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Inverse photoemission studies of two quasi-one-dimensional overlayer systems

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Abstract

Photoemission studies of Au chains on Si(111) surfaces, Si(111)–Au(5×2), have identified a 'metallic' surface band parallel to the chains. The signature of the metallic nature is strong emission intensity at the Fermi level. In contrast, it has recently been established that bulk quasi-one-dimensional systems have vanishing emission intensity at the Fermi level. To further our understanding of these low dimensional systems, we have studied Si(111)–Au(5×2) and Si(111)–In(4×1), which is also quasi-one-dimensional, with inverse photoemission. © 1998 Elsevier Science B.V.

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1. Introduction

In the study of bulk quasi-one-dimensional (1D) systems with photoemission, one of the most intriguing results of the last decade has been the discovery that, even in the metallic state, there is vanishing emission intensity at the Fermi level [1–4]. A consensus has emerged that this is a natural consequence of the fact that quasiparticles do not form in these systems and that Fermi liquid theory [5,6], which has been successful in describing the metallic state in many metals in 3D, and also some strongly anisotropic quasi-2D metals [7], is inappropriate. In light of this, it is interesting that recent photoemission studies of quasi-1D Au chains on Si(111) surfaces [8] have identified strong emission intensity at the Fermi level. This arises from a partially filled 'metallic' surface band which is visible in photoemission spectra collected parallel to the Au chains. This begs the question, *do quasi-1D systems on surfaces belong to the same physical class as bulk quasi-one-dimensional systems*? To gain more information about these systems, we have performed inverse photoemission studies of the Si(111)–Au(5×2) and the Si(111)–In(4×1) quasi-1D systems. These studies complement the photoemission studies that have already been performed on both the Au [8] and In [9] overlayer systems and they give us a more complete picture of the electronic structure of these interesting systems.

2. Inverse photoemission detector

The inverse photoemission experiments were performed with a high sensitivity bandpass photon detector [10] and a low-energy electron gun [11]. The detector is a Geiger–Müller tube which utilizes dimethyl ether as the detection gas and a MgF_2 entrance window to produce a bandpass centered on 10.6 eV. The detector bandwidth was estimated to be

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< 0.6 eV FWHM from the Fermi level onset of evaporated polycrystalline Au.

3. Gold chains on silicon

The C_{3v} symmetry of the Si(111) surface allows three equivalent 5×2 domains to form. However, a vicinal miscut encourages a single domain to grow with the Au chains parallel to the surface steps [12]. The samples were p-type Si(111) wafers, with resistivities of $\approx 5\ \Omega$ cm, miscut by $4 \pm 0.5°$ towards $[11\bar{2}]$. On the vicinal wafers, the [111] axis of the sample is rotated from the surface normal. To compensate, the sample was mounted in a sample holder that counter-rotated the sample by 4° to align the [111] surface with the face of the sample holder.

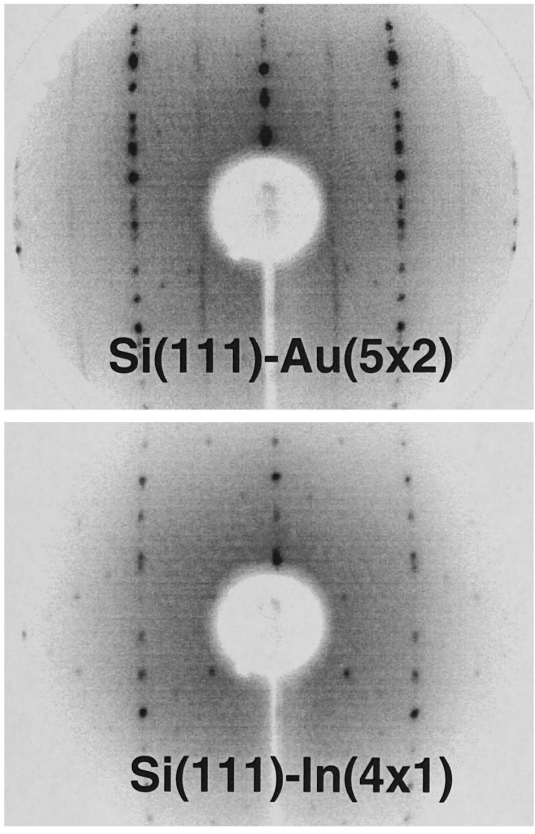

Fig. 1. (a) A LEED image of a single domain Si(111)–Au(5×2) sample collected at 82 eV. The $[11\bar{2}]$ direction is upwards. (b) A LEED image of a predominantly single domain Si(111)–In(4×1) sample collected at 79 eV. $[\bar{1}\bar{1}2]$ is upwards. In both cases the metal chains are horizontal.

