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# Properties of ultrathin Pb layers on the Ni(111) face

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The atomic structure and morphology of ultrathin Pb layers deposited on the Ni(111) face in ultrahigh vacuum at the substrate temperature ranging from 150 to 910 K were investigated with the use of Auger electron spectroscopy (AES), low-energy electron diffraction, and directional elastic peak electron spectroscopy (DEPES). The analysis of AES measurements indicates that the two-dimensional growth of the first Pb monolayer takes place for the substrate temperature $180\ \mathrm{K} < T < 480\ \mathrm{K}$. For $180\ \mathrm{K} < T < 270\ \mathrm{K}$, flat two- and three-atomic layer islands (''wedding cakes'') seem to grow after the completion of the first lead monolayer. Then, the three-dimensional growth starts. DEPES measurements indicate that for layers deposited at $T = 150$ K a twin interface $abcBACBAC$ is formed between the Ni substrate and the Pb layer.

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## I. INTRODUCTION

Ultrathin lead layers deposited on the Ni(111) face were investigated in a number of works.[1–5] Those investigations were limited to lead layers deposited at room temperature and were mainly concentrated on the first and, to some extent, on the second lead layer. In low-energy electron diffraction (LEED) patterns, $(1\times1)$, $(3\times3)$, and $(4\times4)$ structures appeared in succession during the increase of coverage with lead up to the full monolayer (about $1.1\times10^{15}$ Pb atoms/cm$^2$). The presence of these single overlayer structures was confirmed[3] by impact-collision ion scattering spectroscopy (ICISS) results. The annealing of a lead layer at 873 K led to a stable $p(\sqrt{3}\times\sqrt{3})R30°$ structure ascribed, on the basis of ICISS (Ref. 4) and medium-energy ion scattering (MEIS) (Ref. 5) measurements, to the formation of a surface (confined to the first atomic layer) Ni-Pb alloy. Results of Auger electron spectroscopy (AES) measurements presented in Refs. 1, 3 were interpreted as evidence of the Stranski-Krastanov growth mode.

In the present paper, we show the results of investigating the atomic structure and morphology of ultrathin Pb layers deposited on the Ni(111) face in ultrahigh vacuum at the substrate temperature ranging from 150 to 910 K. These results were obtained with the use of LEED, AES, and directional elastic peak electron spectroscopy (DEPES). Good quality LEED patterns corresponding to all the structures reported in literature were observed. Owing to the method used for the recording of AES data, the dependencies of AES signals, for both the substrate and adsorbate, on the adsorbate deposition time [we will call these dependencies AES($t$) plots] are represented by as many as 40 to 60 experimental points for every monolayer of the adsorbate. Thus, the shapes of particular AES($t$) plots could be determined more precisely than in the case of such AES($t$) plots presented in Refs. 1,3, where one monolayer is represented only by five or ten points. It will be shown below that such a precise determination of the AES($t$) plots shape gives new important information about the growth mode. In addition, in our measurements the adsorbate deposition is not interrupted for the recording of Auger peaks. Thus, our AES($t$) plots concern a continuous growth of the deposited layer, and it is simpler to describe this process theoretically than the step-by-step deposition commonly used. The extension of our measurements over a wide temperature range resulted in finding out a strong dependence of the growth mode of the succeeding lead layers on the substrate temperature which, to the best of our knowledge, has not been reported in the literature for the Pb/Ni(111) system. DEPES results indicate that a twin interface is formed between the Ni substrate and Pb layer which can be described as an $abcBACBAC$ sequence.

