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Electron-stimulated desorption of Na atoms from the surface of the intermetallic compound CsAu

© Yu.A. Kuznetsov, M.N. Lapushkin¶

Ioffe Institute,
St. Petersburg, Russia
¶ E-mail: lapushkin@ms.ioffe.ru

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Electron-stimulated desorption of Na atoms from the surface of the 2D intermetallic compound CsAu was studied upon deposition of Na atoms on it. It was shown that up to a coverage of 0.35 monolayers of Na atoms, the electron-stimulated desorption of Na atoms is associated with the excitation of the core levels of the adsorbate Cs 5s and Au 5$p_{3/2}$ and with the excitation of the core level of the adsorbent Na 2p, and at coverage more than 0.4 monolayers of Na, the electron-stimulated desorption of Na atoms is associated only with the excitation of the core levels of the adsorbate Au 5$p_{3/2}$. Two peaks were found in the distribution of the kinetic energies of the desorbed Na atoms, the position and intensity of which depend on the coverage with Na atoms. A model of electron-stimulated desorption of Na atoms associated with the excitation of the core levels of Cs 5s, Au 5$p_{3/2}$ and Na 2p is proposed.

Keywords: electron-stimulated desorption, intermetallic, adsorption, sodium, CsAu.

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1. Introduction

The intermetallic compounds (alloys) are used for long time. Bronze was the first intermetallic alloy known by humanity. Use of intermetallic compounds was caused by the obvious reason: intermetallic compound (alloys) of two and more metals have properties not characteristic for the initial metals. At present time the intermetallic compounds find large application in different branches of science, industry, engineering and medicine [1–5]. Among the intermetallic compounds the compounds of gold with alkali metals stand out. Two of them can form semiconductor compounds (CsAu, RbAu) [6,7]. But during transition to two-dimensional layers the intermetallic compounds Li_xAu_y [8], Na_xAu_y [9] and K_xAu_y [10] also can be semiconductors. Just the possibility of transition from metal into semiconductor causes interest to their study.

In the middle of previous century it was shown [6] that intermetallic compound CsAu is wide-band semiconductor with band gap 2.6 eV. Formation of semiconductor compound CsAu during deposition of Cs atoms on Au was considered in some papers: during deposition on gold film deposited on W [11], on gold film sputtered on surface Ru(100) [12], on surface of massive gold [13,14].

The intermetallic compound Na_xAu_y, like CsAu, is formed at room temperatures with stoichiometry of compounds which depends on the formation conditions [15–17].

In [18] Na adsorption on Au(111) surface was studied by the scanning tunneling microscopy. It was shown that upon coverage with sodium with thickness 0.46 of the monolayer (ML) a poorly ordered hexagonal structure is formed, this is due to interaction between adatoms Na and surface atoms Au, and simultaneous bond weakening between first and second layers of Au. In [15], sodium atoms adsorption on Au(111) was studied in a wide temperature range. In particular, it was shown that at room temperature during deposition of Na atoms to coverages 0.4 ML thick the sodium adsorption on chevron structure of gold surface occurs. Depostion of 0.46 ML sodium results in the transformation of the Au(111) chevron surface structure into a poorly ordered domain structure with isotropically compressed surface layer and hexagonal symmetry. The intermetallic compound $NaAu_2$ is formed during depostion of more then one Na monolayer. In [16] it is shown that at small concentrations as compared to sodium, and pressure below 1 GPa Na_2Au is formed, at pressure in range 1−60 GPa Na_3Au is formed. The intermetallic compound formation on the gold film on tungsten (Au/W), used during study of the electron-stimulated desorption (ESD) of atoms, can differ from the intermetallic compound formation on massive sample of Au, and also depend on gold facet exit to surface.

In [19] effect of sodium adsorption on the gold film 2.7 nm thick, deposited on the tungsten substrate, was studied. Depostion of Na atom on thin film Au on W leads to changes in spectra of valence band and core levels of gold and tungsten. The spectra analysis showed that Na atoms in the surface layer are in neutral state, and diffusion of Na atoms deep into Au film results in formation of intermetallic compound Na_xAu_y. Heating of Au film with formed intermetallic compound Na_xAu_y at temperature of 640 K leads to partial desorption of atoms Na and Au from the surface layer of intermetallic compound Na_xAu_y.