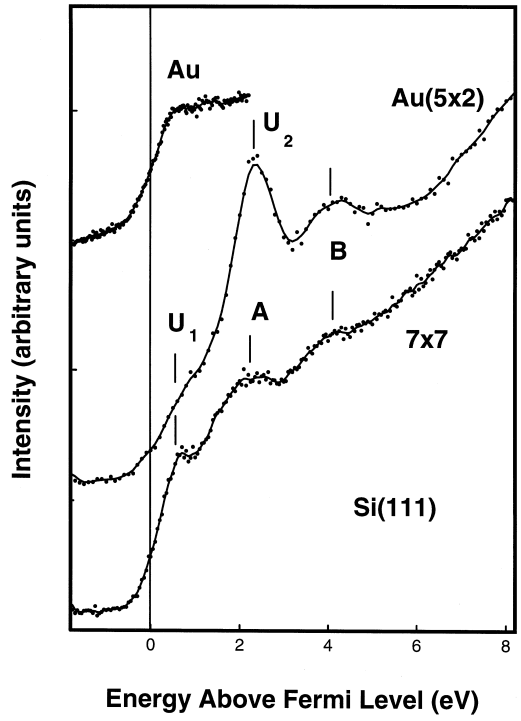

Fig. 2. Inverse photoemission spectra from: evaporated polycrystalline gold, Si(111)–Au(5×2) and Si(111)(7×7). The points are unsmoothed raw data. The Au(5×2) and (7×7) spectra have not been scaled, to allow comparison of absolute intensities.

Clean, well ordered Si(111) surfaces were prepared by flashing the Si sample to ≈ 1100°C by direct current heating. All currents were applied in the $[\bar{1}\bar{1}2]$ (uphill) direction to avoid step bunching. At electron energies of 40 eV the 1×1 LEED beams were split, indicating that the steps formed a regularly spaced superlattice.

The overlayers were formed by depositing Au onto a Si(111) surface that was heated to ≈ 650°C. Coverages were measured to lie in the range 0.40–0.45 ML, where 1 ML is defined to be the areal atomic density of an ideally terminated Si(111) plane (7.83×10^{14} cm^{-2}).

A LEED pattern from the Si(111)–Au(5×2) surface is presented in Fig. 1a. The $[11\bar{2}]$ direction is upwards and the electron energy is 80 eV. The image has 5×1 symmetry with half-order streaks. It has previously been suggested that the half-order streaks are a consequence of lack of registry between Au chains [13,14] and STM studies have indeed found row slippage [12,15,16].

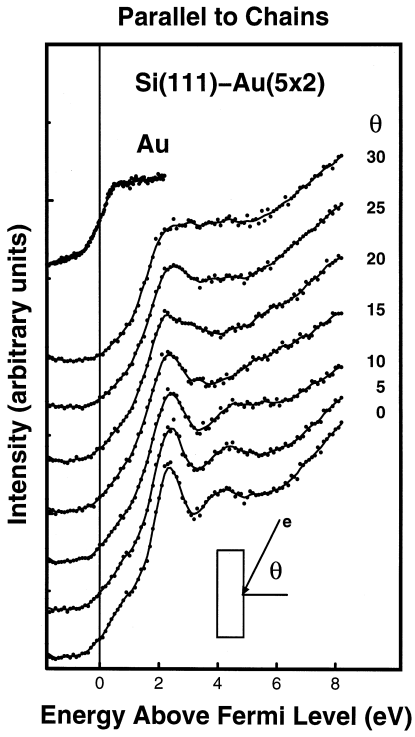

Fig. 3. Inverse photoemission spectra collected parallel to the Au chains, from single domain Si(111)–Au(5×2). The numbers along the r.h.s. are the electron incidence angles in degrees. No states cross the Fermi level.

