Metallic Nature of the α-Sn/Ge(111) Surface down to 2.5 K

Stefano Colonna,* Fabio Ronci, and Antonio Cricenti+
ISM CNR, Via del Fosso del Cavaliere 100, I-00133 Roma, Italy

Guy Le Lay‡
CINaM-CNRS, Campus de Luminy, Case 913, F-13288 Marseille Cedex 09, France
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Low temperature (down to 2.5 K) scanning tunneling microscopy (STM) and spectroscopy (STS) measurements are presented to assess the nature of the α-Sn/Ge(111) surface. Bias-dependent STM and STS measurements have been used to demonstrate that such a surface preserves a metallic 3×3 reconstruction at very low temperature. A tip-surface interaction mechanism becomes active below about 20 K at the α-Sn/Ge(111) surface, resulting in an apparent unbuckled $(\sqrt{3} \times \sqrt{3})$ reconstruction when filled states STM images are acquired with tunneling currents higher than 0.2 nA.

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According to the scale localization theory in a two-dimensional electron system (2DES) the metallic conductivity should be suppressed at low temperature [1]. However, a number of experimental results have suggested that finite conductivity is possible in different 2DESs [2]. In fact, the metal to insulator transition (MIT) in 2DESs remains largely a mystery. To address this issue the electron-electron interactions in a strictly 2D system has been the focus of interest at some prototypical semiconductor surfaces, which are an ideal playground for the analysis of critical phenomena. This is especially the case at the one third of a monolayer of group IV adatoms on Ge(111) and Si(111) substrates (the so-called α phase). The α-Sn/Ge(111) surface and its Pb/Ge(111), Pb/Si(111) and Sn/Si(111) partners are especially interesting for their triangular surface crystal lattice supposed to be a good model system for potential electron correlation effects. Except for the α-Sn/Si(111) interface, which maintains the $(\sqrt{3} \times \sqrt{3})R30°$ periodicity (for brevity $\sqrt{3}$ hereafter) at temperature as low as 2.3 K [3], the α phase of the isoelectronic Sn/Ge(111), Pb/Ge(111), and Pb/Si(111) interfaces has been extensively studied for their $\sqrt{3} \leftrightarrow 3 \times 3$ reversible phase transition below a critical temperature [4–6]. The electronic structure of the 3×3 reconstructed surfaces presents a narrow metallic conduction band believed to be prone to split into two subbands giving rise to an insulating surface. This idea has driven extensive research for electronic transitions at low temperature (below the critical temperature of the $\sqrt{3} \leftrightarrow 3 \times 3$ transition). Theoretical calculations have predicted a correlation-driven magnetic Mott metal-to-insulator transition and even a superconductive phase upon doping [7]. However, calculations reported in Ref. [8] have shown that the electron correlation effects on the 3×3Sn/Ge(111) surface could be merely responsible for a width reduction of the conduction band at the surface preserving its metallic character. Several recent experimental observations have been interpreted following the idea of the existence of a low temperature phase. In the case of the α-Sn/Ge(111) [9] and α-Sn/Si(111) [10] the arising of a low temperature Mott insulating phase was reported on the basis of scanning tunneling microscopy (STM) and photoelectron spectroscopy measurements (PES), whereas for the Pb/Ge(111) interface a glasslike phase was claimed below 76 K [11]. However, this observed glasslike phase has been disputed by Brihuega *et al.* [12] who explained this phenomenon as induced by a hindered surface conductivity at low temperature. The α-Sn/Ge(111) has been the object of a large number of studies aimed at determining the exact crystallographic arrangement, the electronic structure, and the mechanism of the $\sqrt{3} \leftrightarrow 3 \times 3$ transition [13–19]. A renewed interest for the α-Sn/Ge(111) surface has been stimulated by the discovery of a new $\sqrt{3}$ phase below 25 K (hereafter referred as LT-$\sqrt{3}$) attributed to a 2D MIT [9]. Even though the discovery of a 2D correlation-driven MIT is an exciting possibility, special care is necessary in the study of the α-Sn/Ge(111) and the related interfaces. In fact, the ground state of these interfaces is the result of a fine balance of the surface electronic and elastic energy. As a consequence, it is always necessary to carefully take into consideration possible probe-surface interaction effects in studies conducted by STM. In this Letter we report on low temperature (down to 2.5 K) STM STS measurements aimed to assess the nature of the Sn/Ge(111) surface.