The adsorption of Na atoms on the surface of a predeposited gold film with a deposition dose of more than 3.3 monolayers on the surface of textured W(100) was studied using thermal desorption spectroscopy, and it was shown that Na atoms are desorbed in three phases: from a monolayer Na film, a multilayer sodium film, and from the intermetallic compound Na_xAu_y [20]

$NaAu_2$ is received in melt of gold with sodium at temperature of 1200°C according to Bridgeman method [17]. Also formation of Na_xAu_y is possible at temperature 1000 K during depostion of Na atoms on surface of gold tape [21]. The so produced layer of Na_xAu_y is wide-band semiconductor with a band gap of 2.7 eV [21]. On Na_xAu_y surface different catalyzed reactions take place. [22] showed that during diethylamine adsorption as a result of catalitic reactions on Na_xAu_y surface the products are formed with molecular mass by several times exceeding the molecular mass of diethylamine.

Formation of two-dimensional semiconductor layer Na_xAu_y during deposition of Na atoms on Au surface was studied in [9] by method of electron-stimulated desorption of Na atoms. ESD of atoms at energy of desorption activating atoms below 200 eV is observed only from surface of semiconductors and dielectrics, and from surface of metal it is not observed due to fast energy dissipation [23–26].

Currently the following scheme is suggested to form intermetallic compounds of gold with alkali metals (AM) during Am atoms depostion on surface of 2D-layer of gold at temperatures close to room temperatures, which can differ from the formation of intermetallic compound on surface of massive gold sample. During AM atoms deposition on gold surface at sumonolayer coverages with AM atoms the intermetallic compound Au-AM is not formed. Only at deposition dose of AM atoms exceeding one monolayer the 2D-layer of intermetallic compound Au-AM is formed, at that on surface only monolayer of adsorbed AM atoms presents.

The phase diagram of system gold–sodium is known [27]. At room temperature and normal pressure the existence of intermetallic compounds Na_2Au, NaAu and $NaAu_2$ is shown.

In paper the objective is set to study using electron-stimulated desorption the adsorption of alkali metal atoms on surface of intermetallic compound Au with other alkali metal using example of Na atoms adsorption on CsAu surface.

2. Experiment

2.1. Materials

A target was textured tungsten ribbon with predominant yield to a face (100) surface. At $T = 2000$ K and under pressure of $1 \cdot 10^{-9}$ Torr, the ribbons were preheated for 5 h by passing alternating current for yield to the surface, predominantly, of the face (100) [9], and after that they were cleaned of carbon by annealing at $T = 1800$ K in oxygen under pressure $1 \cdot 10^{-6}$ Torr for 3 h. The ribbon cleaning was finished after oxygen pumping-out by oxygen desorption at $T = 2200$ K for 3 min.

Gold was sputtered onto the ribbon from a straight-channel tungsten tube, which contained pieces of gold foil with purity of 99.99%. The tube was in parallel to the ribbon and it had holes for uniform deposition of gold along the tape.

The source of atoms flow of cesium and sodium was standard tube straight-channel evaporators, they contain thermally decomposable mixture of cesium (or sodium) chromate with zirconium and aluminum. The tubes of the diameter of 3 mm had several length-wise holes for uniform deposition of caesium (or sodium) along the target. Purity of the flow of deposited atoms was monitored by means of a quadrupole mass-spectrometer MC7302.

The target was irradiated with electrons in a stationary mode. An electron emitter was a polycrystalline tungsten filament of the diameter of 0.15 mm, which was located in parallel to the target. The current density of the irradiating electrons did not exceed 10^{-5} A/cm^2 at the electron energy of 100 eV, so the electron irradiation of the target did not cause its noticeable heating.

2.2. Preparation of samples

The following method was used to prepare the studied samples: on clean tungsten tape at room temperature the gold film was sputtered with thickness of 5 monolayers. Then on Au layer at $T = 300$ K Cs atoms were deposited with dose corresponding to deposition of 5 monolayers leading to the formation of intermetallic compound CsAu [11]. Surface layer Cs was removed upon heating to temperature $T = 320$ K for 80 min [11] to obtain clean surface of CsAu. Then on CsAu surface the atoms of sodium were sputtered in amount necessary to measure ESD (the amount varied in different experiments). After the measurements, the tungsten surface was cleaned and prepared again.

