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Electrical conductance at initial stage in epitaxial growth of Pb on modified Si(1 1 1) surface

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Abstract

The electrical conductance and RHEED intensities as a function of the coverage have been measured during Pb depositions at 105 K on Si(1 1 1)–(6 × 6)Au with up to 4.2 ML of annealed Pb. The experiments show the strong influence of used substrates on the behavior of the conductance during the epitaxy of Pb atoms, especially for very initial stage of growth. Oscillations of the conductance during the layer-by-layer growth are correlated with RHEED intensity oscillations. The analysis of the conductance behavior is made according to the theory described by Trivedi and Aschcroft [N. Trivedi, N. Aschcroft, Phys. Rev. B 38 (1988) 12298].

Keywords: Electrical transport measurements; Electron transport; Thin film

1. Introduction

The transport properties of a clean, well ordered surface with deposited unreactive metals are an important topic of surface physics. Since the detailed surface structure may modify the transport mechanism dramatically, the atomic structure and transport properties have to be measured simultaneously. Most studies of electronic transport in such structures have been conducted for metals coverage higher than 1 monolayer (ML) [1–5]. Jałochowski et al. concentrated on quantum size effect studies in thin epitaxial Pb and Pb–In films grown on Si(1 1 1) reconstructed surface [1,2]. Pfennigstorf et al. [3,4] studied electronic transport in ultrathin epitaxial Pb films on Si(1 1 1)-7 × 7 and on Si(111)–$\sqrt{3} \times \sqrt{3}$Pb surfaces. These authors obtained useful information about conduction mechanisms for the coverage higher than 1 ML. They did not mention the initial decreasing in the conductance for the coverage lower than 1 ML. Hasegawa and Ino monitored the conductance dependence on the substrate–surface structures and epitaxial growth modes at initial stages of Ag and Au depositions on a Si(1 1 1) surface at room temperatures [5]. Authors paid little attention to the conductance changes for the coverage lower than 1 ML. Recently, Pfennigstorf et al. [6] have concentrated on the correlation of structural properties with the measured conductance. The conductance of a Pb film during deposition at 15 K on a film of tenths ML, which had been annealed (recrystallized), showed the strong decrease during the first half monolayer. The behavior of the conductivity as a function of film thickness is a result of the competition between the thickness increment and the roughness variation. Also the electron mean free path as well as the Fermi energy and electron density are changed (oscillate) during the layer-by-layer growth but for thicker films these functions tend to constant values which correspond to the bulk material [7,8]. Understanding of the electron transport characteristics for low coverage is still limited.

In this paper, we focus on the measurements of the electrical conductance at initial stage in epitaxial growth of Pb on Si(1 1 1)–(6 × 6)Au/Pb (i.e. Si(1 1 1)–(6 × 6)Au with up to 4.2 ML of annealed Pb) surface using four-point probe.

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By relating these measurements to the observed simultaneously reflection high energy electron diffraction (RHEED) intensity changes we have obtained a better understanding of the origin of the conductance behavior. The analysis of the conductance behavior is made according to the theory described by Trivedi and Aschcroft [7].

