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Optical anisotropy of Pb nanowires on Si(533) surface

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Ultrathin Pb films on Si(533)/Au surface deposited at low temperatures are investigated by STM, RHEED, and by optical reflectance difference spectroscopy (RDS). In particular, we report the observation by RDS of an anisotropy in the optical reflectance of the Si(533) surface covered by Pb nanowires. It is found that the optical anisotropy originates from anisotropy of electrical conductivity of the Si(533) substrate and the anisotropy of Pb nanowires formed on this surface.

1. Introduction

The vicinal surfaces of Si(111) show unusual properties. The surface of the crystal cut at proper angle and in specific azimuth exhibits one-dimensional order determined by step bunches [1] or single steps [2], [3]. The distribution of steps can be controlled by careful annealing of the clean surface or by annealing of the surface with a predeposited metallic layer. If density of foreign atoms is equal to that necessary to produce a specific superstructure on each terrace, then the whole surface can be thermodynamically stabilized and the vicinal surface shows perfect order in a macroscopic sense. Such approach was successfully applied to Si(755) [2] and Si(533) [3] surfaces. The perfectly ordered surface was achieved after the deposition of Au necessary to create a single monoatomic chain of Au atoms on each terrace. Any deviation from that optimal amount of Au caused worsening of the order and created step bunches.

The surface with one-dimensional order is convenient to produce one-dimensional nanostructures. As it was recently shown [4] Pb deposited on the ordered Si(533) at the temperature slightly lower than room temperature formed arrays of exceptionally long and perfectly parallel nanowires. Both, substrate and the nanowires are highly anisotropic, with principal axes parallel and perpendicular to the nanowires.

Reflectance difference spectroscopy (RDS) is a powerful tool for studying anisotropic surfaces [5], [6]. However, up to now, arrays of the nanowires have not been investigated by RDS. As nondestructive and well established this method seems to be very well suited for *in situ* studying of one-dimensional nanostructures structures on semiconducting surfaces.

In this paper we present an experimental investigation by RDS of an anisotropy in the optical reflectance of the well ordered Si(533) surface with metallic nanowires. We show that the observed strong anisotropy of the optical properties originates from metallic structures which grown aligned in one principal axis and form an array of nanowires.

2. Experimental

Samples were prepared and investigated in two UHV chambers. First chamber was equipped with a RHEED apparatus and a gas-flow UHV liquid helium cryostat for cooling samples during film deposition and during optical measurements. The vacuum system was also furnished with a quartz film-thickness monitor. The reflectance difference (RD) technique consists in the measurements of a relative change in the substrate reflectance upon thin film deposition: $\Delta R/R = (R^{Si+Pb} - R^{Si})/R^{Si}$ where R^{Si} and R^{Si+Pb} are the reflectances of a bare substrate and a substrate covered by a Pb deposit, respectively. An angle of incidence of the light beam ϕ was equal to 45°. The apparatus consisted of a quartz halogen lamp, a prism monochromator, a polarizer, a PbS photoconductor for light energies below 1.2 eV and a Si-photodiode above this energy. The temperature of the detectors was stabilized at –10 °C. The light beam was chopped with frequency equal to 17.8 Hz. A lock-in technique was employed to recover the signal. Light either s-polarized or p-polarized, entered the UHV chamber through a fused silica window and was focused on a sample. Convergence of the light beam at the sample was below 0.7°. The specularly reflected beam, after passing the second fused silica window, was focused on the detector. Intensity of the reflected light was recorded during thin film deposition. The rate of Pb deposition was about 1 monoatomic layer (ML) of Pb(111) per minute. Typically about 600 points of data were collected for each sample with the final thickness of about 4 nm. The whole optical set-up was optimized in order to achieve a high stability of the signal and high accuracy of the measurements. The RD data were recorded during the deposition of Pb as a function of thin film thickness d at fixed photon energies $h\nu$ within the range of $h\nu = 0.5$–3 eV. The reflectance measurements were performed for two orientations of the sample in respect to the light incident plane. In the first experiment the Si steps were in the incident plane, *e.g.*, light electric field vector **E** was perpendicular to the Si step edges. In the second experiment the sample was rotated 90 deg and **E** was parallel to the steps.

The second chamber, equipped similarly, instead of optics contained the low temperature STM (type Omicron VT). The base pressure in both UHV systems was 7×10^{-11} mbar. The Si(533) samples with the dimensions of $18\times4\times0.6$ mm^3 for optical studies and with $10\times1.5\times0.4$ mm^3 for STM measurements, were fabricated from high-resistivity Si crystal. The samples were oriented with a standard X-ray diffractometry technique with the accuracy of ±0.05 deg.