2.3. Experimental setup

The studies described in the present paper were carried out in an ultrahigh-vacuum unit „Spectrometer ESD“, and the experimental scheme is shown in Figure 1. During the experiment, the set-up pressure was below $5 \cdot 10^{-10}$ Torr. The experiments were carried out at temperature of $T = 300$ K. The tungsten ribbon with formed layer CsAu and deposition coverage Na was irradiated by electron beam with energy in range 0−200 eV. Power delivered by the electron beam did not changed the ribbon temperature. The concentration of Cs and Na on the surface has been determined by the time of constant-flux deposition, whose intensity was measured by a surface ionization current on ribbon made of refractory metal [28,29]. The desorbing atoms of cesium were ionized using heated tungsten ribbon, and desorbing atoms of sodium were ionized using heated iridium ribbon. The ESD atoms yield magnitude, q, is

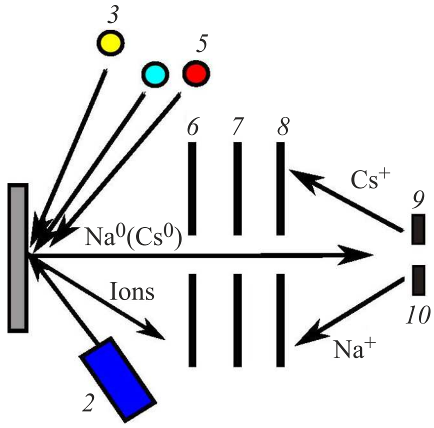

Figure 1. Experiment diagram. *1* — tungsten tape, *2* — electrons source, *3, 4* and *5* — evaporators of Au, Cs and Na, *6, 7* — ion retaining electrodes, desorbing at electron irradiation of sample, *8* — collector of ions, *9* — surface ionization tungsten ribbon, *10* — surface ionization iridium ribbon.

considered to be a density of desorbing atom flux to flux density of the ESD-exciting electrons. „Spectrometer ESD" unit allowed recording the ESD yield of the atoms q, measuring the dependence of this yield q on the energy of the exciting electrons and the number of Eu and Ge atoms deposited on the substrate.

3. Results and discussion

3.1. Results

Figure 2 shows the measured at $T = 300\,\mathrm{K}$ dependence of yield of EDS atoms Cs from surface of 2D-layer of intermetallic compound CsAu on energy of bombarding electrons E_{ex} after removal of the surface layer Cs (see diagram in Figure 3, *b*). In dependence of yield of EDS of Cs atoms we observe one wide resonance peak with maximum at 65.6 eV. The peak position coincides with study results of ESD of system Cs/CsAu provided in paper [8], where the peak is associated with excitation of core level of Au $5p_{3/2}$ (bond energy $E_b = 57.2\,\mathrm{eV}$). No peak corresponding to level excitation Au $5p_{1/2}$ ($E_b = 74.2\,\mathrm{eV}$) is registered. This is due to the fact that when the core level Au $5p_{1/2}$ is excited, the formed hole is neutralized by electron transition from level Cs $5d$ (bond energy of doublet $E_b = 77.5$ and 79.8 eV). Full width at half maximum of peak Γ is 10.8 eV.