## II. EXPERIMENT

The measurements were carried out in a metal ultrahigh-vacuum chamber with the base pressure of $2\times10^{-8}$ Pa. The chamber was equipped with the reverse view LEED optics (OCI Vacuum Microengineering) which was used for both LEED and AES measurements. The Ni(111) crystal was mounted on a manipulator with facilities for its rotation around axes parallel and perpendicular to its surface. The sample could be heated to 950 K and cooled to 150 K. The crystal temperature was measured by using an NiCr-Ni thermocouple. The crystal was cleaned by potassium ion bombardment from a zeolite source[6] (5 $\mu$A, 660 eV) and by simultaneous annealing at 900 K until the carbon and sulphur peaks became invisible in AES spectrum and a LEED pattern of the clean Ni(111) face with sharp spots and low background was obtained.[7] This method of cleaning was successfully used in our laboratory for other systems, e.g., Ag/Ni(111),[8] Co/Cu(111),[9] and AuCu alloys.[10] It should be pointed out that in investigation of the adsorption of potassium on nickel surfaces[11,12] potassium was found to be removed completely during annealing at 900 and 1000 K. Lead (99.999%) was evaporated onto the crystal surface from a quartz crucible. In directional elastic peak electron spectroscopy,[13] the dependence of the elastic peak intensity in the secondary electron spectrum on the incidence angle of the primary electron beam (DEPES profile) is measured. The intensity maxima appear when the primary beam is parallel to one of densely packed rows of atoms in the sample under investigation because of forward focusing and channeling of primary electrons along such rows and atomic planes parallel to the given row, respectively. Namely, these two mechanisms result in the concentration of primary electrons on