The samples were cleaned in UHV by flashing for a few seconds to about 1500 K. A direct resistive heating was used. In order to produce a well ordered Si(533) surface

0.28 ML of Au was deposited onto the Si(533) and annealed for 1 min at about 950 K, and then the temperature was gradually lowered to about 500 K within 3 min. This process was controlled by RHEED and STM.

3. Results and discussion

3.1. Crystal structure of Si(533) substrate and Pb nanowires

Fresh Si(533) surface obtained after mechanical treatment consisting in polishing and subsequent cleaning in the UHV environment was rough and composed of a variety of facets. A perfect order of the Si(533) surface was achieved after technological treatment used previously during RHEED studies of Si(755) [2] and Si(533) crystal [3]. In this process a key factor is to create one-dimensional chains of Au atoms on each Si(111) terrace. For the Si(533) surface the proper amount of Au necessary to induce such order is equal to 0.28 ML of Au, where ML is counted in units equivalent to 1/2 atom density of Si(111) bilayer. Figure 1**a** shows STM topographic image of

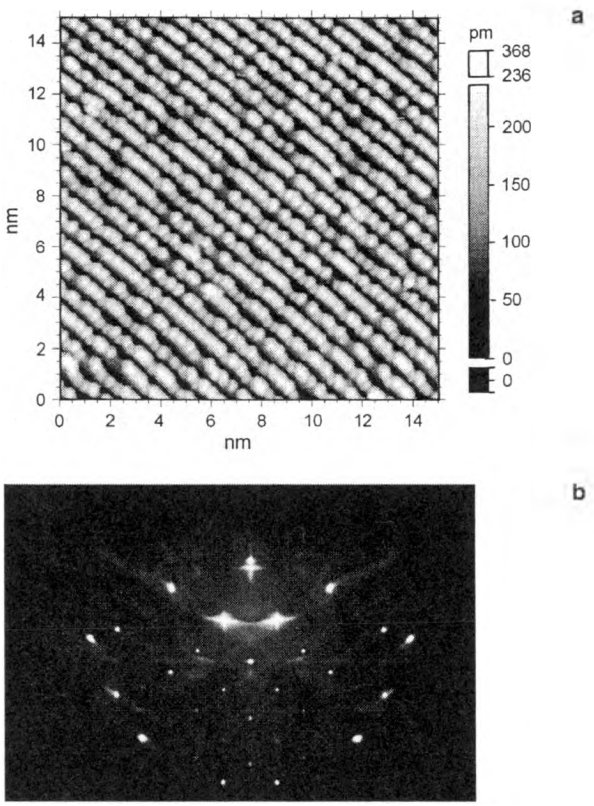


Fig. 1. STM topographic image of Si(533) after the deposition of 0.28 ML of Au and annealing (**a**). Corresponding RHEED pattern with electron beam aligned perpendicular to the terrace edges. All visible bright spots originate from the intersection of the RHEED Ewald sphere with reciprocal lattice of the perfectly ordered Si(533) surface (**b**).

Si(533) after deposition of 0.28 ML of Au and annealing. One-dimensional structures running along [$\bar{1}$10] azimuth are clearly visible. The exact position of Au atoms forming chains is not known but certainly their role in the creation of uniformly distributed parallel features is crucial. Corresponding RHEED pattern with electron beam aligned perpendicular to the terrace edges is shown in Fig. 1**b**. Here all visible bright spots originate from the intersection of the RHEED Ewald sphere with a reciprocal lattice of the perfectly ordered Si(533) surface.

The growth of Pb on the ordered Si(533) varies strongly with the temperature of the substrate. At about 265 K very long and parallel aligned Pb nanowires with a triangular cross-section are growing [4], after reaching a critical Pb thickness of about 1.2 ML of Pb(111). Their length reaches several micrometers. The wires are surrounded by approximately 1 ML of Pb wetting layer on the one-dimensional terraces of Si(533). Figure 2**a** shows STM topographic image of Si(533) surface between large scale Pb nanowires, after the deposition of about 2 ML of Pb. These fine nanowires are not homogeneous in their length but are all aligned parallel to the terraces edges. After nucleation of the long and larger nanowires during further Pb