After sodium atoms deposition on surface of 2D-layer of intermetallic compound CsAu ESD of Na atoms is observed even at coverage with sodium atoms of less than one monolayer, as this is observed during deposition of alkali metal atoms on gold surface. Figure 2 shows the measured at $T = 300\,\mathrm{K}$ dependence of yield of ESD of Na atoms on energy of bombarding electrons from surface of CsAu layer on surface of which 0.23 ML of sodium is deposited. Corresponding diagram of 2D-layer is given in Figure 3, *c*. In dependence of yield of EDS of Na atoms there are two peaks with maximums at about 42 and 63.3 eV. Poorly resolved peak in the excitation energy region 24–54 eV can be decomposed into two: with maximum at energy $E_{\mathrm{ex\,max}}$ 34.6 and 44.8 eV. Peak with energy 34.6 eV can be associated with excitation of core level Cs $5s$ ($E_b = 22.7\,\mathrm{eV}$), peak with $E_{\mathrm{ex\,max}} = 44.8\,\mathrm{eV}$ can be associated with excitation of core level Na $2p$ ($E_b = 30.8\,\mathrm{eV}$), peak with $E_{\mathrm{ex\,max}} = 63.3\,\mathrm{eV}$ can be associated with excitation of core level Au $5p_{3/2}$ ($E_b = 57.2\,\mathrm{eV}$). Full width at half maximum Γ of peaks with maxima at energy 34.6 and 44.8 eV respectively is 10.4 and 10.6 eV. Note that area under the peak at $E_{\mathrm{ex\,max}} = 44.8\,\mathrm{eV}$ by 1.6 times exceeds the area under peak at $E_{\mathrm{ex\,max}} = 34.6\,\mathrm{eV}$. This may be associated with both the difference in the spectral dependences of the excitation probability of these levels and with the number of atoms of Cs layer in CsAu and adsorbed Na atoms. The area of peak associated with excitation of Au is $5p_{3/2}$, Area of peak associated with excitation of Au $5p_{3/2}$, decreases by 25%, its maximum is shifted towards lower energies to value 63.3 eV, and width significantly decreases (by 3.4 eV) as compared to clean surface of CsAu. We can suppose that additional channel of hole neutralization is formed at level of Au $5p_{3/2}$ due to electron transition from the core level of Na $2s$ ($E_b = 63.5\,\mathrm{eV}$). The various Auger processes occurring in the Na/CsAu interface lead to the electron capturing at the formed local level, which is located below the bottom of the conduction band near it, by the adsorbed sodium atom, as proposed in the ESD model of the system Cs/CsAu [30]. Deposition of one Na monolayer onto CsAu surface with removed top layer leads to that in dependence of yield of ESD of Na atoms on excitation energy only one peak

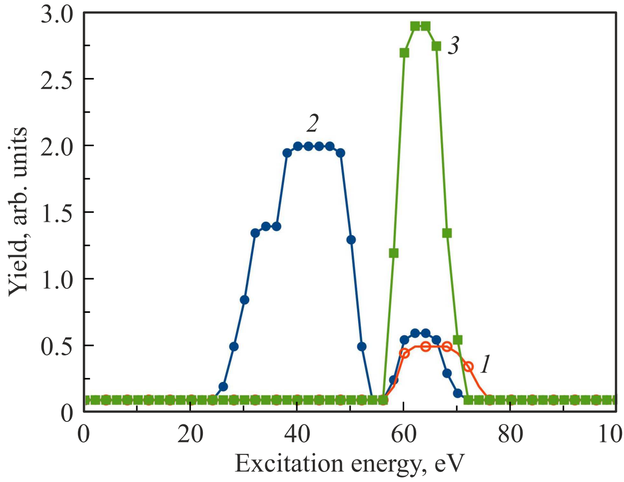

Figure 2. Yield of atoms of Cs (*1*) and Na (*2, 3*) at ESD from CsAu surface at $T = 300\,\mathrm{K}$ depending on energy of bombarding electrons E_{ex}. After CsAu (*1*) heating, after deposition of 0.23 ML of atoms Na (*2*) and after deposition of 0.95 ML atoms of Na (*3*).

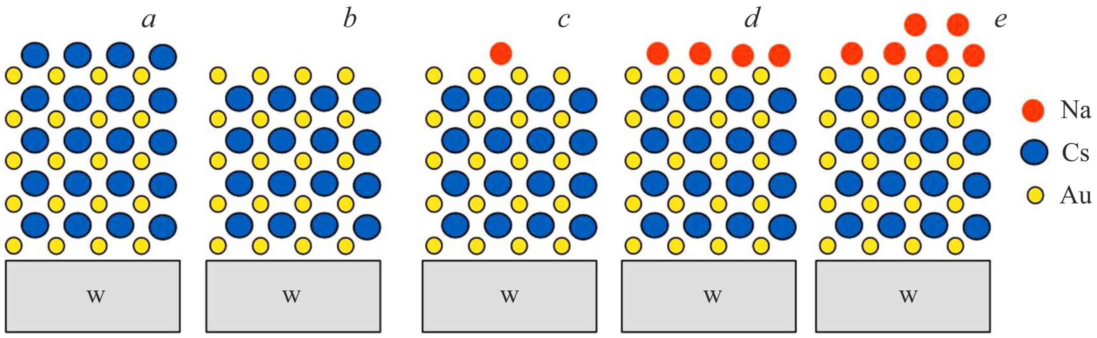

Figure 3. 2D-layer CsAu after formation of layer (*a*), after heating at temperature 320 K for 80 min (*b*), after deposition of 0.25 monolayer of Na atoms (*c*), after deposition of 1.0 monolayer of Na atoms (*d*) and after deposition of over 1 monolayer of Na atoms (*e*).

