

Adsorption of superheavy elements on metal surfaces

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The adsorption of radioactive superheavy nuclei in a thermochromatographic column is one possible method to detect these nuclei and to compare their volatility with known elements. The important physical quantity which controls this process is the binding energy. The theoretical prediction of it is very complicated, but we present here a first ab initio calculation of these quantities for the element 112 and for its homologue Hg on an Au(100) surface. Our main task is to calculate the difference in the adsorption energies between these two elements. We performed the calculations of the binding energies using a relativistic molecular program [1] developed in our group, which solves the relativistic Khon-Sham equations for various density functionals. To solve them the molecular orbitals are taken as a linear combination of atomic orbitals (MO-LCAO).

We employ optimized basis sets obtained from dimer calculations with the same program, which include the additional functions $6p$ and $5f$ for Au and Hg, and $7p$ and $6f$ for element 112. The obtained binding energies (Table 1) differ slightly from those given in [2] because we employed a more extended basis set which included additional nf functions. These results can be interpreted in terms of binding when the surface is approximated by only one atom.

System	Binding energy [eV]		Distance [a.u.]
	rLDA	GGA	
HgAu	-1.03	-0.55	4.9
112Au	-0.93	-0.41	5.0

Table 1: Binding energy and bond distance (rLDA) for the HgAu and 112Au dimers

In our calculations for the adsorption we approximate the surface by moderate clusters (with the number of atoms between 9 and 16). Three possible positions (top, hollow and bridge) of the ad-atom on the cluster are considered. The distances between the atoms of the clusters are kept fixed to their bulk values. In the case of top and hollow positions the C_{4v} symmetry was used, while the C_{2v} symmetry was used for the bridge position.

Type of ad-atom	Position of the ad-atom	Binding energy [eV]	Distance [a.u.] to the surface nearest neighbour	
			surface	nearest neighbour
Hg	top	-0.60	5.0	5.0
	bridge	-1.15	4.3	5.1
	hollow	-1.04	3.5	5.2
Element 112	top	-0.67	5.2	5.2
	bridge	-1.08	4.5	5.3
	hollow	-0.99	3.8	5.4

Table 2: Binding energy and bond distances of Hg and element 112 on the Au clusters (rLDA)

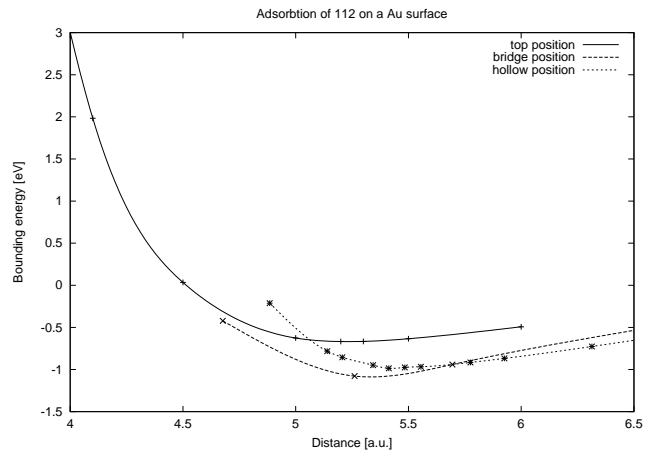


Figure 1: The potential energy curves for the adsorption of element 112 on the Au clusters.

Fig. 1 presents the potential energy curves (rLDA) of element 112 on these three clusters as function of the distance to the nearest neighbours. The values which describe the minima are summarized in Table 2, which also includes the corresponding values for Hg. One can see that for both Hg and element 112 the bridge position on the Au cluster is preferential, with the binding of element 112 being by 0.06 eV weaker than that of Hg.

The values for the top position show the reversed trend from Hg to element 112. This is probably an artifact because the cluster for the top position is too small (only 9 atoms) to lead to good results. Here further calculations with a bigger cluster are necessary. Nevertheless, our experience shows [3] that the bridge position may stay preferential even for larger clusters. Our calculated GGA values for the binding energy of Hg and element 112 on the Au cluster in the bridge position are 0.65 eV and 0.56 eV, respectively. The value for Hg could be compared with the adsorption enthalpy of Hg on the gold surface of -1.01 eV estimated on the basis of experimental data [4]. Results of our calculations suggest that the adsorption enthalpy of element 112 on the Au surface should be about 0.1 eV (~ 10 kJ/mol) smaller than that of Hg.

References

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