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LETTER TO THE EDITOR

Discontinuities in the properties of thin lead films on gold (110) surfaces

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**Abstract.** Simulations show that lead forms a single adlayer on the gold (110) surface until a saturation coverage,  $\theta_c$ , of less than one Pb atom/gold surface atom is reached. Additional atoms at slightly above this saturation coverage initiate a second adlayer. Discontinuities are seen in the energy of the adlayer and its structural properties at the saturation coverage.

Thin metal-on-metal films have been the subject of much experimental research [1, 2]; in part, because they provide an important test of our understanding of how atomic interactions are modified at surfaces and, in part, because of their potential technological applications.

Experimental studies of lead (Pb) on the gold (Au) (110) surface observe that the lead forms a single simple adlayer until a saturation coverage of  $\theta = 6/7 \approx 0.86$  is reached [2]. Here we report simulations of the ground state configurations of Pb on Au(110). The results are consistent with the experimental results in showing that a saturation coverage,  $\theta_c$ , occurs at less than one Pb atom/surface Au atom. In addition, the simulations reported here clearly show that the saturation coverage is associated with discontinuities in the energy of the first adlayer and the atomic spacing.

A simulation technique in which the temperature is gradually reduced in a series of steps [4], based on molecular dynamics, was used to obtain the ground state configurations of Pb on the Au(110) surface and the underlying gold substrate for coverages in the range  $0.5 \geq \theta \geq 1.1$ . The atomic interactions were parametrized using the embedded-atom method (EAM). Following the prescription given by Mei *et al* [3], the energy of atom  $i$  due to the surrounding atoms is assumed to consist of two parts, a two-body interaction  $\phi$  and an embedding term  $F(\rho_i)$  which depends on the local electron density  $\rho_i$

$$E_i = F(\rho_i) + \frac{1}{2} \sum_{j(\neq i)} \phi(r_{ij}). \tag{1}$$

The embedding function and two-body interaction are expressed in terms of a number of adjustable parameters which can be fitted to bulk properties of the solid. In the prescription of Mei *et al* [3] these take the following forms:

$$F(\rho) = -E_c \left[ 1 - \frac{\alpha}{\beta} \ln \left( \frac{\rho}{\rho_e} \right) \right] \left[ \frac{\rho}{\rho_e} \right]^{\frac{\alpha}{\beta}} - \frac{1}{2} \sum_m s_m \phi \left( p_m r_{1e} \left[ 1 - \frac{1}{\beta} \ln \left( \frac{\rho}{\rho_e} \right) \right] \right) \tag{2}$$

$$\phi(r) = \phi_e \left[ 1 + \delta \left( \frac{r}{r_{1e}} - 1 \right) \right] \exp \left[ -\gamma \left( \frac{r}{r_{1e}} - 1 \right) \right]. \tag{3}$$

$E_c$ ,  $r_{1e}$  and  $\phi_e$  are the cohesive energy, equilibrium nearest-neighbour distance and value of the two-body interaction at the equilibrium nearest-neighbour distance respectively.  $s_m$

and  $p_m$  are the number of atoms in the  $m$ th shell of atoms surrounding atom  $i$  and the equilibrium value of the ratio of the distance between atom  $i$  and an atom in the  $m$ th shell and the nearest-neighbour distance, respectively. In the present work only first- and second-neighbour shells were included in the sum. The embedded-atom method and simulated annealing method used in this study have been fully described elsewhere [4, 5].