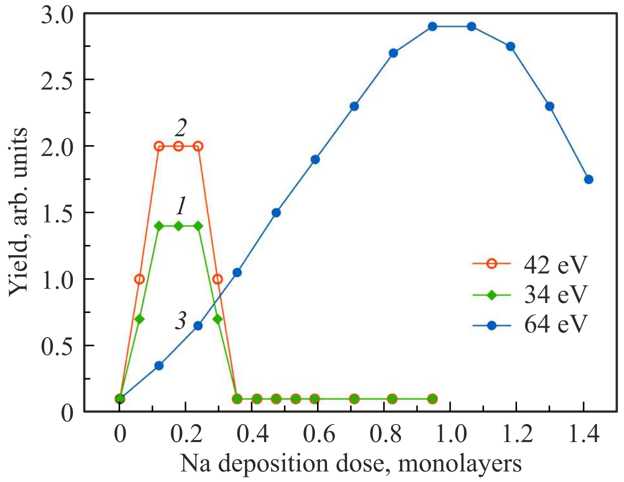

Figure 4. Yield of Na atoms during ESD from CsAu surface at $T = 300$ K vs. dose of sputtered Na: at energies of bombarding electrons 34 (*1*), 42 (*2*) and 64 eV (*3*).

remains, it corresponds to excitation of core level Au $5p_{3/2}$, its intensity increases (Figure 2).

In contrast to the ESD of Na and Cs atoms, when these alkali metals are adsorbed on the Au surface [9,11], in our case ESD of Na atoms is observed at submonolayer coverages Na (Figure 2). This is associated with the fact that adsorption of Na atoms occurs of already formed semiconductor 2D-layer CsAu. During adsorption of atoms Na and Cs on Au surface the adsorbed layer Na (Cs) does not participate in intermetallic compound formation, and, therefore, at such submonolayer coverages ESD of alkali metals is not observed.

Figure 4 shows dependence of yield of ESD of Na atoms from CsAu surface at $T = 300$ K on dose of deposited Na at excitation energy 34, 42 and 64 eV. Energies selection is connected with presence of tree peaks in dependence of yield of ESD of Na atoms, that correspond to excitation of core levels Cs $5s$, Na $2p$, Au $5p_{3/2}$. Let's consider dependence of yield of ESD of sodium atoms in case of excitation of core level Au $5\mathrm{p}_{3/2}$. It is obvious that at excitation energy 64 eV yield of ESD of sodium atoms is observed in full studied range of dose of deposited sodium. As adsorption of sodium atoms occurs to surface of intermetallic compound CsAu, then ESD of sodium atoms is observed already at 0.12 ML.

Increase in dose of deposition of sodium atoms leads to linear increasing of yield of ESD of sodium atoms to dose of deposition of sodium 0.87 ML. After reaching the maximum of yield of ESD of sodium atoms at monolayer coverage yield of ESD decreases. Decrease in yield of ESD of sodium atoms after deposition of monolayer coverage is associated with screening effect of the second layer of sodium atoms, preventing desorption of sodium atoms from below located layer of sodium. Same situation was observed during deposition of Na and Cs atoms on adsorbed monolayer of Na and Cs atoms on surface of 2D-layer of gold with thickness 2 monolayers [9,11]. Completely another picture is observed for dependence of yield of ESD of sodium atoms at excitation energy 34 and 42 eV (these energies correspond to excitation of core levels Cs $5s$ and Na $2p$ respectively). ESD of sodium atoms is observed in very narrow range of doses of deposition of sodium: at dose of deposition 0.3 ML it disappears. Maximum yield of ESD of sodium atoms is reached at dose of deposition 0.15 monolayer. Such behavior of ESD of sodium atoms at excitation energy 34 and 42 eV can be explained by features of formation of submonolayer coverages of sodium. At small coverages the adsorption of sodium atoms occurs, most likely, in pit position above caesium atom and between gold atoms. So, it is possible to excite the core levels Cs $5s$ and Na $2p$ with further neutralization of hole at these levels due to different Auger-processes in Na/CsAu interface.

