was subtracted from all spectra. In this series the energy width of the spectra below the Fermi level was restricted to 2.2 eV in order to focus on the dispersion of the two Rb-induced surface states $C1$ and $C2$. The resulting dispersion curves are plotted in Fig. 5. The shaded region in Fig. 5 is the projected bulk band structure from Ref. 39. In the normal-emission spectrum $C1$ and $C2$ are separated by ~1 eV and the emission intensity of these two surface states is rather weak. For small emission angles $C2$ disperses upward very quickly while the energy position of $C1$ is almost unchanged. As the emission angle increases, both states start to disperse downward and they show much higher intensities when they move into the projected bulk band gap. Close to the J' point the two states cannot be resolved in the spectra and they form a sharp pronounced peak at an energy position of -1.5 eV. At higher emission angles $C1$ and $C2$ split again into two well-resolved peaks. Another state, $C3$, is also present in Fig. 4 and the dispersion is shown in Fig. 5. This structure was also observed on the Si(100)2×1-K (RT saturated)[4] and Si(100)2×1-H (monohydride phase)[40] surfaces. A recent band-structure calculation for the Na- and K-saturated Si(100)2×1 surfaces by Krüger and Pollmann[13] gives a band originating from the Si-dimer bond, which shows some similarity with $C3$ along [010] direction. However, further studies are needed in order to clarify the origin of this structure.

It is instructive to compare the RT-saturated Si(100)2×1-Rb surface with other AM-saturated Si(100)2×1 surfaces. In Fig. 6 we present valence-band spectra recorded on RT-saturated Si(100)2×1-Na, -K, -Rb, and -Cs surfaces at the Γ point. The two surface states $C1$ and $C2$ are well resolved in the spectra from the Na-, K-, and Rb-saturated surfaces, while $C2$ cannot be clearly identified for the Cs case. Results from theoretical studies of the Si(100)2×1-Na and Si(100)2×1-K surfaces are consistent with the appearance of the surface states $C1$ and $C2$. In the theoretical studies two completely filled surface bands are found which correspond to hybrid states formed by the Si dangling bonds and the AM valence orbitals.[12,13] The $C1$ state shifts towards the Fermi edge with increasing size of the AM atoms. Also the $C2$ state shifts upwards, but only slightly, when going from Na to Rb. The controversy about the metallicity at the AM-Si interfaces could, in principle, be solved by a careful study of the emission close to the Fermi level. For Na, there is no emission close to the Fermi level which indicates that the surface is semiconducting. For K, a clear structure is observed near the Fermi level. The appearance of this structure was accompanied by an overall energy shift of the spectrum by ~0.2 eV toward the higher-binding-energy side. A plausible explanation for the energy shift is that the minimum of an empty surface band on the K-saturated surface starts to become occupied. This surface band located above the surface band gap was previously discussed in an inverse and direct photoemission study.[20] A systematic study of the adsorption of different AM atoms (Li, Na, and K) on the Si(100)2×1 surface has been performed with inverse and direct photoemission by Johansson and Reihl.[20,21,41] An AM-induced unoccupied state was observed in their inverse photoemission spectra (IPES). This state was consistent with the results of the theoretical studies, in which a strong unoccupied state was found with the charge density localized at the AM atom in the pedestal position.[13] The IPES results also show that for increasing size of the AM atoms, the energy position of the empty band

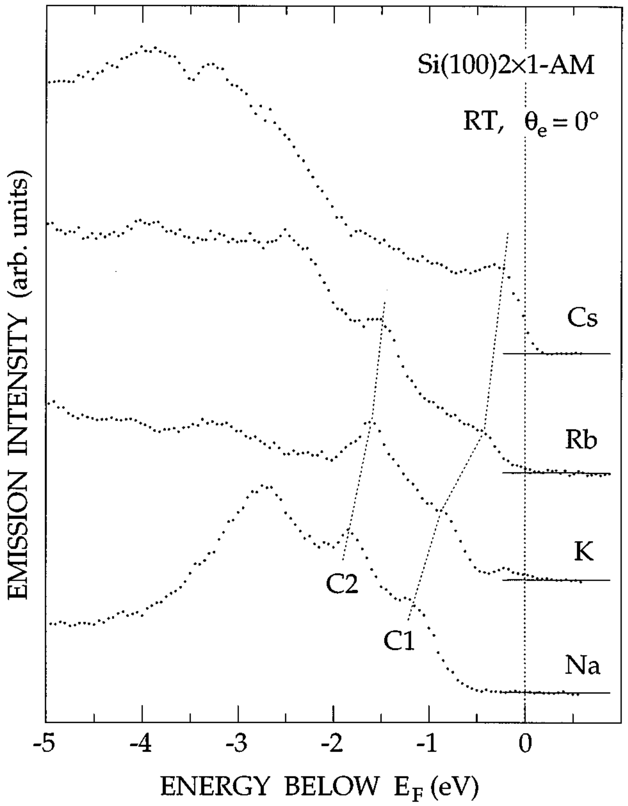

FIG. 6. Comparison between normal-emission valence-band spectra obtained from different RT-saturated Si(100)2×1-AM surfaces recorded with a photon energy of 21.2 eV at RT.