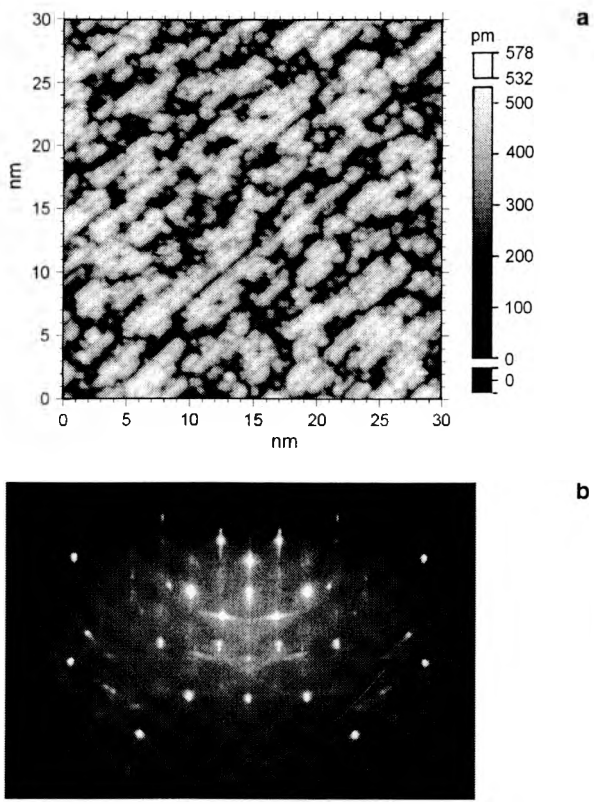


Fig. 2. STM topographic image of Si(533) surface between large scale Pb nanowires after evaporation of about 2 ML of Pb (**a**). In RHEED pattern the larger wires manifest as double spots visible in (**b**). The wetting layer composed of short Pb nanowires contribute here as a fuzzy background.

deposition all incoming excessive Pb atoms are attached to these wires and the thickness of the surrounding wetting layer with small nanowires remains unchanged. In RHEED pattern the larger wires manifest as double spots visible in Fig. 2**b**. The wetting layer composed of short Pb nanowires contribute here as a fuzzy background.

At lower temperatures these long wires are growing more densely. Their length as well as the width and the thickness become smaller and, at the temperature below 120 K, they closely cover the whole substrate. During evaporation of Pb at temperatures higher than room temperature the wires become less elongated, more bulky and, finally resemble islands.

3.2. Reflectance difference

In a classic description a substrate and film immersed in a vacuum are characterized by local and isotropic dielectric functions $\varepsilon^{s}(\omega)$ and $\varepsilon^{f}(\omega)$, respectively. Applying the Fresnel laws derived from the Maxwell equations and continuity arguments, McIntyre and Aspnes [7] showed that when thickness of a film d is small with respect to the wavelength λ of incident light then for s-polarization the RD is

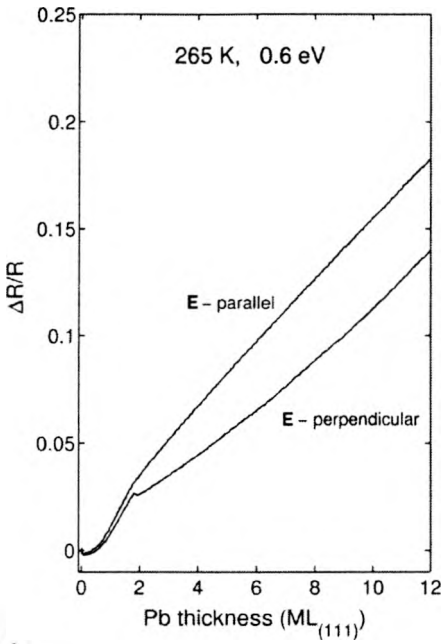
$$\frac{\Delta R}{R} = \frac{R_s^{\mathrm{Si+Pb}} - R_s^{\mathrm{Si}}}{R_s^{\mathrm{Si}}} = \frac{8\pi d}{\lambda}\cos\phi \,\mathrm{Im}\left(\frac{\varepsilon^{\mathrm{Pb}} - \varepsilon^{\mathrm{Si}}}{\varepsilon^{\mathrm{Si}} - 1}\right), \tag{1}$$

with $\varepsilon^{\mathrm{Pb}}$ and $\varepsilon^{\mathrm{Si}}$ being complex dielectric function of Pb overlayer and the substrate, respectively. In our case the expected structural anisotropy of the substrate and of the Pb overlayer leads to anisotropy of the RD data.

Figure 3 shows variation of $\Delta R/R$ of Pb growing on Si(533) at 265 K. The light with $h\nu = 0.6$ eV is s-polarized. The lower curve corresponds to the vector **E** aligned perpendicular to Pb wires, whereas the upper one corresponds to their parallel orientation. Two curves show similar variation up to critical thickness of Pb equal to about 1.8 ML where sudden anisotropy of optical properties occurs. At this critical thickness Pb begins to crystallize [4] and forms on the Si(533) an array of parallel connducting nanowires. Simulataneously, about 1 ML thick wetting layer of Pb remains on the substrate between nanowires.