2. Experimental setup and results

The measurements were performed in a UHV chamber with a base pressure in order of 5×10^{-11} Torr. The structure of the substrate and the deposition of Pb were monitored by the RHEED system. An n-type Si(1 1 1) wafer of around 25 Ω cm resistivity and $18 \times 4 \times 0.4\ mm^3$ size was mounted on a pair of Mo rods and clamped with Ta stripes. Electrical conductivity was measured in situ by four-point probe method. An alternating current: $I =$ 2 μA, 17 Hz was sent through the outer-most Ta clamps contacts, while the voltage ac was measured across the inner two W wires kept in elastic contact with the wafer. The voltage electrode spacing of 1.5 mm were used in the experiments. The sample holder was mounted to SuperTran-VP continuous flow cryostat cold finger (Janis Research Comp. Inc.). The temperature of the sample has been measured with the Au–Fe chromel thermocouple in touch with a wafer. Before each measurement run, the surface was cleaned to obtain a clear Si(1 1 1)-7 × 7 RHEED pattern, by few flash heating for 5 s with a direct current of 13.5 A through the sample. In order to prepare the Si(1 1 1)–(6 × 6)Au surface structure, 1.3 ML of Au were deposited on Si(1 1 1)-7 × 7 superstructure. Annealing for 1 min at about 950 K and slow cooling to room temperature (10 K/min) resulted in the appearance of a sharp (6 × 6)Au superstructure RHEED pattern. Si(1 1 1)–(6 × 6)Au with up to 4.2 ML of Pb deposited at 105 K have been heated up to 300 K in order to increase crystalline order. After recrystallization the surface has been cooled to about 100 K for further deposition of Pb. The conductance has been measured simultaneously with deposition. The amount of deposited material in units of monolayer ($ML = 7.8 \times 10^{14}\ atoms/cm^2$) was monitored with a quartz crystal oscillator. RHEED specular beam intensity oscillation during monolayer-by-monolayer growth has been used for calibration. Pb was evaporated from Ta crucible at rate 0.02 ML/min.

Fig. 1 shows the RHEED specular beam intensity changes during Pb deposition at 105 K on Si(1 1 1)–(6 × 6)Au surface at a glancing angle 0.5° in the azimuth of Si(1 1 1) surface (upper panel). The letters A–E mark the coverage at which deposition of Pb was terminated. After annealing up to 300 K and cooling down to 100 K the conductance increases due to improved crystalline order of the layer. For example in the case of the substrate Si(1 1 1)–(6 × 6)Au with deposited 4.2 ML Pb after annealing the conductance increases by 40% at 100 K. Moreover, the conductance of the substrate Si(1 1 1)–(6 × 6)Au with annealed Pb is about two times larger than the conductance

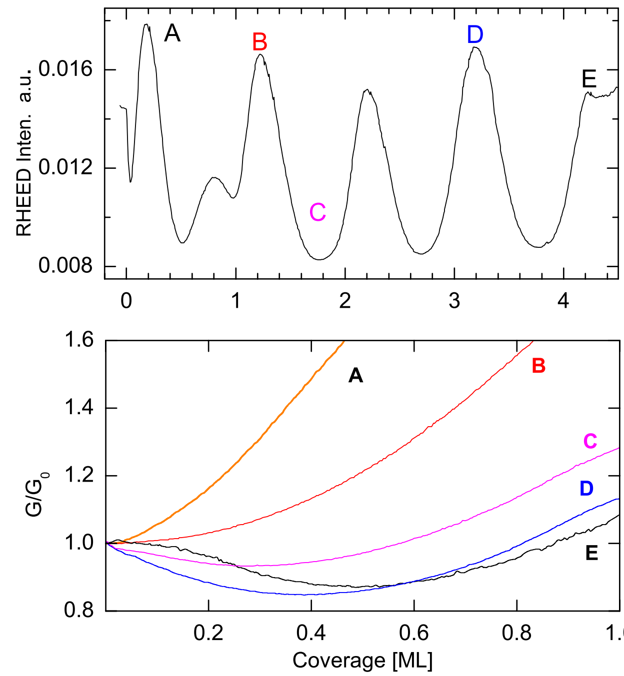

Fig. 1. RHEED specular intensity (upper panel) during growth of Pb on Si(1 1 1)–(6 × 6)Au surface and the relative conductance (lower panel) during growth of Pb on Si(1 1 1)–(6 × 6)Au surface with annealed Pb layers. The letters A–E correspond to the coverage 0.2, 1.2, 1.8, 3.2 and 4.2 ML, respectively, at which deposition was terminated and Pb was annealed.