at $\bar{\Gamma}$ moves downwards and the surface band gap decreases. The dispersion of the empty band increased from no dispersion for Li to 1.4 eV for K, which indicates a stronger AM-AM interaction and a weaker AM-Si interaction when the size of the AM atoms increases. From the IPES studies we expect to find an experimental surface band gap for Rb and Cs of only a few tenths of an eV. The occupied surface-state peak at the $\bar{\Gamma}$ point, $C1$, will thus be located very close in energy to the minimum of the empty band. A metallization due to occupation of this minimum as proposed for the Si(100)2×1-K surface will thus lead to emission very close to the occupied surface-state peak for the 2×1-Rb and 2×1-Cs. In fact, the observation of the upward shift of the $C1$ state and the enhanced emission intensity observed at the Fermi level for Rb and Cs (see Fig. 6) agree indeed with the above hypothesis. The spectrum of the Si(100)2×1-Rb surface is similar to that of the K-adsorbed surface except that structure $C1$ is closer to the Fermi level. A slightly asymmetric line shape has been observed in Si and Rb core-level spectra at the room-temperature saturation coverage. The singularity index of the Doniach-Šunjić line shape required in the curve fittings for the Si $2p$ and Rb $3d$ spectra are 0.05 ±0.01 and 0.07±0.01, respectively. These values are similar to the results from the 2×1-K surfaces but smaller than that of the 2×1-Cs surfaces. For the Si(100)2×1-Cs surface, the rather strong emission intensity near the Fermi level and the strong asymmetric tails in both the Si $2p$ and Cs $4d$ spectra indicate a strong metallic character even though the surface is not metallic in the true sense since the surface most likely has a nonzero surface band gap.[29]

In Fig. 7 we compare the dispersions of the $C1$ and $C2$ bands for the Si(100)2×1-Rb surface (solid lines) to the corresponding dispersions for the Si(100)2×1-K (dashed lines) and Si(100)2×1-Cs (dotted lines) surfaces obtained from Ref. 4. For the Cs-adsorbed surface the $C2$ band is only identified for $k_{\parallel}$ values larger than ~ 0.7 Å^{-1}. For the K-adsorbed surface, on the other hand, these two bands can only be distinguished for small $k_{\parallel}$ values. Except for these details the dispersions are qualitatively quite similar for all three surfaces. It is also noticeable that the $C1$ band shifts upward from K to Cs, which is consistent with the observation in Fig. 6. The similarity of the dispersion curves indicates that the Si(100)2×1-Rb surface must have an atomic structure similar to that of the 2×1-K and 2×1-Cs surfaces. In our previous core-level studies,[24,29] two components were found in the K $3p$ and Cs $4d$ spectra which were interpreted as evidence for two different adsorption sites. Together with the observations of 2×1 LEED patterns, the RT-saturated Si(100)2×1-AM surfaces are likely to have a coverage close to one full monolayer with two different adsorption sites. This is consistent with the proposed double-layer model which leads to a semiconducting surface but due to the decrease in the surface band gap the Si(100)2×1-Cs surface appears more metallic in photoemission than the Si(100) surface saturated with the other alkali metals.

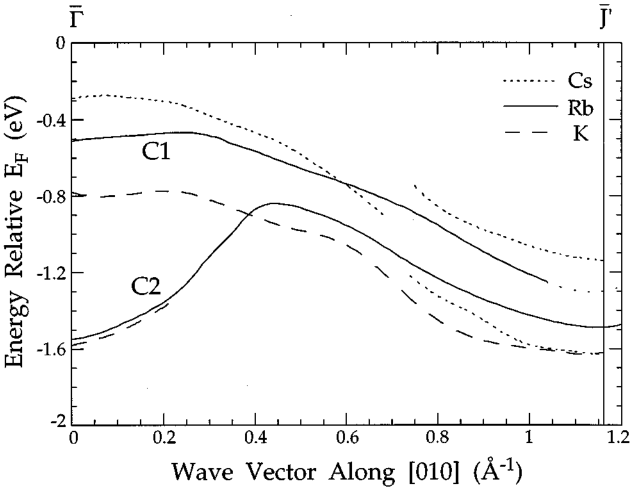

FIG. 7. Comparison between the experimental surface-state dispersions obtained in the present study for the Si(100)2×1-Rb surface (solid lines) and the 2×1-Cs (dotted lines) and the 2×1-K (dashed lines) surfaces presented in Ref. 4.

IV. CONCLUSIONS

In summary, Rb adsorption on the Si(100)2×1 surface has been studied with angle-resolved photoemission. A series of valence-band spectra for different Rb coverages is presented. In the initial stage of Rb coverage a strong surface-state F appears at the Fermi level due to a partial occupation of a normally empty surface band. The onset of occupation leads to an abrupt change in the band bending by shifting the Fermi-level pinning position upwards. For a RT saturated Si(100)2×1-Rb surface two Rb-induced surface states $C1$ and $C2$ are resolved with energies of -0.55 and -1.55 eV at $\bar{\Gamma}$, respectively. The dispersions of $C1$ and $C2$ were determined along the [010] azimuthal direction. A systematic comparison to other RT saturated Si(100)2×1-AM (Na, K, and Cs) surfaces was done. Our results indicate that the Si(100)2×1-Rb surface has an atomic structure that is similar to the other 2×1-AM surfaces, i.e., the double-layer model is favored by our results.

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