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Atomic and electronic structure of the $(2\sqrt{3} \times 2\sqrt{3})R30^\circ$ strained Sn reconstruction on Ge/Si(111)

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Abstract

Motivated by the different behaviour of Sn/Si(111) and Sn/Ge(111) in their metal-insulator transition, we have explored the possibility of growing Sn on an ultra-thin Ge layer strained on top of a Si(111) substrate. We have demonstrated by scanning tunneling microscopy and low energy electron diffraction that a $(2\sqrt{3} \times 2\sqrt{3})R30^\circ$ reconstruction can be stabilized under adequate growth conditions. The size of the reconstructed domains increases progressively up to a coverage of 1.3 monolayer of Sn, as determined by a combined study of scanning tunneling microscopy and core level spectroscopy. This coverage differs from that of Sn/Si(111) and Sn/Ge(111) exhibiting Mott phases. Angle resolved photoemission shows that the highly strained reconstruction is a band insulator, with a surface state dispersing roughly between 1300 and 2300 meV of binding energy.

PACS numbers:

Semiconductor surfaces present narrow surface bandwidths, and are thus excellent candidates to search for correlation induced metal-insulator transitions^{1,2}. Interestingly, electronic transitions as a function of the temperature have been observed: a Mott transition on the Sn/Ge(111) interface³ and a similar transition on Sn/Si(111)^{4,5}. These correlated surfaces appear for a Sn coverage of 1/3 ML, where all the T4 sites are occupied with Sn atoms of three different heights⁶. Despite this similarity, there are some differences in the two systems. Sn/Ge shows a structural transition from a (3×3) towards a $(\sqrt{3} \times \sqrt{3})R30^\circ$ at 30 K, concomitantly with the Mott transition^{3,7}. In turn, Sn/Si exhibits conductivity changes below 100 K⁴, but no structural transition has ever been observed down to 6 K⁸: a $(\sqrt{3} \times \sqrt{3})R30^\circ$ symmetry is always observed, which indicates a flat surface and equivalent Sn atoms. Since the Sn 4d core level exhibits more than one component even at low temperatures and the band structure shows a (3×3) symmetry⁹, quantum fluctuations between two configurations have been evoked¹⁰. The interest on Sn/Si has recently been renewed due to the successful combination of the dynamically screened interaction (or the GW theory) and Dynamical Mean Field Theory calculations¹¹ as well as the eventuality of magnetic order¹².

While extensive work has been performed on both Sn/Si(111) and Sn/Ge(111) systems, many issues remain still unsolved. In particular, the different behaviors as a function of temperature as well as the driving force of the metal-insulator transition are not yet understood. The transition may be triggered either by the lattice parameter change due to the elastic deformation or by the charge screening. In order to distinguish between these two effects, we propose the original approach of exploring the growth of artificial systems with the electronic properties of one overlayer (Sn/Ge) and the elastic properties of another (Sn/Si). Such a study may help to disentangle between the elastic or the electronic contributions in phase transitions. Before reaching this stage, it is necessary to demonstrate the growth of a strained interface (Sn/Ge/Si) and to study its similarity with the well-known reconstructions observed on either Sn/Ge(111) and Sn/Si(111) surfaces. This is precisely the aim of this article, where we present a growth study of Sn on ultrathin strained Ge films on a Si(111) substrate in the seek for a stable reconstruction. After determining a stable reconstruction and studying its electronic properties, we have analyzed the similarity of the reconstruction to α phases of Sn/Si and Sn/Ge. To the best of our knowledge this is the first attempt to study the evaporation of Sn on strained Ge/Si(111)- (5×5) as usually Sn is directly adsorbed on the Ge(111)-c (2×8) or on the Si(111)- (7×7) reconstructions.