of Si(1 1 1)–(6 × 6)Au substrate. On such prepared surface further deposition of Pb was carried out. The coverage dependence of the relative conductance G/G_0 during deposition of Pb on different substrates is shown in Fig. 1, the lower panel. Here G_0 means the conductance of the system with annealed Pb atoms, i.e. Si(1 1 1)–(6 × 6)Au/Pb. For the substrate Si(1 1 1)–(6 × 6)Au/⩽1.2 ML Pb, i.e. (6 × 6)Au with less than 1.2 ML of annealed Pb, the relative conductance increases with increasing Pb coverage (cf. lines A and B). It suggests that annealed Pb atoms form small, few-atom clusters which are situated in (6 × 6)Au cells, [11]. In such a case small amount of deposited Pb atoms cannot disturb the conductance as the electron mean free path is few times smaller than for the bulk material. On the other hand, the decrease of the conductance starts from the beginning of the Pb deposition on the substrate with recrystallized Pb coverage higher than 1.2 ML (cf. lines C–E). The conductance reaches a minimum value and afterwards increases. In this case annealed Pb atoms form rather smooth surface, especially for integer monolayer, and the electron mean free path is close to the bulk one. Deposition of Pb atoms on such prepared structure causes the roughness of the surface to increase and the conductance to decrease. But for the higher coverage these added atoms form 1 ML high clusters and cause the conductivity increases.

The RHEED experiments confirm the above conclusions. In Fig. 2 we show the conductance (the lines start from the value of 1 for the coverage equals to 0) and

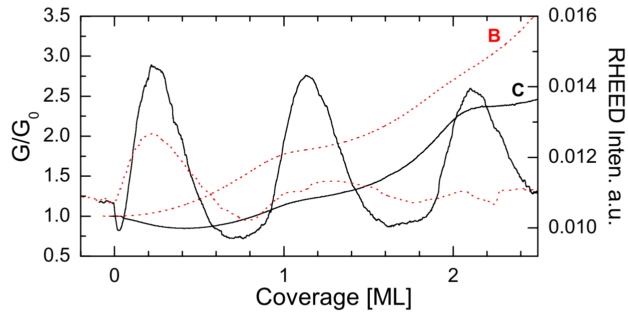

Fig. 2. RHEED specular intensity (right side) and relative conductance (left side) during growth of Pb on Si(1 1 1)–(6 × 6)Au surface with deposited and annealed 1.2 (broken lines) and 1.8 ML (solid lines) of Pb, respectively. The relative conductance curves start from the value of 1 (for the coverage equals to 0).

RHEED intensity of specular spot dependence on Pb coverage on Si(1 1 1)–(6 × 6)Au/1.2 ML Pb (broken lines) and Si(1 1 1)–(6 × 6)Au/1.8 ML Pb (solid lines) (i.e. (6 × 6)Au with 1.2 and 1.8 ML of annealed Pb) surfaces, respectively. In both cases the oscillations of the RHEED start from the beginning (layer-by-layer growth), but for the substrate with recrystallized Pb coverage equal or lower than 1.2 ML these oscillations go out with the Pb coverage, cf. the broken RHEED line. In this case the thickness dependence of the conductance is not monotonic. It should be noted that for the substrate with more than 1.2 ML of annealed Pb, the conductance oscillations (quantum size effect) have been observed.

3. Theoretical description

To explain the behavior of the conductance of Si(1 1 1)-(6 × 6)Au/Pb during deposition of Pb atoms we have used the theory by Trivedi and Aschroft [7]. The conductance of thin films in the presence of bulk impurity scattering and surface roughness scattering can be expressed as follows:

$$\sigma_{xx} = \frac{e^2 k_F}{\hbar \pi^2} \frac{1}{\kappa} \sum_{m=1}^{n_c} \frac{1 - m^2/\kappa^2}{\frac{2n_c+1}{k_F l_0 \kappa} + \left(\frac{\delta d}{d}\right)^2 \frac{s(n_c) m^2}{3\kappa}}, \quad (1)$$

where k_F is the Fermi wave vector, $\kappa = k_F d/\pi$, l_0 is the electron mean free path of the system, $n_c = \mathrm{Int}(\kappa)$ and

