TDS Study of Potassium Adsorption on Al(111) and fivefold Al-Pd-Mn

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Comments

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TDS Study of Potassium Adsorption on Al(111) and fivefold Al-Pd-Mn

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ABSTRACT

We have used thermal desorption spectroscopy to carry out a comparative study of potassium adsorption on Al(111) and on the fivefold Al-Pd-Mn surface. Potassium adsorption on the quasicrystal was found to be different than on Al(111). The potassium monolayer desorbed from fivefold Al-Pd-Mn at lower temperatures than from Al(111). Potassium is known to form a dense monolayer on Al(111), with an ideal coverage of 0.33, but for the monolayer on fivefold Al Pd Mn we find that the saturation coverage is only one twelfth.

INTRODUCTION

Alkali metal adsorption has been studied during the last twenty years on a large variety of surfaces, especially metal and semiconductor surfaces¹,². Known to decrease the work function, alkali metals are able to influence the surface electronic structure, even leading to adsorption induced reconstruction of the structurally less-stable surfaces³. Potassium is known as a promotor in the field of catalysis, increasing the sticking coefficient and dissociation of small molecules, catalyzing the oxidation of semiconductors, for example⁴.

There is an ongoing discussion about the surface electronic structure of quasicrystals. A particular issue is whether the surface density of states has a pseudogap at the Fermi level as is the case for the bulk density of states⁵,⁶,⁷ and whether this pseudogap can affect chemical interactions with other materials, such as wetting. For this reason, we chose to study the interaction between a strong electron donor--the alkali metal potassium--and the quasicrystal surface, for which a low electron density is postulated. The Al(111) surface was chosen for comparison because in the topmost layer of the fivefold (5f) Al-Pd-Mn surface, only Al atoms are found, and a surface structure model shows that the lateral density of the two topmost layers taken together is similar to that of the Al(111) surface⁸.

EXPERIMENTAL DETAILS

A single grain Al-Pd-Mn Bridgman sample was oriented by Laue X-ray diffraction along the fivefold axis. The surface was diamond polished down to 1 µm. Inductively coupled plasma atomic emission spectroscopy of an adjacent piece showed the bulk composition to be Al₇₂.₂Pd₁₉.₁Mn₈.₇. A single grain Al sample oriented along the [111]-axis was prepared in the same manner. The sample dimensions were about 7 × 6 × 1.5 mm³. The experiments were carried out in an ultrahigh vacuum chamber with a base pressure of 8 × 10⁻¹¹ Torr. A 5 × 5 mm² sample area was cleaned by sputtering and annealing as described in detail elsewhere⁹.
limits of the annealing temperature were 800 K for the Al-Pd-Mn sample and 750 K for the Al sample. In order to accommodate W/5%Re-W/26%Re thermocouples, holes were drilled into the side of the samples, in the case of the quasicrystalline sample by spark erosion. Temperature measurement inside the sample was necessary due to the dynamic conditions of the thermal desorption experiment. The thermal desorption experiment required relatively fast heating and cooling. Therefore, the samples were mounted with Ta wires on a Ta foil, which was spot welded to Ta rods. The sample holder was cooled by liquid nitrogen and heated resistively. The sample was heated with rates between 1 and 4 K/s. The Al sample could be cooled down to 120 K, whereas the Al-Pd-Mn sample reached only 150 K as its lowest temperature.

The sample temperatures for Al-Pd-Mn cannot be compared directly to temperatures given in the literature, because in most other experiments the temperature has not been measured at the sample, as it was here. As an example of the discrepancies which can result, we found that at a sample temperature of 800 K, the temperature of the Ta foil was 890 K, when measured under static conditions.

Potassium was deposited onto the surface with a commercial SAES getter source which had been sufficiently outgassed before use. For thermal desorption experiments, a quadrupole mass spectrometer (UTI) was used. The ionizer assembly was shielded by a stainless steel mask with a meshed net window \(3 \times 5 \text{ mm}^2\) in size.

**DISCUSSION**

Thermal desorption traces with increasing K exposures were measured. Fig. 1 shows the monolayer saturation, and the beginning of the multilayer growth. The onset temperature for the multilayer desorption was 300 K. The monolayer desorption peak maximum for complete monolayer coverage was 450 K. A small peak at higher temperatures was due to oxygen stabilized K patches, and its position varied with the number of thermal desorption traces that were taken after initial surface cleaning. For a surface that was free of contamination, no K was detectable with Auger electron spectroscopy after heating up to 700 K.
Fig. 2 shows the monolayer desorption peak maximum shifting about 20 K for coverages below monolayer saturation. For monolayer saturation, the peak maximum was 450 K and a shoulder appeared at 465 K. The same asymmetric peak shape has been reported in the literature, although there the maximum at 450 K was much sharper and the shoulder appeared at 475 K\textsuperscript{10}. For the K/Al(111) system, it was reported that the K adatoms condense into islands already at low coverages\textsuperscript{11}. The islands exhibit in general a \((3^{1/2} \times 3^{1/2})R30^\circ\) structure, with adsorbate positions changing with temperature. The K adatoms occupy on-top sites at 90 K and substitutional sites at 300 K\textsuperscript{12}, this phase transformation is irreversible because of the adsorption energy being larger for intermixed K than for surface K. A dense monolayer is formed at a coverage of 0.33\textsuperscript{11}, coverage being defined as the ratio of the number of adsorbate atoms to the number of the atoms in the top layer of the unreconstructed surface.

The thermal desorption trace for a monolayer saturation exposure of K on 5f Al-Pd-Mn is shown in fig. 3. For this measurement the holder was heated with a constant rate and the relation between the sample and holder temperatures was recorded as shown in the insert. This resulted in a nearly constant heating rate of 4 K/s for sample temperatures above 350 K. Multilayer and monolayer desorption peaks were observed, but the corrected temperature scale was not valid in the low temperature range where the multilayer peak occurred. Multilayer desorption is independent of the substrate and the onset temperature is known from the literature to be around 300 K. The low temperature peak did not saturate with increased exposures and was therefore identified as multilayer peak. The monolayer peak at 390 K reached saturation for very low exposures. No K was detectable with Auger electron spectroscopy after heating up to 700 K.

The monolayer on 5f Al-Pd-Mn desorbed at temperatures 60 K lower than the monolayer on Al(111), as shown in fig. 4. Therefore we conclude that K binds more weakly to the 5f Al-Pd-Mn surface and does not intermix as it does in the case of Al(111), where it occupies substitutional sites above room temperature. This could mean that the vacancy formation energy for the quasicrystal surface is significantly higher than for Al(111).

K11.4.3
Because the sputtered areas were the same for both samples the peak areas can be compared quantitatively. For monolayer saturation coverage the monolayer peak area for Al(111) was twelve times the monolayer peak area for 5f Al-Pd-Mn. Therefore we conclude that the density of the K monolayer on 5f Al-Pd-Mn is only one twelfth of than on Al(111). The few atoms on the surface may adsorb at a small number of energetically favorable sites on the 5f Al-Pd-Mn surface.

CONCLUSIONS

We find that potassium binds only weakly to the 5f Al-Pd-Mn surface, and that the density of the first layer is quite low. There is no evidence for the intermixing that occurs on Al(111). The low density of the first layer implies that K adsorption occurs only at a few select sites. Depending on whether these adsorption sites are intrinsic to the quasiperiodic structure or statistically-arranged defect sites, this system might be a candidate for a quasiperiodic adsorbate superstructure. The weak interaction of K with the quasicrystal may reflect the sensitivity of this ionic adsorbate to the low density of states at the surface.

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