FIG. 1: (Color online) STM images obtained after deposition of 2-3 ML layers of Ge on Si(111) at 500°C: (a) 100 nm \times 100 nm ($V = -2$ V et $I = 1$ nA) and (b) 15 nm \times 15 nm with the (5×5) unit cell. STM images obtained after deposition of 1.3 ML of Sn on Ge/Si(111): (c) 400°C, 100 nm \times 100 nm (-2 V and 1 nA), (d) 400°C, 15 nm \times 15 nm (-1.9 V and 0.4 nA) (e) 300°C, 100 nm \times 100 nm (-1.9 V and 0.4 nA) and (f) 300°C, 15 nm \times 15 nm (-1.9 V and 0.7 nA). The unit cell of the $2\sqrt{3}$ reconstruction is indicated.

The experiments were carried out in two different ultra-high vacuum chambers with a base pressure during the experiments in the low 10^{-10} mbar range. The Si(111) substrates were n-doped with a doping level of 3×10^{16} cm $^{-3}$. Scanning tunnelling microscopy (STM) measurements were performed with a modified low temperature Omicron microscope operating at RT. A negative bias voltage probes the filled states of the sample. Images were analysed using WsxM software¹³. The photoemission measurements were performed at the CASSIOPEE beamline of SOLEIL synchrotron. Incoming radiation and the Scienta SES-4000 detector subtend a 45° angle. Light polarization was in the plane defined by these two directions and the detector slit was perpendicular to them. Band structure measurements were performed at $h\nu = 29$ eV and the core level of Sn 4d was acquired at $h\nu = 80$ eV in normal and grazing emission angle. Our preparation of Si(111)- (7×7) and Ge/Si(111)- (5×5) has been described elsewhere^{14,15}.

FIG. 2: (Color online) LEED pattern of (a) Ge/Si(111)- (5×5) reconstruction (corresponding to fig. 1a), (b) the non-optimal reconstruction of fig. 1c and (c) the optimized Sn/Ge/Si(111)- $(2\sqrt{3} \times 2\sqrt{3})R30^\circ$ reconstruction at a beam energy of 60 eV.

Figure 1(a) shows an STM image (100 nm \times 100 nm) obtained upon deposition of 2-3 ML Ge on top of a Si(111)- (7×7) surface at 500°C. The surface consists of terraces having a characteristic width of about 30 nm separated by steps. It is well known that Ge has a larger lattice constant than Si, the misfit being 4.2%. The growth of Ge on Si thus follows the Stranski-Krastanow mode: a 2D wetting layer is observed before the formation of 3D islands. In our case, 3D islands do not appear, indicating that the Ge

film is fully strained and that elastic strain relaxation has not started yet. A (5×5) reconstruction¹⁶ is obtained as shown by a high resolution STM image (Fig. 1(b)) and by low energy electron diffraction (LEED) (Fig. 2(a)). The (5×5) symmetry is observed at different surface locations, demonstrating that uniform, ultrathin strained Ge films can be obtained on Si(111) surfaces. This is an important prerequisite for further Sn deposition. In the following, we studied the growth of Sn as a function of coverage between 0.15 ML and 2 ML and deposition temperatures between 25°C and 500°C. The aim is to determine whether a commensurable surface structure can be formed when Sn is deposited on top of Ge/Si(111)- (5×5) surfaces. When the surface is maintained at 400°C during Sn deposition, the surface does not exhibit a particular reconstruction as shown in Figs. 1 (c)-(d) and by LEED in fig. 2b. On the other hand, when Sn is grown at 300°C, a $(2\sqrt{3} \times 2\sqrt{3})R30^\circ$ reconstruction ($2\sqrt{3}$ in short) appears as shown by STM in Fig. 1(e)-(f) and by LEED in Fig. 2(c). A large scale STM image (Fig. 1(e)) shows that the $2\sqrt{3}$ reconstruction is not homogeneous but rather consists of domains, of increasing size with the Sn coverage. The surface appears to be completely covered by $2\sqrt{3}$ reconstructed domains for a Sn coverage of about 1.3 ML. Finally, when Sn is deposited at substrate temperatures lower than 300°C, the STM images (not shown here) reveal a disordered surface. We thus conclude that 300°C is the optimal temperature to organize Sn adatoms on top of Ge/Si(111)- (5×5) surfaces. We then perform a coverage dependent study at this particular temperature. For Sn coverages in the 0.1-0.2 ML range, traces of the (5×5) reconstruction are observed by LEED. The $2\sqrt{3}$ diffraction spots increase their intensity up to 1.3 ML and beyond this coverage a (1×1) symmetry is observed, corresponding to the bulk diffraction pattern as the surface is disordered. STM images of the optimal reconstruction consist of circular bright spots arranged in a $2\sqrt{3}$ periodicity, strictly different from the elongated bright features observed on Sn/Si(111)- $2\sqrt{3}$ which appears for 14 Sn atoms per unit cell¹⁷. The observed reconstruction must thus correspond to a significantly different structure. The domain size of the optimal reconstruction is only around 100 nm² as we expect that the relaxation of the surface strain prevents the formation of larger coherent domains (fig. 1e). The limited domain size can also be observed on the high background of the LEED pattern (fig. 2).