Figure 5 presents the normalized distributions by kinetic energies E_{kin} of Na atoms during ESD from surface of 2D-layer CsAu with removed top layer Cs after deposition of Na atoms at temperature 300 K for energy of bombarding electrons 42 and 64 eV after deposition of Na atoms. During deposition of 0.2 ML of sodium atoms in spectrum two high-energy peaks are visible: with maxima at energy $E_{\mathrm{kin\,max}} = 0.4$ eV and $E_{\mathrm{kin\,max}} = 0.52$ eV. Generally, low-energy peaks during ESD of alkali metal atoms have maxi-

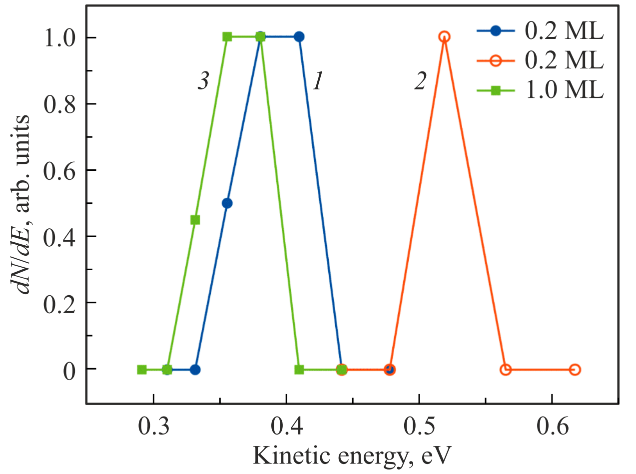

Figure 5. Normalized distributions by kinetic energies E_{kin} of Na atoms at ESD from surface of 2D-layer CsAu with removed top layer Cs after deposition of Na atoms at Na coverages 0.23 (*1, 2*) and 0.95 ML (*3*). Energies of bombarding electrons are 64 (*1, 3*) and 42 eV (*2*).

mum at $E_{\text{kin}} < 0.2$ eV [9,11]. These peaks are associated with excitation of desorption of Na atoms from surface layer of adsorbed Na atoms. As it was stated above, at these excitation energies ESD of Na atoms is associated with excitation of core levels Na $2p$ and Au $5p_{3/2}$. coverage increasing of sodium atoms to monolayer leads to offset of peak maximum at $E_{\text{kin max}} = 0.4$ eV towards lower kinetic energies by 0.06 eV. Peak with $E_{\text{kin max}} = 0.52$ eV is not observed.

The presence of two peaks can be explained by the fact that the adsorption of Na atoms occurs at different adsorption sites on CsAu surface. Peak with $E_{\text{kin max}} = 0.4$ eV is most likely associated with formed 2D-layer of Na atoms. With increasing of Na atoms coverage due to lateral interaction of Na atoms the distance decreases between Na atoms, and the distance increases between submonolayer coverage Na and CsAu surface.

3.2. Model

Figure 6 shows diagram of processes occurred in electronic structure of system Na–CsAu. Electronic irradiation of sample leads to electron excitation from core level Au $5p$ (*1*) to local state E_{L0} near bottom of conduction band E_{CBM}, lowered in field of formed core hole Au $5p^+$ from conduction band into band gap of semiconductor CsAu. The formed hole Au $5p^+$ is neutralized by electrons from level Cs $5s$ (*2*) or from valence band CsAu (*3*). The hole Au $5p^+$ neutralization by transition from level Cs $5p$ is prohibited by selection rules. Further different Auger-processes occur as result of which all formed holes are neutralized, the processes comprise filling of all holes (*4*), with excitation of electrons of valence band (E_{VBM} — top of valence band) in vacuum or into conduction band (*5*) due to realized energy. Hole Au $5p^+$ neutralization returns the local state E_{L0} into conduction band (*6*). Further electrons in the conduction band can be captured by potential field of core Na^+ with formation of neutral atom Na^0 in adsorbed layer Na (*7*). Atom Na^0, formed at that and increased by volume as compared with ion Na^+ is ejected from the adsorbed layer Na on surface CsAu.