$$s(n_c) = (2n_c + 1)(n_c + 1)n_c/(3\kappa^3). \quad (2)$$

The function δd is the root-mean-square deviation of the mean film thickness d. This function can be obtained from the RHEED intensity oscillations during the growth of the layer [1,9] and is given by [10]:

$$(\delta d)^2 = \sum_{n=0}^{\infty} (n - t/\tau)^2 (\Theta_n - \Theta_{n+1}), \quad (3)$$

where τ is the deposition time of one layer (in calculations we set $\tau = 1$) and Θ_n is the coverage of the nth layer. Here we consider only the Pb coverage lower than 1 ML (the second layer is forbidden) and in this case the coverage function can be obtained analytically, $\Theta_{n+1} = 1$ and $\Theta_{i>n+1} = 0$, where n is the number of full substrate layers on which Pb atoms are deposited. Also the roughness function $(\delta d)^2$ has analytical parabolic form with $\delta d(t = 0) = \delta d(t = 1) = 0$ and the maximal value equals to 0.5 which corresponds to the half-coverage of the layer. In the calculations we assume that the thickness of the considered system can be expressed in the following form:

$$d = nd_0 + td_0, \quad t \in \langle 0, 1 \rangle, \quad (4)$$

where t means the Pb coverage and nd_0 corresponds to annealed Pb layers (or Si(1 1 1)–(6 × 6)Au/Pb layers). We express all distances in d_0 units and k_F in $1/d_0$ units. Moreover, we assume that for the substrate Si(1 1 1)–(6 × 6)Au/(<1.2 ML Pb) the electron mean free path is limited mainly by the dimension of (6 × 6)Au cells and, as was deduced from our experiment, it is a few times smaller than the mean free path for the substrate Si(1 1 1)–(6 × 6)Au/(>1.2 ML Pb).

In Fig. 3 we show the relative conductance as a function of the Pb coverage for two kinds of substrates, i.e. n-annealed Pb layers (upper panel) and (6 × 6)Au with n-annealed Pb layers (lower panel) – see inside figures. Here we consider the same effective d_0 for (6 × 6)Au and for one Pb layer. It is interesting that for $n = 1$ layer of annealed Pb the relative conductance increases with increasing Pb coverage – for both substrates. In this case we set $l_0 = 7$, which e.g. for $d_0 = 2.5$ Å corresponds to the mean free path $l \simeq 18$ Å. For $n \geqslant 2$ and greater mean free path, $l_0 = 25$ (which corresponds to the bulk one), the conductance decreases at the beginning and then for the higher coverage

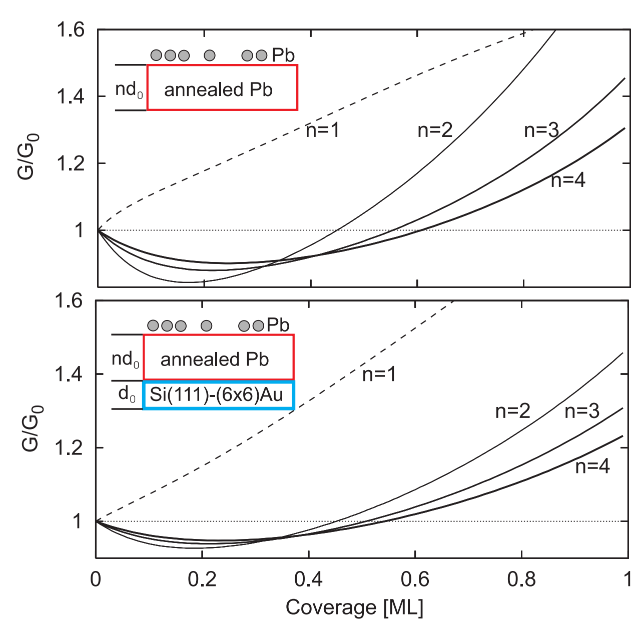

Fig. 3. The relative conductance versus the coverage of Pb atoms (up to 1 ML) on the surface consisted of n-annealed Pb layers (upper panel) and (6 × 6)Au with n-annealed Pb layers (lower panel). The mean free path is $l_0 = 25$ for $n = 2,3,4$ and $l_0 = 7$ for $n = 1$, $k_F = \pi$. To place the inside figure in the upper panel the broken line was divided by the factor 8 and shifted to 1.

it increases. It is worth mentioning that the minimum of the conductance appears when the electron mean free path is rather large (equal about the mean free path of the bulk material). For very small l_0 this minimum does not appear.