After growing the strained $(2\sqrt{3} \times 2\sqrt{3})R30^\circ$ Sn reconstruction, we were interested in determining the degree of similarity with Sn/Ge(111) and Sn/Si(111) reconstructions. A first difference is the coverage needed to fully cover the surface. In unstrained surfaces, the

FIG. 3: (Color online) Sn 4d core level of Sn/Ge/Si(111)-($2\sqrt{3} \times 2\sqrt{3}$)R30° prepared at 300°C ($h\nu = 80$ eV) at (a) normal emission and (b) at 45° off normal emission. Spectra are fitted with a spin-orbit splitting of 1.05 eV, a branching ratio of 0.74, a Lorentzian width of 0.07 eV and a gaussian width of 0.4 eV. Two components separated by 310 meV with an intensity ratio 1:10 are observed.

phase exhibiting metal-insulator transitions is the α phase which corresponds to 0.33 ML while on Ge/Si(111) 1.3 ML Sn are required to uniformly cover the surface. Besides reporting on the coverage, Sn 4d core level determines the number of inequivalent Sn atoms on the surface, which is a crucial information to settle a model for the reconstruction. Fig. 3 shows the Sn 4d core level at $h\nu = 80$ eV of Sn/Ge/Si(111)-($2\sqrt{3} \times 2\sqrt{3}$)R30°. The lineshape is fitted with a major component (red dashed line in Fig. 3) and a minor component (blue dashed line in Fig. 3) at (310 ± 10) meV higher binding energy. The intensity ratio of the minor component is about 10% of the major one, both at normal and at 45° off normal emission. The intensity independence of the emission angle indicates that both components are originated at a similar depth from the surface. The interdiffusion of Sn on the subsurface region must not be significant and the higher binding energy component can be related to surface defects, probably at step edges, where STM images show some bright regions. We cannot however exclude the presence of other subsurface Sn components not visible at a more bulk sensitive photon energy.

FIG. 4: (Color online) (a) Band structure of the Sn/Ge/Si(111)-($2\sqrt{3} \times 2\sqrt{3}$)R30° reconstruction measured at $h\nu = 29$ eV (darker colors correspond to higher intensity). The bulk bands are identified with the label B and a surface state is indicated by SS. Lines placed at a lower binding energy than the bands are a guideline to the eye to follow the band dispersion.