The α-Sn/Ge(111) surface was prepared following a standard procedure in a UHV system [19] housing a low temperature STM (Omicron LT-STM). The reported STM images were all obtained in the constant tunneling current mode. The tunneling spectra were acquired during the image scan holding the tip scan movement on a regularly spaced grid of 80×80 points spread over the scanning area. The reported spectra are the result of the average of the spectra collected on 10×10 nm^2 images cutting out surface defects. The $I(V)$ curves [20] were numerically derived and the normalized spectra were obtained by dividing the differential conductance (dI/dV) by the total conductance (I/V), giving a direct measure of the surface density of electronic states [21].

In Fig. 1 STM images of the Sn/Ge(111) surface acquired at 80 and 10 K, (tunneling parameters: ± 1 V; 1.0 nA) are presented. The empty states (ES) and filled states (FS) images collected at 80 K show the well-known complementary decoration of the surface, exhibiting a honeycomb and a hexagonal pattern, respectively, typical of the 3×3 reconstruction. Decreasing the temperature to 10 K the FS image shows an apparent flat surface, i.e., all of the adatoms appear practically equivalent, in agreement with previously reported studies [9]. The observation of this LT-$\sqrt{3}$ reconstruction below 25 K was presented as the experimental evidence of a low temperature phase transition to a Mott insulating phase. Surprisingly, looking at the 10 K ES STM image in Fig. 1, a sharp 3×3 periodicity is still visible. This inconsistency between the ES and FS images calls for a more accurate investigation in order to understand the real nature of the α-Sn/Ge(111) surface at low temperature. In particular, a close examination of the possible interaction effects between the STM tip and the investigated surface becomes necessary. In Fig. 2 a sequence of constant current images collected at different gap voltages (sample temperature 5 K, tunneling current 0.2 nA) is reported. These images exhibit a continuous motif change as a function of the measurement parameters. At very low gap voltage the ES images show a faint patterning reminiscent of the honeycomb decoration of the 3×3 reconstruction. By increasing the gap voltage the 3×3 honeycomb pattern clearly emerges in the ES images up to $+1.2$ V. Further increasing the gap voltage the ES images exhibit a continuous transition to a hexagonal pattern passing through a flat appearance around $+1.6$ V. This behavior as a function of the sample bias was already experimentally observed at 78 K for the α-Sn/Ge(111) interface and simulated through *GW* calculations of the surface electronic structure [22]. FS images, on the other hand, show at very low negative bias a decoration similar to the ES images and an apparently undistorted surface at 0.2 V as predicted by the *GW* calculations [22]. Increasing the negative gap voltage, at variance with the measurements at 78 K and the *GW* calculations, the images appear disordered with a low corrugation and almost unbuckled around -1.0 V sample bias. At higher negative voltage the surface appears disordered with small patches of 3×3 reconstruction. The excellent agreement between the ES series here reported, the analogous series measured at 78 K, and the STM images simulated from *GW* calculations [22], which do not incorporate strong electron correlation effects, demonstrate that a one-electron picture for the α-Sn/Ge(111) surface is correct. Hence, the flat image around -1.0 V gap voltage has to be considered as an imaging effect. To understand the influence of the tip during the image acquisition we report in Fig. 3 the FS STM images collected at different tunneling currents (gap voltage -1.0 V, sample temperature 5 K) along with the related 2D-Fourier transforms. The STM image collected at 1.0 nA exhibits a clear $\sqrt{3}$ periodicity confirmed by the related Fourier transform too; decreasing the tunneling current to 0.2 nA, the 3×3 periodicity becomes again visible in both the topographic image and the Fourier transform; further decreasing the current down to 0.05 nA no changes are observed in the STM image, indicating that below 0.2 nA the expected topography is recovered. The most direct way to verify whether the proposed metal to insulator transition takes place at the α-Sn/Ge(111) surface or not is to perform a low temperature tunneling spectroscopy study. In fact, a gap opening is expected in the case of a MIT. In Fig. 4 we report tunneling spectra of the α-Sn/Ge(111) surface collected at temperatures ranging from 20 K to 2.5 K (current set point 0.1 nA) along with representative STM images of the surface at the same temperature. Clearly, the derivative