In order to observe how the formation of Pb nanowires influences the optical anisotropy we have deposited Pb onto Si(533) prepared in the same manner as previously but held at 105 K. The results displays Fig. 4. In this case the knick at about 1.8 ML of Pb is not visible; a fact which indicates that the surface morphology does not change rapidly. Both curves follow almost the same path. Clearly the anisotropy of optical properties is much smaller.

It seems to be reasonable to link optical anisotropy with the formation of metallic nanowires. As it is visible in Fig. 3, the formation of Pb nanowires accompanies the decrease in of the light intensity when vector **E** is perpendicular to the wires, and this process begins when the wires formation starts. This behaviour can be explained as follows. At the critical thickness, when nucleation of nanowires begins, the local density of Pb atoms between nanowires decrease. Large amount of Pb is transferred



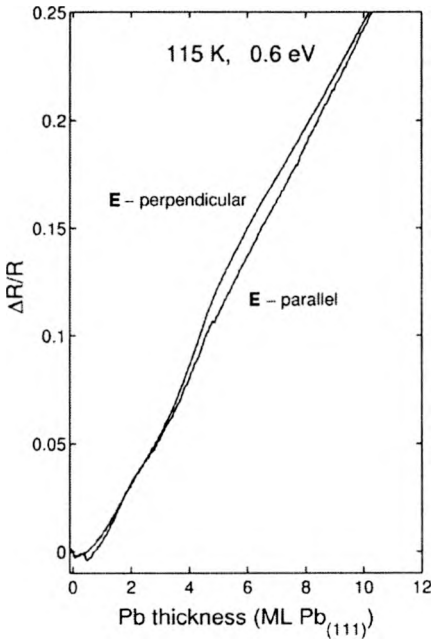


Fig. 3. Variation of $\Delta R/R$ of Pb growing on Si(533) at 265 K. The light with $h\nu = 0.6$ eV is s-polarized. The upper curve is for vector **E** aligned perpendicular to Pb wires, whereas the lower one corresponds to sample orientation with wires oriented parallel to **E**.

Fig. 4. Variation of $\Delta R/R$ of Pb growing on Si(533) at 115 K. The light with $h\nu = 0.6$ eV is s-polarized.

to nanowires. This process reduces the amount of metallic medium interacting with the light electric field **E** directed perpendicular to the wires. For the light with the wavelength λ several times larger than the width of the wire, the wires are too thin to induce noticeable currents perpendicular to their length. As a result, the intensity of reflected light decreases. After rotation of the sample, when vector **E** is parallel to the wire length, the electric field of the light can induce currents along the wires.

This assumption is confirmed by negligible anisotropy observed during deposition of Pb on a cooled substrate, when the formation of nanowires is suppressed. In other experiments, during the deposition of Pb at 265 K, we have observed that the anisotropy for energy of $h\nu$ equal to 3 eV vanishes. This observation suggests that the main factor responsible for the observed optical anisotropy is the intraband Drude absorption, not interband transitions [8].

4. Summary

In summary, we have shown that the formation of parallel aligned nanowires of Pb on the anisotropic Si(533) substrate is responsible for the observed optical anisotropy. The anisotropy appears as a difference in optical reflectance for the s-polarized light with vector **E** parallel and perpendicular to the wires. This large optical anisotropy

originates from surface conductivities which are different for two principal symmetry directions of the Si(533) itself and the surface covered with Pb.

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References

[1] PHANEUF R.J., WILLIAMS E.D., Phys. Rev. B **41** (1990), 2991.
[2] JAŁOCHOWSKI M., STRÓŻAK M., ZDYB R., Surf. Sci. **375** (1997), 203.
[3] ZDYB R., STROZAK M., JAŁOCHOWSKI M., Vacuum **63** (2001), 107.
[4] JAŁOCHOWSKI M., BAUER E., Progr. Surf. Sci. **67** (2001), 79.
[5] BORENSZTEIN Y., MOCHAN W.L., TARRIBA J., BARRERA R.G., TADJEDDINE A., Phys. Rev. Lett. **71** (1993), 2334.
[6] CHANDOLA S., POWER J.R., FARRELL T., WEIGHTMAN P., MCGILP J.F., Appl. Surf. Sci. **123** (1998), 233.
[7] MCINTYRE J.D.E., ASPNES D.E., Surf. Sci. **24** (1971), 417.
[8] LILJENVALL H.G., MATHEWSON A.G., MYERS H.P., Philos. Mag. **22** (1970), 243.

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