Figure 4 shows the band structure of the Sn/Ge/Si(111)- $2\sqrt{3}$ surface measured along the surface high symmetry directions ($h\nu = 29$ eV). The Sn adsorption and the subsequent $2\sqrt{3}$ reconstruction suppresses the metallic surface state of the Ge/Si(111)-(5×5)surface¹⁵, but it may induce different surfaces states. Several bands can be identified in Fig. 4 such as

FIG. 5: (Color online) Energy distribution curves as a function of the photon energy at $k_x=0.26 \text{ \AA}^{-1}$ along $\overline{\Gamma K}$ direction. The continuous (green) line indicates a non-dispersing state as a function of k_z . The dashed (blue) lines are a guide for the eye to identify dispersing and non-dispersing states as a function of k_z .

those marked by B, which correspond to bulk bands¹⁸ and another feature marked by SS (identified by a yellow dashed line) dispersing between 1300 and 2300 meV, which could possibly correspond to a surface state. In order to confirm the assignment of these features, we vary the photon energy, i.e. k_z . Figure 5 shows the energy dispersion curves taken in a reciprocal point with the same projection along the $\overline{\Gamma K}$ at different photon energies ranging from 21 eV to 32 eV. We can identify two states which disperse along k_z (as their binding energy varies with respect the blue dashed lines) and they thus correspond to bulk states. Another state does not disperse perpendicularly to the surface, i.e. its binding energy is always constant as it follows the green solid line. We can thus conclude that the SS feature must correspond to a surface state induced by the adsorption of Sn. Since 16 Sn adatoms are present in the unit cell, SS is probably not a single surface state but rather consists of a manifold of non-metallic surface states. Finally, we observe that the SS feature does not intersect the Fermi level at any point of $(2\sqrt{3} \times 2\sqrt{3})R30^\circ$ Brillouin zone, so the reconstruction is insulating. The non-metallicity of the surface is thus not associated with a Mott insulator as in Sn/Ge(111) or in Sn/Si(111), where the unit cell contains only one atom (i.e. one electron). In Sn/Ge/Si(111), the number of atoms in the unit cell is even, as the number of available electrons, so all the observed bands are completely filled. In consequence, Sn/Ge/Si(111) does not allow to conclude about the driving force of the Mott transition in Sn/Ge(111) and in Sn/Si(111) but it appears instead as a playground for exploring strain effects in the electronic properties.

In conclusion, we have successfully stabilized a $(2\sqrt{3} \times 2\sqrt{3})R30^\circ$ reconstruction during deposition of Sn at 300°C on strained Ge/Si(111)- (5×5) surface. At this particular temperature, the reconstruction consists of domains having a maximum size of 100 nm², probably limited by the underlying strain of the substrate. Core level spectroscopy shows one component associated with the reconstruction indicating a similar chemical environment for all the

Sn atoms probed by 80 eV incoming photons. As the number of atoms per unit cell is 16, they are probably distributed in more than one layer. The lower Sn atoms can be bonded to the dangling bonds of the (5×5) reconstruction and stabilized afterwards by clustering. The electronic structure is insulating, with one surface feature that must be associated with several unresolved surface states. The binding energy of these surface states is extremely high to be associated with Hubbard bands. The $(2\sqrt{3} \times 2\sqrt{3})R30^\circ$ reconstruction is very different from Sn/Si and Sn/Ge, with a different coverage and an even number of electrons per unit cell, being thus a band insulator. Further experimental and theoretical work would be needed to propose a structural model for the $(2\sqrt{3} \times 2\sqrt{3})R30^\circ$ reconstruction observed on Sn/Ge/Si(111) surfaces. The model should be compatible with a coverage of 16 atoms per unit cell, an insulating surface under strain and similar chemical shifts in the last layer of Sn atoms.