The Au substrate consisted of seven atomic layers. The upper layer was taken to be the surface layer and periodic boundary conditions were applied on edges parallel to the surface. The lowest two layers were fixed in the bulk equilibrium atomic positions for gold to simulate the effects of bulk layers in a thick substrate. Surface areas between  $10 \times 6$  and  $12 \times 8$  (atomic spacings<sup>2</sup>) in the  $[1\bar{1}0]$  and  $[001]$  directions, respectively, were used. For each value of coverage the system was started with the gold atoms close to the equilibrium positions at initial temperatures of  $T \approx 100$  K. Time steps of 0.625 fs were used and typically cooling took place over a simulated time of 20 ps with the system temperature reduced every 4 ps. Although this procedure is expected to find a local minimum in the energy it is not guaranteed to find a global minimum. To ensure that the global minimum had been found the simulations were repeated three to five times starting from different initial configurations of lead atoms in each case. Data were collected at various temperatures during the cooling process and extrapolated to zero temperature by linear regression.

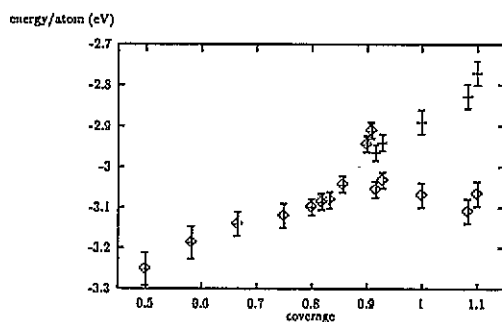


Figure 1. Variation of the energy/atom with coverage for Pb atoms in the whole Pb adsorbate (+) and in the first layer only (◇).

Figure 1 shows the average energy/atom of the adsorbed Pb atoms as a function of coverage. It is not possible to clearly discern any feature that would indicate that the saturation coverage for a single adlayer has been exceeded. However, visual inspection of the configurations reached by the simulations at coverages above  $\approx 0.9$  showed that some of the Pb atoms lay on top of the first adlayer. A plot of the energy/atom for atoms in the first adlayer of lead only, as a function of coverage, exhibits a substantial discontinuity in the energy at  $\theta = 0.91 \pm 0.01$  (figure 1). If the coverage is further increased, up to  $\theta = 1.1$ , the energy/atom of the first adlayer remains essentially constant.

Discontinuities are also observed in structural properties such as the average nearest-neighbour distance between Pb atoms lying in the same trough on the gold surface (figure 2). This distance initially decreases as more atoms enter the first adlayer but increases sharply at  $\theta_c$ . Similarly the average spacing between the gold surface layer and atoms in the first Pb adlayer initially increases with increasing coverage but shows a sharp drop at  $\theta_c$ .

Visual inspection of the configurations reached in the simulations, at each of the coverages considered, makes clear the physical process that causes these discontinuities (figure 3). The Au(110) (unreconstructed) surface is a  $1 \times 1$  surface in a rectangular array with a nearest-neighbour distance of 2.88 Å in the  $[1\bar{1}0]$  direction and a nearest-

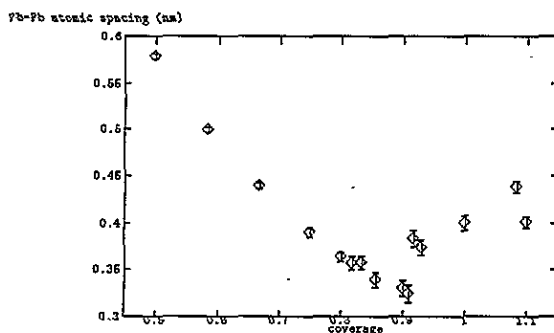


Figure 2. Variation in the average Pb-Pb spacing in the  $[1\bar{1}0]$  direction for atoms in the first adlayer only.