In Fig. 2, we present a comparison of two normal-incidence inverse photoemission spectra taken from the Si(111)(7×7) and the Si(111)–Au(5×2) systems. These probe the $\bar{\Gamma}$ point of the surface Brillouin zone. Also shown is the Fermi level emission from freshly evaporated polycrystalline Au which was in electrical contact with the sample. The first thing to note is that on the Si(111)–Au(5×2) surface there are no unoccupied surface states at the Fermi level. Also note the similarity in binding energy of the state labeled U_1 on the Si(111)(7×7) spectrum and the corresponding state on Si(111)–Au(5×2). On Si(111)(7×7), the features labeled A and B have been identified as bulk transitions [17]. Furthermore, the feature marked U_2 on the Si(111)–Au(5×2) spectrum is very similar to a feature that is produced by Si–Au bonding on the Si(111)–Au$(\sqrt{3}\times\sqrt{3})$R30° system [17] and it overlaps the bulk transition labeled A.

In Fig. 3 we show a sequence of inverse photoemission spectra that were collected by moving the electron gun in the $[1\bar{1}0]$ azimuth, parallel to the Au chains. Once again, there are no states at the Fermi level. Perpendicular to the Au chains, the dispersion of all the features is similar, and so the spectra have not been reproduced here. Again there is no evidence for a band crossing the Fermi level.

4. Indium chains on silicon

Predominantly single domain Si(111)–In(4×1) overlayers were grown on vicinal Si(111) surfaces by depositing In on a surface heated to ≈ 395°C (Fig. 1b). The Si(111) wafers were n-type with a resistivity of $\approx 5\ \Omega$ cm, miscut by 3 ± 0.5° towards $[\bar{1}\bar{1}2]$. Once again, the sample was counter-rotated by the vicinal offcut angle.

In Fig. 4 we show a series of spectra that were

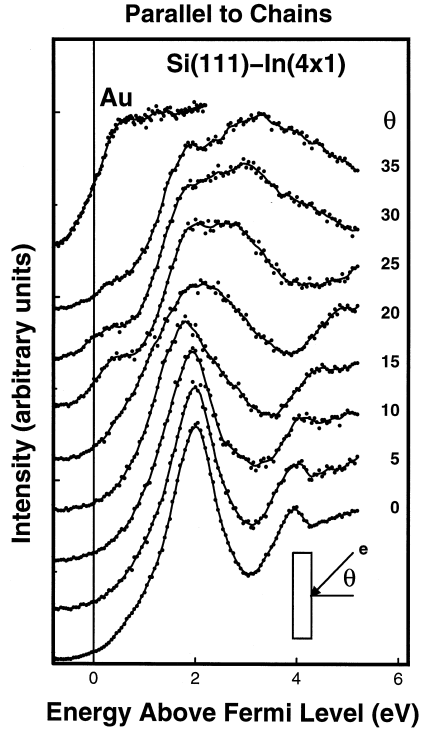

Fig. 4. Inverse photoemission spectra, from single domain Si(111)–In(4×1), collected parallel to the In chains. The numbers along the r.h.s. of the figure are the electron incidence angles. Notice the strong Fermi level emission, beginning at 25° (~ 0.5 Å^{-1}).

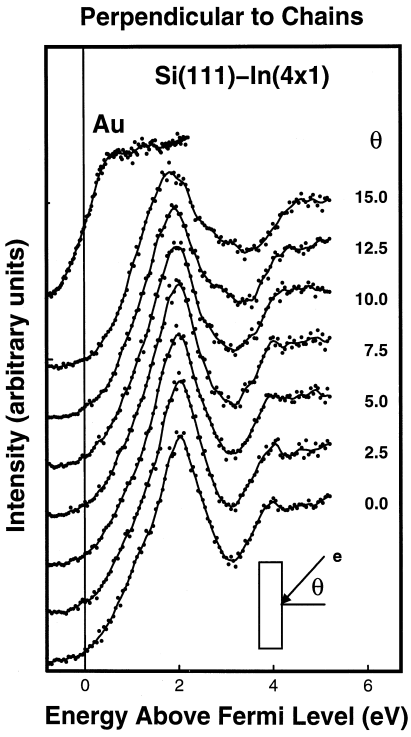

Fig. 5. Inverse photoemission spectra collected perpendicular to the In chain direction, from single domain Si(111)–In(4×1). The numbers along the r.h.s. of the figure are the electron incidence angles. Notice the lack of emission at the Fermi level. The zone boundary in this direction is close to 10°.