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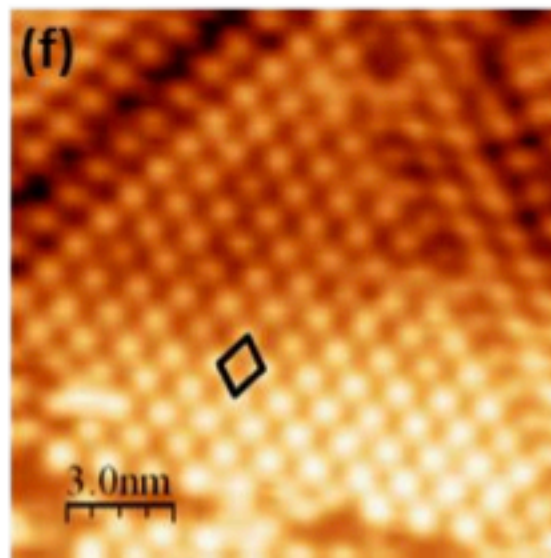
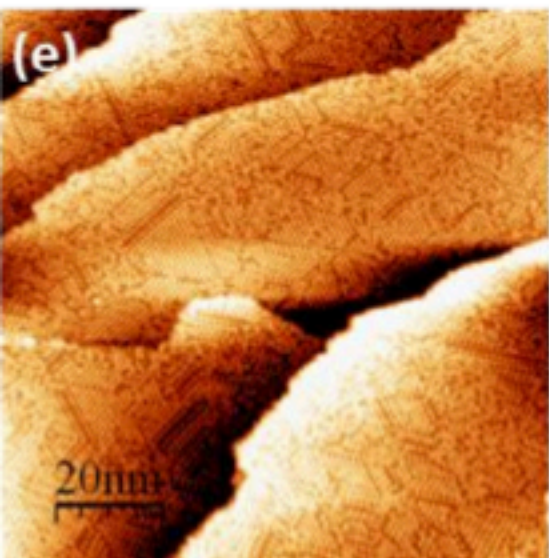
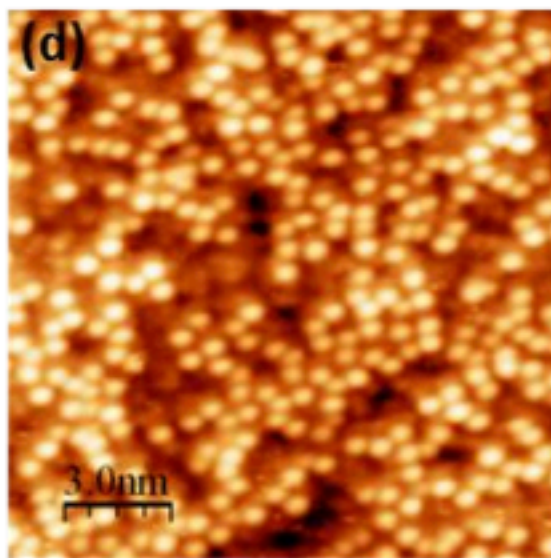
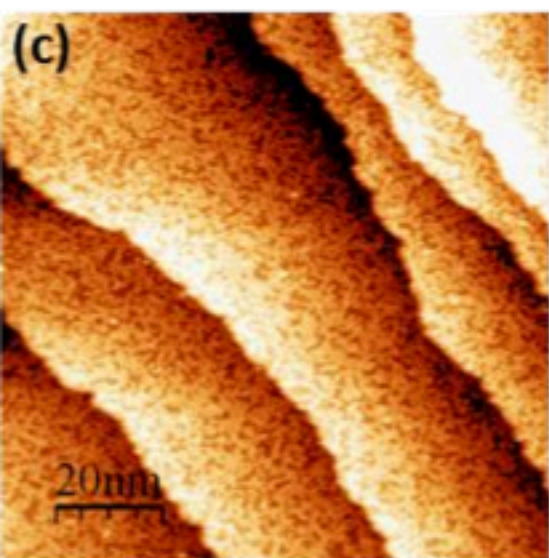
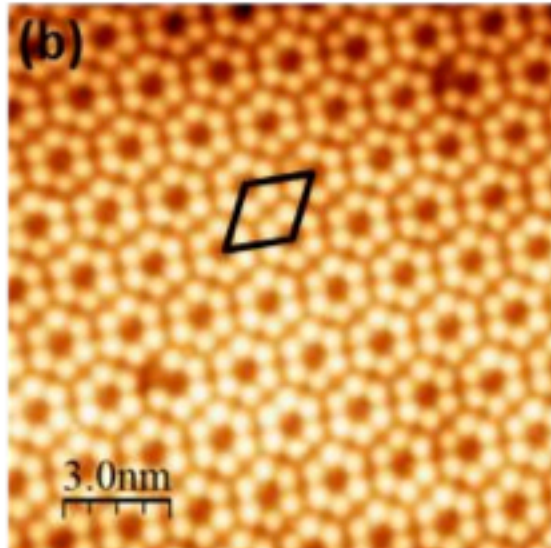
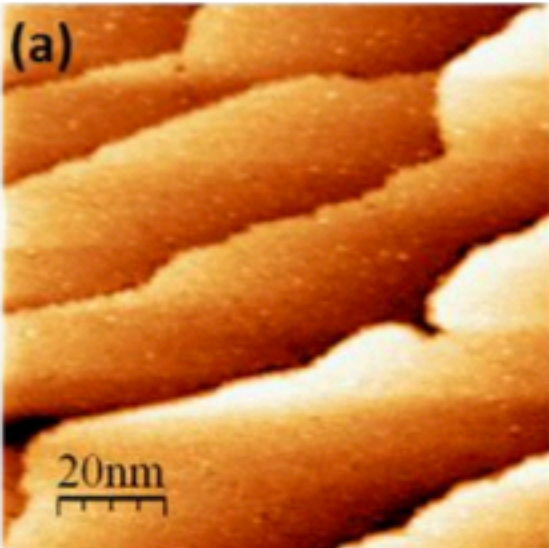
