FIRST-PRINCIPLES CALCULATIONS OF NANOSTRUCTURED SURFACES

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I will review some of the complex surface reconstructions that have been studied recently in our group. I will mainly focus of the quasi-one-dimensional reconstructions formed by the deposition of submonolayer amounts of gold on Si(111) and vicinal Si(111) surfaces [1]. If possible, I will briefly present preliminary results on the structure and instabilities induced by a monolayer of water on NaCl(001) and its implications in two very different scenarios: AFM visualization of the NaCl(001) surface at 30-40% relative humidity and atmospheric chemistry [2].

The gold atoms on vicinal Si(111) surfaces form very thin wires, and therefore the electronic structure (as measured with angle resolved photoemission, ARP) exhibits a strong one-dimensional behaviour. An example of these systems that has attracted much attention in recent years is the stepped Si(557)-Au surface. In this case the gold atoms are known to form one monatomic wire in the middle of the terrace of each step. The wires run parallel to the step edges. If the structure of other surfaces is similar we can get an interesting playground: increasing or decreasing the miscut angle of the initial silicon substrate and, therefore, the size of the steps, we can change the interaction between the gold chains in neighbouring steps. In this way, it is believe that it would be possible to control de degree of one-dimensionality of these systems, and study the transition between two-dimensional and one-dimensional behaviours. Furthermore, since the silicon substrate is relatively rigid, there is hope that it would be possible to overcome the Peierls distortion and it would be possible to obtain truly one-dimensional metals in these systems. For these reasons these surfaces have attracted a lot of attention, both theoretically and experimentally, in recent years. We have study some of these reconstructions using first-principles electronic structure calculations, and will present some our results for three characteristic systems: the stepped Si(557)-Au and Si(553)-Au surfaces, and the flat Si(111)-(5x2)-Au reconstructions.

The photoemission spectrum of the Si(557)-Au surface is dominated by a one-dimensional band. This band is found to split in two peaks near the Fermi energy. This was initially interpreted as a signature of the spin-charge separation characteristic of a one-dimensional metal [3]. Although new experimental evidence indicated that the bands were not completely consistent with the theoretical predictions for a one-dimensional electron gas, the origin of these *twin* bands was a mystery for several years. Our recent calculations [4] show that it is the spin-orbit splitting what produces these two bands. These spin-splitting may have relevance for future device applications since simple arguments show that such splitting should occur in any surface band formed from heavy atoms. We have also shown that the apparent Peierls-like transition observed in this surface [5] using scanning tunnelling microscopy might be explained as a result of the dynamical fluctuations of the step-edge structure, which are quenched as the temperature is decreased [4]. The implications of the fluctuations for the observed metal-insulator transition [5] will be also discussed in our contributions.

The stepped Si(553)-Au surface has also attracted a lot of attention in recent times. Its ARP spectrum is very similar to that of the Si(557)-Au surface and is dominated by two neighbouring one-dimensional bands with approximately ½ occupation. However, a very peculiar one-dimensional band with a ¼ occupation is also observed [6]. Neither the structure

of this surface, nor the origin of this band is known. We have examined the energetics and the band structure of several structural models constructed from a plausible analogy with the structure of the Si(557)-Au surface [7] and ruled out a model proposed recently from the analysis of X-ray diffraction experiments [8]

Finally we analyze the Si(111)-(5x2)-Au reconstruction with our density functional calculations. In spite of the fact that this reconstruction has been studied with several experimental techniques since the late sixties, its structure is still a matter of debate. Due to the large size of the reconstructions, only very recently the first electronic structure calculations of the surface appeared. Based on this type of calculations, S. C. Erwin has proposed a structural model for this reconstruction [9]. We have studied the relative stability and the electronic band structure of several different models, including the Mark-Plass and Erwin model [10].We have found a new model which, at least within the theoretical approach used, is more stable than any other proposal to date.

References:

[1] J. N. Crain *et al.*, Phys. Rev. B **69**, 125401 (2004), and references therein.
[2] P. Cabrera-Sanfelix, A. Arnau, G. Darling and D. Sánchez-Portal, in preparation.
[3] P. Segovia, D. Purdie, M. Hengsberger, and Y. Baer, Nature (London) **402,** 504 (1999).
[4] D. Sánchez-Portal, S. Riikonen, and R. M. Martin, Phys. Rev. Lett. **93**, 146803 (2004).
[5] J. R. Ahn, H. W. Yeom, H. S. Yoon, and I.-W. Lyo, Phys. Rev. Lett. **91**, 196402 (2003).
[6] J. N. Crain *et al.*, Phys. Rev. Lett. **90**, 176805 (2002).
[7] S. Riikonen and D. Sánchez-Portal, Nanotechnology **16**, S218 (2005)
[8] S. Riikonen and D. Sánchez-Portal, Surface Science **600**, 1201 (2006)
[9] S. C. Erwin, Phys. Rev. Lett. **91**, 206101 (2003)
[10] S. Riikonen and D. Sánchez-Portal, Phys. Rev. B **71**, 235423 (2005)

Figures:

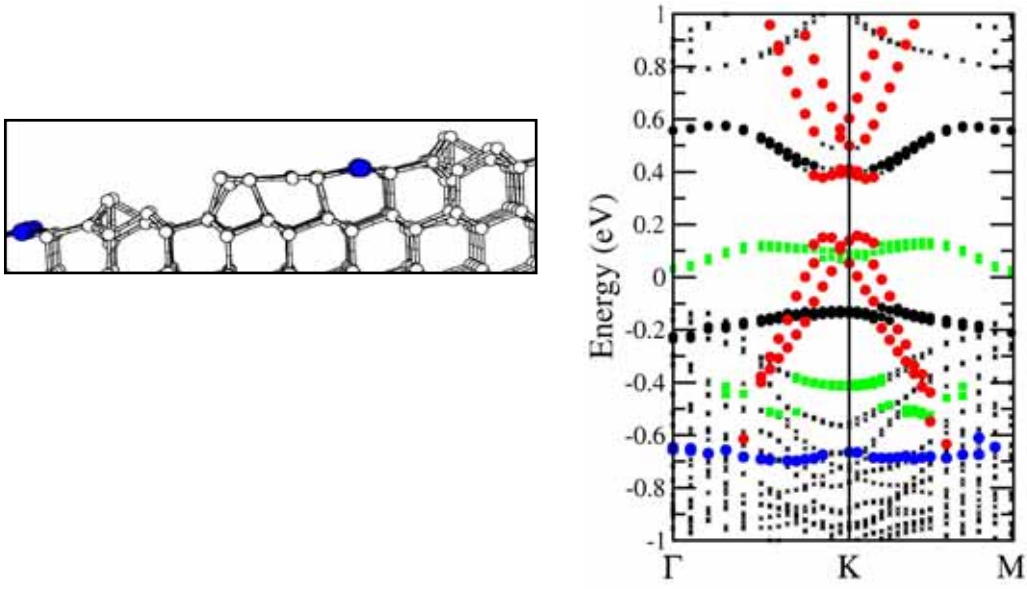

Figure. Predicted structure for the Si(557)-Au along with the calculated band structure once the spin-orbit interaction is included [4]. The large solid circles represent the gold atoms.