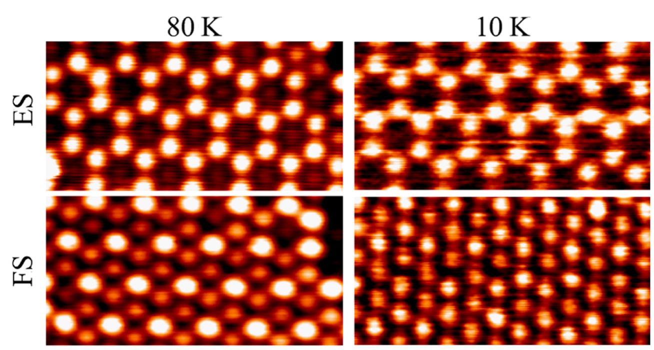

FIG. 1 (color online). Constant current images (7.0×3.5 nm^2) of the Sn/Ge(111) surface collected at ± 1.0 V, 1.0 nA for 80 and 10 K sample temperature.

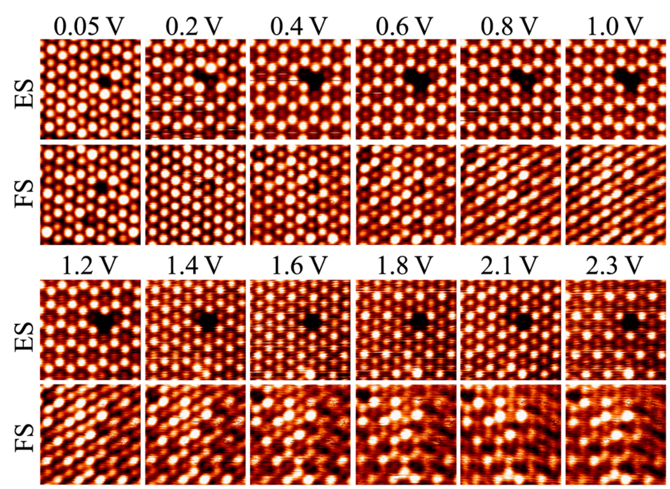

FIG. 2 (color online). Sequence of 5×5 nm^2 constant current (0.2 nA) images of the αSn/Ge(111) surface at 5 K at various gap voltages.

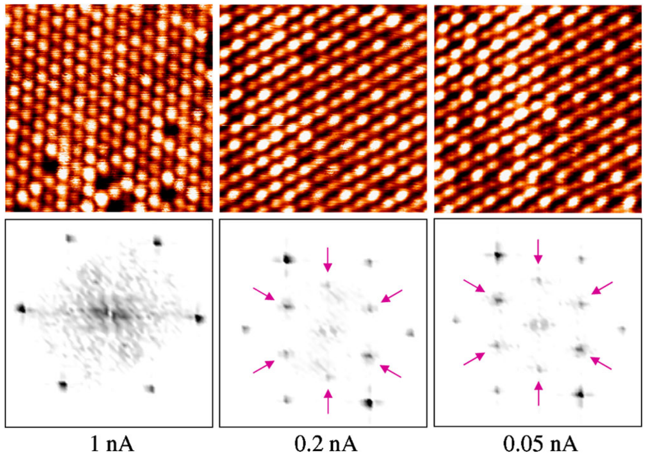

FIG. 3 (color online). Constant current 10×10 nm^2 FS images of the α-Sn/Ge(111) surface (-1.0 V gap voltage) at 5 K for various tunneling currents. In the lower panels are reported the Fourier transforms of the topographic images, the arrows indicating the 3×3 related spots.