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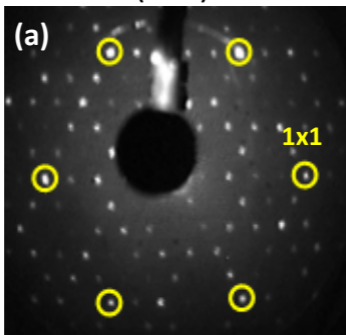
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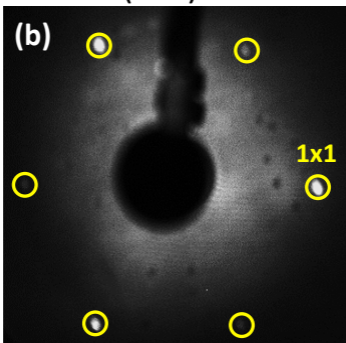
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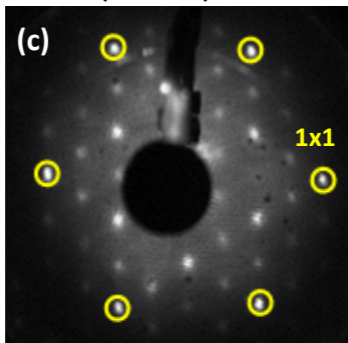
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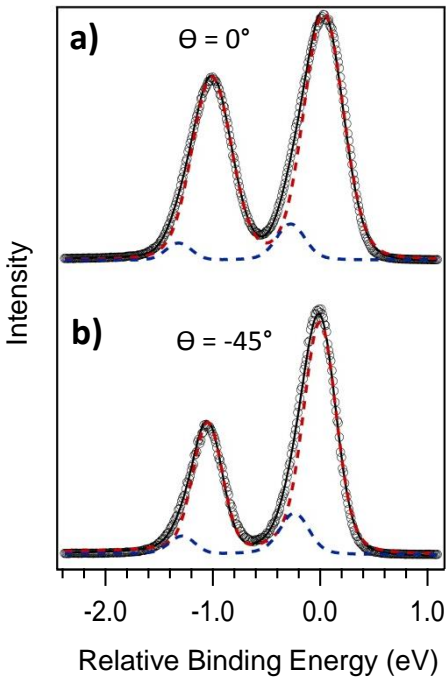