Electronic irradiation of sample leads to electron excitation from core level Na $2p$ (*8*) to local state E_{L1} near bottom of conduction band, lowered from conduction band in field of formed core hole Na $2p^+$ into band gap of semiconductor CsAu. The formed hole Na $2p^+$ is neutralized by electrons from the valence band CsAu (*9*) with electron transition from valence band into vacuum (*5*). Hole Na $2p^+$ neutralization due to transition from level Cs $5s$ is unlikely due to large distance between atoms Na and Cs. Hole Na $2p^+$ neutralization returns the local state E_{L1} into conduction band (*10*). Further, electrons in the conduction band can be captured by potential field of core Na^+ with formation of neutral atom Na^0 in adsorbed layer Na (*7*). Atom Na^0, formed at that and increased by volume (as compared with ion Na^+) is ejected from the adsorbed layer Na on surface CsAu.

Same processes lead to desorption of atoms Cs (they are not shown in diagram). In this case the electron in conduction band is captured by the potential field of core Cs^+ with formation of the neutral atom Cs^0 in 2D-layer

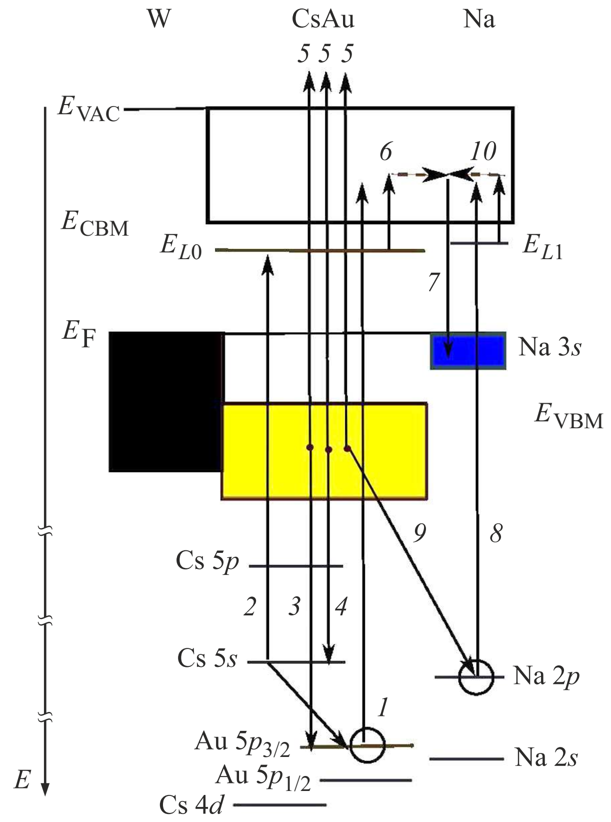

Figure 6. Diagram of processes of electron-stimulated desorption in Na/CsAu interface on tungsten substrate.

CsAu. Atom Cs^0, formed at that and increased by volume (as compared with ion Cs^+) is ejected from 2D-layer CsAu.

Note that all processes occur in Na/CsAu interface.

4. Conclusion

For the first time the electron-stimulated desorption of alkali metal atoms from surface of intermetallic compound of gold with another alkali metal is studied on the example of adsorption system Na/CsAu. the model of ESD of Na atoms is proposed, illustrating the electron transitions in the process of ESD of sodium atoms.

ESD process covers excitation of core levels of adsorbent CsAu (Cs $5s$ and Au $5p_{3/2}$), and excitation of core levels in layer of adsorbed sodium (Na $2p$). ESD of Na atoms upon excitation of level Au $5p_{3/2}$ is observed in full range of doses of deposition of sodium. Excitation of core levels Cs $5s$ and Na $2p$ during ESD of Na atoms is observed only at small doses of deposition of Na, this is associated with decrease in distance between adatoms Na and neutralization of excited states due to states in adsorbed layer. Decrease in yield of ESD of Na atoms at large doses of deposition of Na is due to that second layer of Na atoms prevents desorption of Na atoms from Na/CsAu interface. Distribution by kinetic energies shows two peaks associated with desorption of Na atoms from Na layer in Na/CsAu interface.

Conflict of interest

The authors declare that they have no conflict of interest.

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