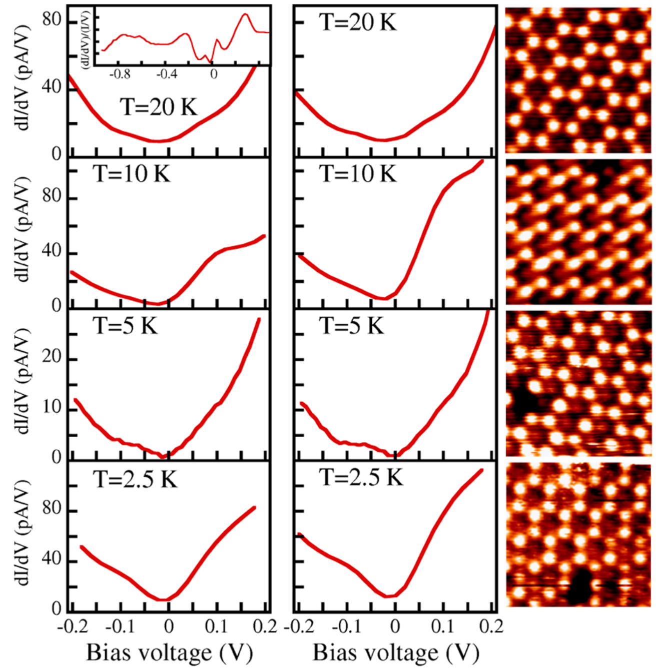

FIG. 4 (color online). Derivative of the tunneling spectra collected at 20 K to 2.5 K. Left panels spectra collected on ES images ($+1.0$ V); middle panels spectra collected on FS images (-1.0 V). The right panels show representative 5×5 nm^2 ES images collected at the different temperatures ($+1.0$ V). Tunneling current 0.1 nA (measurements at 5 K tunneling current 0.05 nA). In the inset, the $(dI/dV)/(I/V)$ spectrum of the α-Sn/Ge(111) ES 20 K measurement is reported.