The results obtained for the substrate Si(1 1 1)–(6 × 6)Au/Pb – the lower panel in Fig. 3 – are in good agreement with the experiment, cf. Fig. 1, the lower panel. For the substrate with n annealed Pb layers – upper panel – this agreement is somewhat worse as the curve for $n = 1$ increases very rapidly (in Fig. 3 this curve is divided by the factor 8) and also the curve for $n = 2$ increases too fast in comparison to experimental data.

4. Conclusions

The electrical conductance at initial stage in epitaxial growth of Pb on the surface Si(1 1 1)–(6 × 6)Au/Pb (with up to 4.2 ML of annealed Pb) has been measured using four-point probe. In comparison with similar experiments, e.g. [6], where only the conductance minimum was observed, our results show that for the substrate with lower than 1.2 ML of annealed Pb the conductance increases with Pb coverage whereas for the substrate Si(1 1 1)–(6 × 6)Au with more than 1.2 ML of annealed Pb the minimum in the conductance appears. This effect can be understood in terms of electron mean free path of these substrates. For the substrate Si(1 1 1)–(6 × 6)Au/>1.2 ML Pb the mean free path is rather large, as in the bulk material and in this case the minimum in conductance as a function of Pb coverage appears. In the case of the substrate Si(1 1 1)–(6 × 6)Au/<1.2 ML Pb the electron mean free path is a few times smaller than in the bulk Pb and the minimum in conductance does not appear – small amount of Pb atoms cannot disturb the conductance. These adatoms cause the conductance to increase from the beginning of the epitaxy. Moreover, the RHEED intensity changes during the growth of the Pb layer show the difference for both surfaces. The oscillations of the RHEED start from the beginning for both kinds of surfaces (layer-by-layer growth) but for the substrate with recrystallized Pb coverage equal or lower than 1.2 ML these oscillations go out with the Pb coverage.

Acknowledgements

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References

[1] M. Jałochowski, E. Bauer, H. Knoppe, G. Lilienkamp, Phys. Rev. B 45 (1992) 13607.
[2] M. Jałochowski, M. Hoffman, E. Bauer, Phys. Rev. B 51 (1995) 7231.
[3] O. Pfennigstorf, K. Lang, H.-L. Günter, M. Henzler, Appl. Surf. Sci. 162 (2000) 537.
[4] O. Pfennigstorf, A. Petkova, H.L. Guenter, M. Henzler, Phys. Rev. B 65 (2002) 45412.
[5] S. Hasegawa, S. Ino, Surf. Sci. 283 (1993) 438.
[6] O. Pfennigstorf, A. Petkova, Z. Kallassy, M. Henzler, Eur. Phys. J. B 30 (2002) 115.
[7] N. Trivedi, N. Aschroft, Phys. Rev. B 38 (1988) 12298.
[8] Z. Wang, S. Wang, S. Shen, S. Zhou, Phys. Rev. B 55 (1997) 10863.
[9] P.I. Cohen, G.S. Petrich, P.R. Pukite, G.J. Whaley, S. Arrott, Surf. Sci. 216 (1989) 222.
[10] M. Horn, V. Götter, M. Henzler, in: P.K. Larsen, P.J. Dobson (Eds.), Reflection High-Energy Electron Diffraction and Reflection Electron Imaging of Surface, Plenum, NY, 1988, p. 463.
[11] M. Jałochowski, Progr. Surf. Sci. 74 (2003) 97.