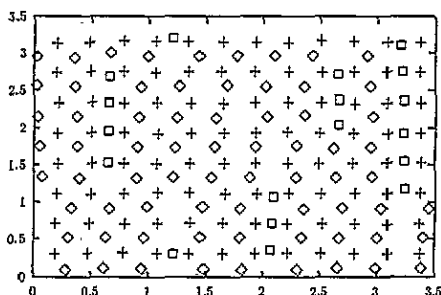
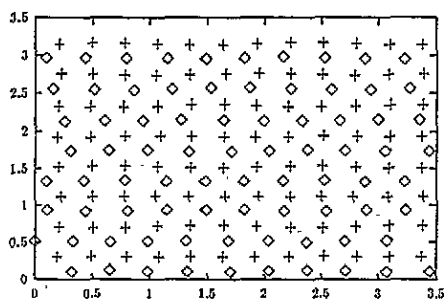


Figure 3. Typical configurations of gold substrate surface atoms (+), first-adlayer lead atoms ( $\diamond$ ) and second-adlayer lead atoms ( $\square$ ) for values of the coverage  $\theta = 0.83 < \theta_c$  (a) and  $\theta = 0.92 > \theta_c$  (b). The scales indicate distances in nm relative to an arbitrary origin in the layer. Note that the second-adlayer atoms tend to be above bridge sites in the gold substrate surface.

neighbour distance of  $4.07 \text{ \AA}$  in the  $[001]$  direction. At the lowest coverages considered the Pb atoms sit in the interstitial sites in the Au surface. As the coverage is increased Pb atoms continue to lie in the troughs between rows of gold atoms in the  $[1\bar{1}0]$  direction. However, Pb atoms do not enter adjacent interstitial sites but rather spread along the trough to maintain the maximum separation. (Perhaps not surprisingly as the interstitial distance is significantly less than the nearest-neighbour Pb-Pb distance of  $3.50 \text{ \AA}$  in bulk lead.) There appears to be no correlation between the Pb atoms in the  $[001]$  direction. As the coverage is further increased the nearest-neighbour distance measured between atoms in the same trough decreases until the saturation coverage is reached. Some buckling of the Pb adlayer is observed as the saturation value is approached. At the saturation coverage

of  $\theta_c = 0.91 \pm 0.01$  some of the Pb atoms move onto the bridge sites between the gold atoms. Where this occurs the bridge atoms, in general, are close to lead atoms in adjacent troughs and these atoms also tend to move out of the first adlayer in order to increase the average distance between Pb atoms. Consequently, as the saturation coverage is exceeded the number of Pb atoms in the first adlayer drops to a value well below its value as the saturation value is approached with the resulting discontinuous change in the energy/atom and nearest-neighbour distance in the first adlayer.

In summary, our simulations have shown that the existence of a saturation coverage for the first adlayer of Pb on the Au(110) surface observed experimentally is associated with discontinuities in the energy/atom of the first adlayer, the nearest-neighbour distance in the layer and the average distance of the layer above the gold surface layer. In particular the number of lead atoms/gold atom in the first adlayer is substantially lower just above the saturation coverage than its value just below the saturation coverage.

The saturation value observed in the simulations  $\theta_c = 0.91 \pm 0.01$  is slightly higher than the value,  $\theta = 6/7 \approx 0.86$ , found experimentally. However, the values reported here are for zero temperature while the experiments appear to have been performed at room temperature. The effect of temperature on the value of the saturation coverage is currently being investigated. Experimentally it is observed that if the coverage is increased sufficiently above one monolayer, a complex series of structures is observed until a AuPb<sub>2</sub> alloy is formed [2]. The higher coverages at which alloying occurs will be studied in future work.

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## References

- [1] Zhang Y and Slavin A J 1992 *J. Vac. Sci. Technol.* **10** 2371
- Ma P and Slavin A J 1993 *J. Vac. Sci. Technol.* **11** 2003
- Jones T L and Venus D 1994 *Surf. Sci.* **302** 126
- [2] Perdureau J, Biberian J-P and Rhead G E 1974 *J. Phys. F: Met. Phys.* **4** 798
- Biberian J-P 1978 *Surf. Sci.* **74** 437
- [3] Mei J, Davenport J W and Fernando G W 1991 *Phys. Rev. B* **43** 4653
- [4] Imeson D 1995 *MSc Thesis* Queen's University
- [5] Imeson D and De'Bell K 1995 unpublished