collected from the Si(111)–In(4×1) surface parallel to the In chains. The presence of a Fermi level crossing, consistent with recent photoemission studies [9], is clearly visible in the spectra. In contrast, inverse photoemission spectra collected perpendicular to the In chains (Fig. 5) show no states at the Fermi level, indicating that the system is truly one dimensional.

5. Electronic structure

Although, the Si(111)–Au(5×2) system has already been described as an 'unusual' system [8], the system may be more unusual than originally thought. First consider the Au chains. The photoemission measurements [8] have detected a small, 500 meV, dispersion of the 5d-band, indicating that the Au wavefunctions are not totally atomic-like. However, the bands associated with the 5d levels do not appear to extend above a binding energy of 3 eV. Therefore, it is unlikely that they strongly contribute to the electronic structure at the Fermi level. Instead it is more likely that the 6s orbitals overlap to form a metallic band. However, there is no evidence from our inverse photoemission experiments for an s-like band above the Fermi level. So the metallic band that has been observed with photoemission [8] may not simply be attributed to the Au chains.

Another possibility is that the metallic band originates from the regions between the Au chains. The electronic structure of the Si(111)(7×7) surface, which acts as a starting point for the Si(111)–Au(5×2) system, is not dissimilar to that of the Si(111)–Au(5×2). For example, the 7×7 system has a relatively flat metallic surface band. The Fermi level is located at the top of the band, making it difficult to detect in inverse photoemission. This is analogous to the Si(111)–Au(5×2) system. In the 7×7 case, both the occupied (S_1) and unoccupied (U_1) bands have been shown to be Si adatom derived. Both ab initio density functional theory calculations [18] and STM studies of the surface [18] have made this association. Consequently the occupied band that crosses the Fermi level may be Si adatom derived. This view would find support from STM studies of the system that have found that the density of intense surface features on Si(111)–Au(5×2) is larger than that calculated from the Au coverage [12]. We note that, although there is spectroscopic evidence for the presence of Si adatoms, until the surface structure is solved, the existence of Si adatoms within or between the Au chains is a conjecture. If Si adatoms are located between the Au chains, the Au chains may inhibit the formation of a Si adatom band perpendicular to the chains. The appealing picture of a metallic band arising from the overlap of Au wavefunctions may be far from the truth.

Turning now to the Si(111)–In(4×1) system. There is a clear Fermi level crossing in the inverse photoemission spectra near $\overline{X}$. STM topographs [19] have revealed a double row structure with a double row separation of 13.4 Å. Once again, the structure of this system is still to be determined. From the photoemission and inverse photoemission studies it is clear that the system is not autocompensated and does not satisfy the electron counting rule [20].

To summarize, we have studied two different metallic quasi-1D systems using inverse photoemission. In the case of Si(111)–Au(5×2), the metallic band is not observed and we must rely on photoemission studies to establish the metallic nature of the system. In contrast, studies of Si(111)–In(4×1) reveal a clear Fermi level crossing near $\overline{X}$. The fact that both systems are metallic and not insulators [21] suggests that the bandwidth of the metallic band (W) is significantly larger than the on-site correlation energy ($W > U$). The electron correlations may also be influenced by the fact that the surface bands may not be truly isolated from the substrate. Unfortunately, neither of the systems have been studied with sufficient resolution to unambiguously determine whether the Fermi level region possesses the characteristic power law behavior, as seen in bulk 1D materials. Although it is presently impossible to perform inverse photoemission with sufficiently high resolution, high resolution photoemission studies ($\Delta E < 40$ meV) of these systems are certainly possible.

Acknowledgements

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