of the I(V) spectra measured on ES and FS images are different from zero around the Fermi level, clearly demonstrating that the surface is metallic at temperatures down to 2.5 K. In the normalized STS spectrum of the α-Sn/Ge(111) (see the inset in Fig. 4) two structures located at about 0.25 and 0.68 eV below the Fermi level are observed. These structures reasonably compare to the ones found in the valence band photoemission spectra [23], the differences probably due to the different k integration for the two techniques [24]. The observation of a 3×3 periodicity and a metallic characteristic at the surface contradict the low temperature MIT proposed in Ref. [9]. In fact, the $\sqrt{3}$ periodicity is observed only in FS images and under specific measurement conditions, namely, high tunneling current, pointing to a tip-surface interaction effect rather than an intrinsic behavior of the sample surface. To explain the observed behavior at very low temperature of the α-Sn/Ge(111) surface it is necessary to envisage a physical mechanism which flattens or makes the surface apparently flat in FS STM images below a critical temperature. A similar behavior has been reported at the α-Pb/Ge(111) surface [12] and explained as the result of a local charging effect due to a reduced surface conductivity at low temperature. Similarly, it is possible to argue that the observed α-Sn/Ge(111) LT-$\sqrt{3}$ phase is not the result of an intrinsic surface transition but rather of an effect induced by STM measurements. It is interesting to note that in our measurements several FS STM images collected at low temperature exhibit many Sn atoms appearing flickering. As demonstrated in previous works a flickering STM image is the fingerprint of fluctuating adatoms [26,27]. It is possible to argue that below 20 K a tip-surface coupling mechanism exists which activates the Sn adatoms fluctuation when FS are probed. A similar behavior is well known in the case of the Si(100) surface, where the (2×1) surface reconstruction observed at high temperature, characterized by a symmetric appearance of the Si dimers, is the result of the thermally activated rocking mode of the dimers. The asymmetric dimers found in the $c(4 \times 2)$ reconstruction of the Si(100) surface are the result of the fluctuation freezing of the dimers below the critical temperature (about 200 K). More recent STM experiments have shown that further decreasing the sample temperature below 40 K, symmetric dimers are again observed as the results of Si atoms fluctuation. It was experimentally demonstrated by STM measurements that the flip-flop frequency, at low temperature, depends on the tunneling current and on the gap voltage polarity [28]. The detection of symmetric dimers at very low temperature was explained by several possible tip-surface interaction mechanisms: surface potential modification [29], electrostatic interaction [30], and inelastic tunneling phenomena [31,32]. In this last case the frequency of the dimers fluctuation was calculated to nonlinearly depend on the sample temperature and tunneling current [31,32]. In particular, at high temperatures the frequency is independent of the current, whereas reducing the sample temperature the frequency decreases and becomes dependent on it because the vibration deexcitation is governed by the surface electronic band population. It is possible to argue that a similar inelastic tunneling process could be active at the α-Sn/Ge(111) surface, giving rise to an apparent flat surface at specific tunneling conditions which could explain the appearance of the LT-$\sqrt{3}$ phase [33]. As a last remark it is interesting to note that the hypothesis of a low temperature transition at the Sn/Ge(111) surface was confirmed by valence band PES measurement [9]. The PES symmetrized spectra showed a gap opening below the critical temperature. This behavior was explained as the fingerprint of a Mott transition. On the contrary, our tunneling spectroscopy measurements exhibit a metallic surface at a temperature as low as 2.5 K. This seeming inconsistency can be explained by taking into account that PES measurements in Ref. [9] were performed along the Γ-K direction of the k space, whereas STS measurements presents also contributions from electronic states located in regions of the reciprocal space different from the Γ-K direction which is the only region explored by PES measurements. In fact, at the Γ point the energy band presents a minimum, so that electronic states with k parallel different from zero contribute to the tunneling current. Based on the reported experimental results the presence of a low temperature transition at the α-Sn/Ge(111) appears untenable. In addition, recent atomic force microscopy (AFM) measurements at the α-Sn/Ge(111) and α-Sn/Si(111) surfaces further support the 3×3 reconstruction as the ground state of the α-Sn/Ge(111) [34]. In fact, AFM images showed a 3×3 periodicity at temperatures as low as 6 K. In summary, by means of bias-dependent STM and STS measurements we demonstrate that the α-Sn/Ge(111) surface preserves the same ground state observed experimentally at 78 K and predicted by *GW* calculations, i.e., the metallic 3×3 reconstruction, casting serious doubts on the recent observation of a MIT below 25 K. STS measurements confirm that the surface metallicity is maintained down to 2.5 K. Moreover, we demonstrate that tip-surface interactions play an important role on the detection of the apparent unbuckled $\sqrt{3}$ reconstruction that is observed on FS STM images obtained below about 20 K with tunneling current higher than 0.2 nA. The metallic nature of this strictly 2DES system down to 2.5 K remains a challenge for the theoreticians community.

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*colonna@ism.cnr.it

†Also at Department of Physics and Astronomy, Vanderbilt University, Nashville, Tennessee 37235-1807, USA; also at NAST Centre, Via della Ricerca Scientifica 1, 00133 Roma, Italy.

‡Also at UFR Sciences de la Matière, Université de Provence, Marseille, France.

[1] E. Abrahams *et al.*, Phys. Rev. Lett. **42**, 673 (1979).

[2] E. Abrahams, S. V. Kravchenko, and M. P. Sarachik, Rev. Mod. Phys. **73**, 251 (2001).

[3] F. Ronci *et al.*, Phys. Rev. Lett. **99**, 166103 (2007).

[4] J. M. Carpinelli *et al.*, Phys. Rev. Lett. **79**, 2859 (1997).