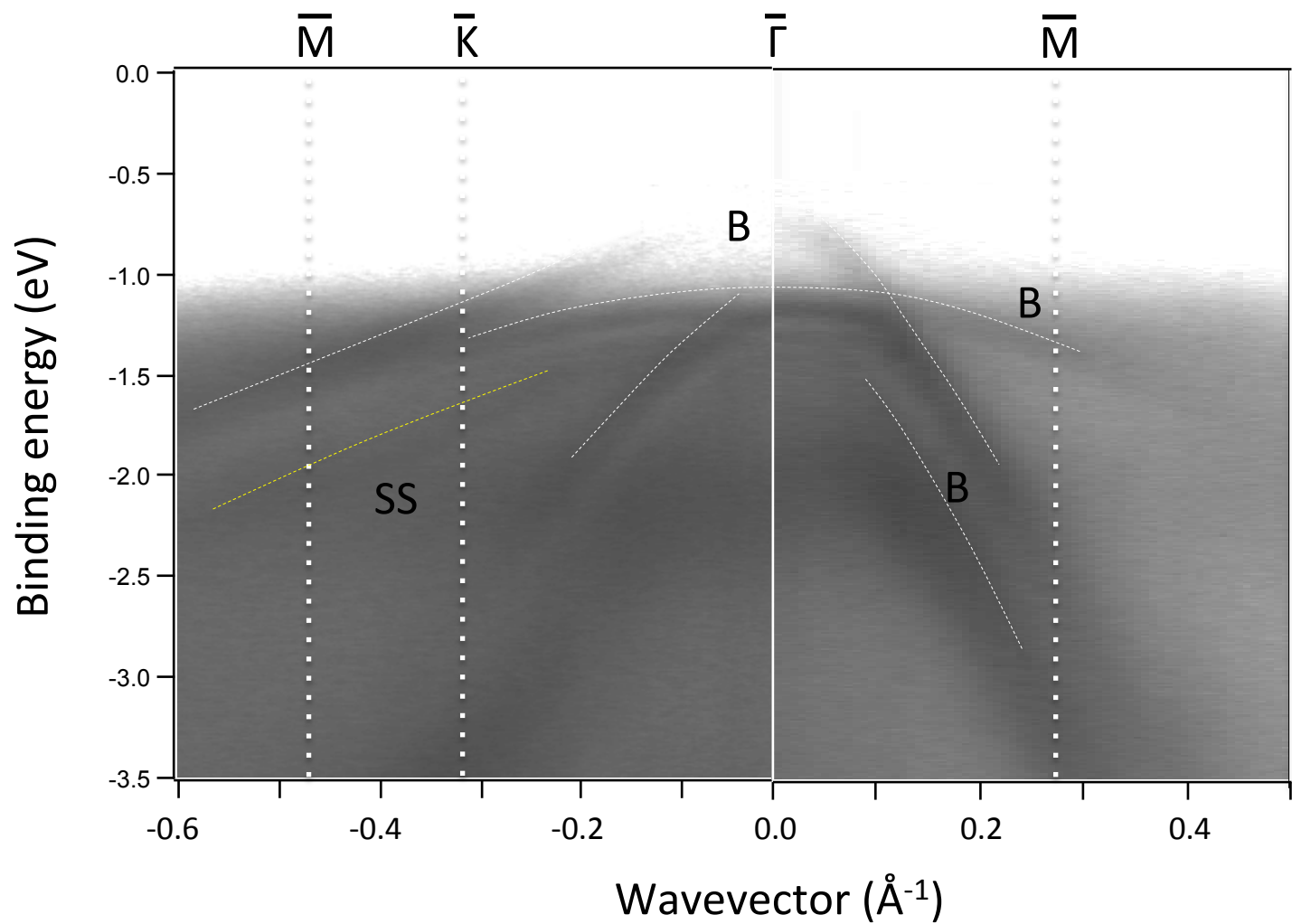
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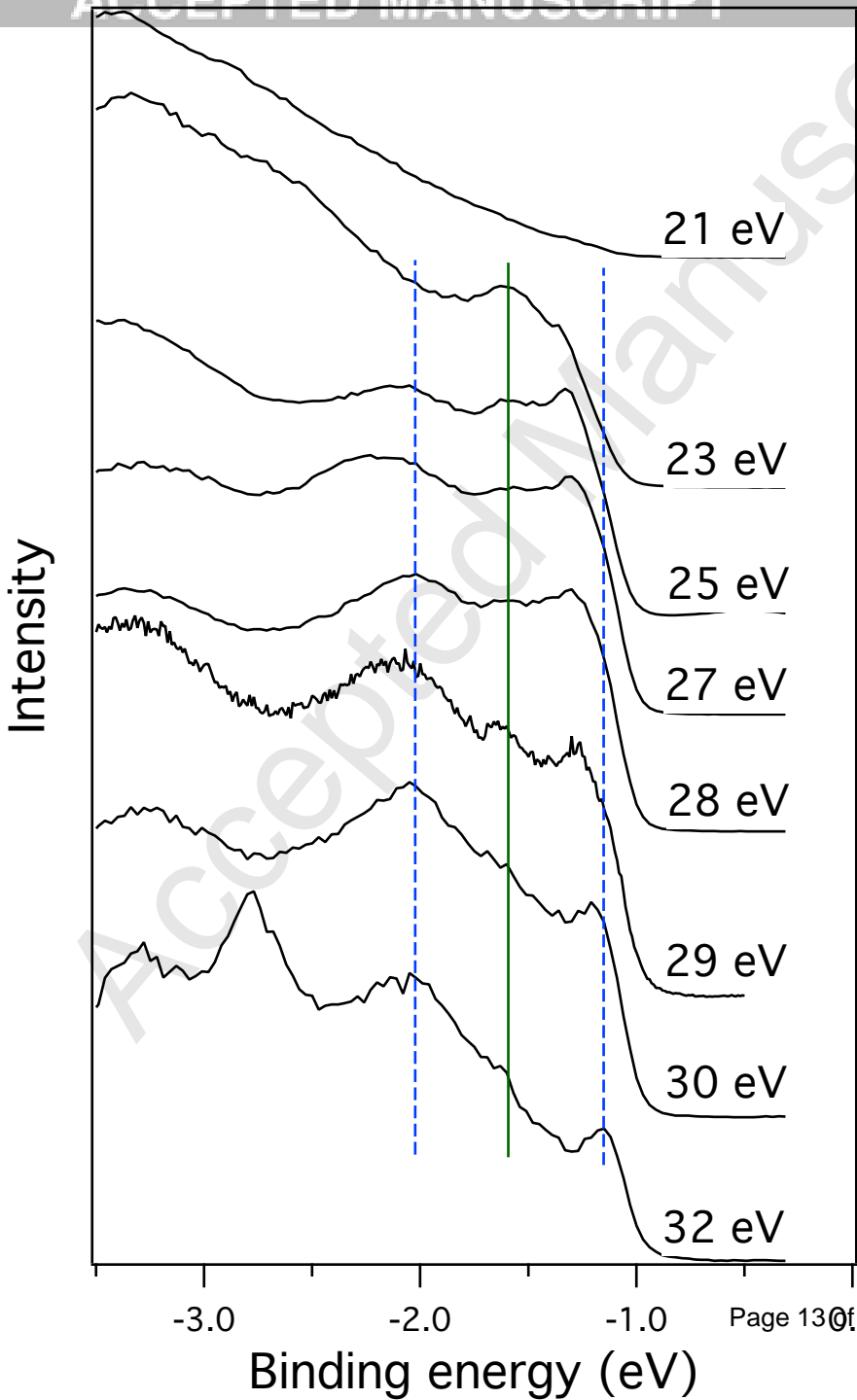


$2(\sqrt{3} \times \sqrt{3})R30^\circ$









Highlights

- * We find a new Sn reconstruction on strained Ge/Si(111)
- * The band structure reveals it is a band insulator.

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