[5] J. M. Carpinelli *et al.*, Nature (London) **381**, 398 (1996).

[6] O. Custance *et al.*, Surf. Sci. **482–485**, 1399 (2001).

[7] G. Profeta and E. Tosatti, Phys. Rev. Lett. **98**, 086401 (2007).

[8] F. Flores *et al.*, Prog. Surf. Sci. **67**, 299 (2001).

[9] R. Cortès *et al.*, Phys. Rev. Lett. **96**, 126103 (2006).

[10] S. Modesti *et al.*, Phys. Rev. Lett. **98**, 126401 (2007).

[11] J. Guo, J. Shi, and E. W. Plummer, Phys. Rev. Lett. **94**, 036105 (2005).

[12] I. Brihuega *et al.*, Phys. Rev. Lett. **95**, 206102 (2005).

[13] O. Bunk *et al.*, Phys. Rev. Lett. **83**, 2226 (1999).

[14] M. Göthelid *et al.*, Surf. Sci. **271**, L357 (1992).

[15] J. Avila *et al.*, Phys. Rev. Lett. **82**, 442 (1999).

[16] J. S. Okasinski *et al.*, Phys. Rev. B **69**, 041401(R) (2004).

[17] M. E. Davila *et al.*, Phys. Rev. B **70**, 241308 (2004).

[18] T. Lee *et al.*, Phys. Rev. Lett. **96**, 046103 (2006).

[19] F. Ronci *et al.*, Phys. Rev. Lett. **95**, 156101 (2005).

[20] The consistency of our measurements was tested by measuring $I(V)$ spectra on a Ge(111) bare surface obtaining an unambiguous gap with null current within the error around E_F.

[21] J. A. Stroscio, R. M. Feenstra, and A. P. Fein, Phys. Rev. Lett. **57**, 2579 (1986).

[22] P. Gori *et al.*, arXiv:0804.0386v1.

[23] R. I. G. Uhrberg and T. Balasubramanian, Phys. Rev. Lett. **81**, 2108 (1998).

[24] It has to be noted that the 5 K STS spectra in Fig. 4 were collected at a lower tunneling current set point. Since there are no observable differences between the spectra collected at 0.10 nA and the one collected at 0.05 nA it is possible to rule out any artifact induced by the tunneling current as in Ref. [25], for example.

[25] V. Ramachandran and R. M. Feenstra, Phys. Rev. Lett. **82**, 1000 (1999).

[26] T. Sato, M. Iwatsuki, and T. Tochihara, J. Electron Microsc. **48**, 1 (1999).

[27] K. Hata, Y. Sainoo, and H. Shigekawa, Phys. Rev. Lett. **86**, 3084 (2001).

[28] S. Yoshida *et al.*, Phys. Rev. B **70**, 235411 (2004).

[29] K. Cho and J. D. Joannopoulos, Phys. Rev. Lett. **71**, 1387 (1993).

[30] K. Seino, W. G. Schmidt, and F. Bechstedt, Phys. Rev. Lett. **93**, 036101 (2004).

[31] H. Kawai *et al.*, J. Phys. Soc. Jpn. **71**, 2192 (2002).

[32] H. Kawai and O. Narikiyo, J. Phys. Soc. Jpn. **73**, 417 (2004).

[33] It is worthwhile noting that the low temperature fluctuation of the Si dimers at the Si(100) surface can also be excited by the electron beam in low energy electron diffraction (LEED) measurements as reported in T. Shirasawa, S. Mizuno, and H. Tochihara, Phys. Rev. Lett. **94**, 195502 (2005). Assuming a similar behavior of the Si(100) and α-Sn/Ge(111) surfaces it is possible to explain why the first STM observation of the LT-$\sqrt{3}$ was supported by LEED patterns showing a $\sqrt{3} \times \sqrt{3}$ periodicity.

[34] I. Yi *et al.*, Appl. Surf. Sci. **253**, 3072 (2007).