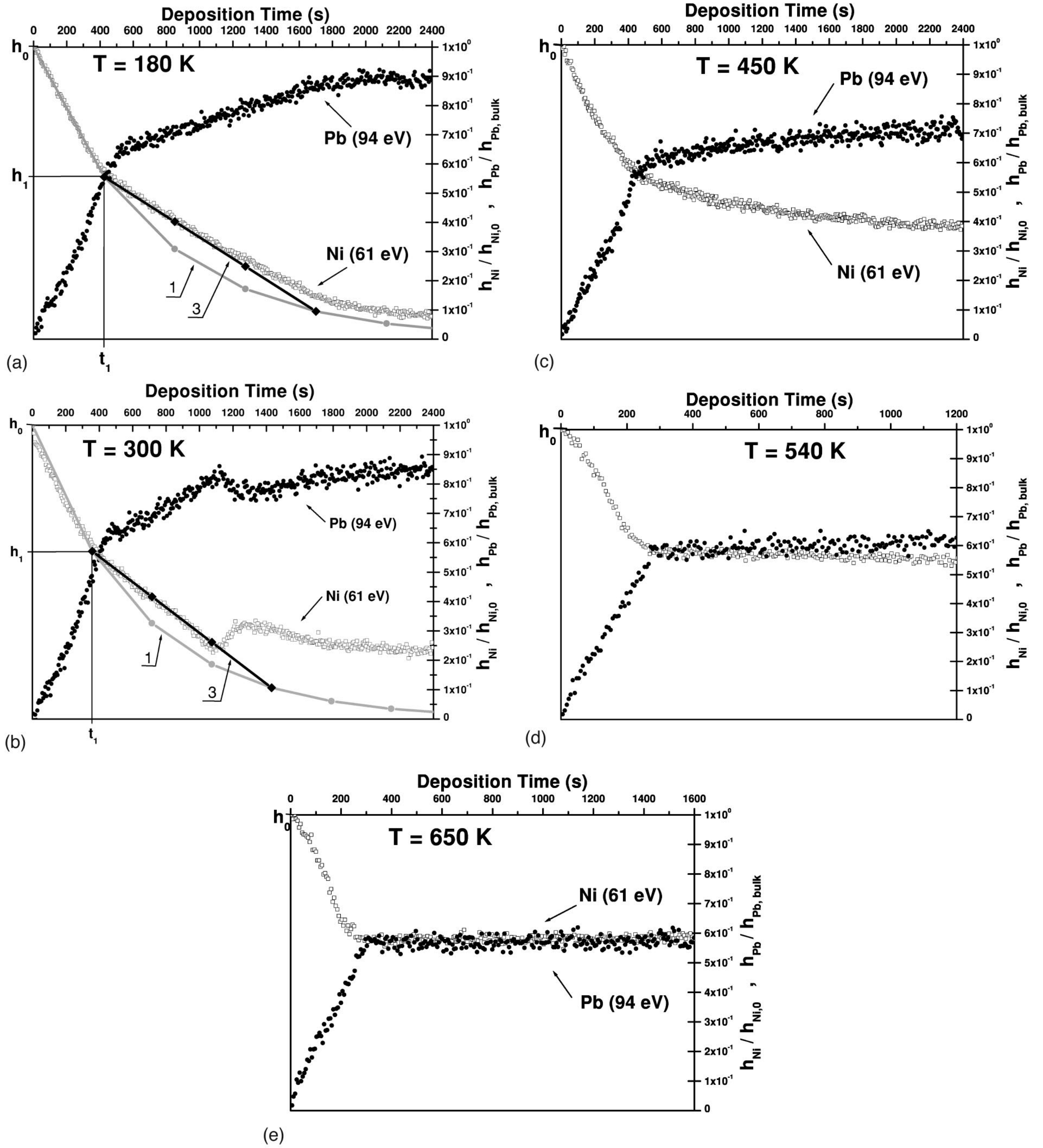

FIG. 1. AES($t$) plots of Ni MVV and Pb NVV peak heights for lead deposition on Ni(111) face at (a) $T=180$ K, (b) $T=300$ K, (c) $T=450$ K, (d) $T=540$ K, (e) $T=650$ K. (1) AES($t$) plot calculated for the Frank–van der Merwe growth, (3) AES($t$) plot calculated for the growth of three-atomic layer thickness flat islands on the first lead layer.

cores of atoms at the subsurface atomic layers which increases the cross section for elastic backscattering of those electrons. For the lead crystal with the (111) face one can expect that three or four atomic layers are responsible for the creation of maxima in profiles B presented in Figs. 2, 3. More information about the mechanism of the creation of these maxima can be found in reviews.[14,15] Positions of such maxima remain unchanged when the electron energy changes from 1000 to 2000 eV. In addition to such maxima, maxima connected with multiple elastic scattering of primary electrons appear in DEPES profiles for some electron energies. These maxima appear, shift, and disappear when the electron energy changes. Only the maxima present at a permanent position for the whole electron energy range (“permanent” maxima) are taken into account in DEPES analysis. For the (111) face of fcc crystals and for the primary beam lying in the plane determined by the $[1\bar{2}1]$-$[\bar{1}2\bar{1}]$ azimuth, permanent maxima are expected to appear simultaneously for [101], [121], and [010] directions (for the primary electron incidence angle $\alpha \cong -35°, 19.5°$, and 55°, respectively).

Owing to the geometry of the system it was possible, during the deposition of lead on the sample surface, either to record the Auger peak heights for the nickel MVV transition at 61 eV and the lead NVV transition at 94 eV under computer control or to record LEED patterns in the computer memory with the use of a CCD camera. The Auger peak heights for nickel and lead were corrected for the background created by the secondary electrons in the low-energy part of the $dN/dE$ versus electron energy curve. To obtain DEPES polar profiles, the sample was rotated around the axis perpendicular to the sample surface to the position (determined with the use of LEED patterns) where the incidence plane of the primary electron beam was parallel to the $[1\bar{2}1]$-$[\bar{1}2\bar{1}]$ azimuth on the sample surface. Next, the sample was steadily rotated around the axis parallel to its surface and the elastic peak height in the $N(E)$ mode was measured, under computer control, at regular intervals, which gives the dependence of the measured height on the rotation angle. Unfortunately, because of the incidence system limitation, the error in determining the incidence angle was as large as 2°.

## III. RESULTS AND DISCUSSION

### A. AES

For substrate temperatures ranging from 180 to 380 K, two linear parts of the AES($t$) plots for 61 eV nickel and 94 eV lead peaks can be distinguished [see Figs. 1(a), 1(b)]. For all 13 AES($t$) plots recorded in this temperature range (only two of them are presented in Fig. 1) straight lines corresponding to the first and second linear parts of the substrate Auger AES($t$) plot (these lines have been obtained by the least-squares method) intersect one another at point $h_1$ corresponding to the value $h_{\mathrm{Ni}}/h_{\mathrm{Ni,\,0}}=0.56$–$0.62$ on the vertical axis and to $t_1=340$–$600$ sec on the horizontal axis for particular AES($t$) plots. The error $\Delta h_1$ of the value $h_1$ determination was estimated for particular AES($t$) plots with the use of errors of coefficients in equations obtained for both linear parts. A typical value of $\Delta h_1$ was equal to about $\pm 0.02$. Thus, the scatter of the experimentally found values of $h_1$ may be only the result of the scatter of experimental points in measured AES($t$) plots. The values of $t_1$ differ remarkably for AES($t$) plots measured at different substrate temperatures. Moreover, for particular AES($t$) plots obtained at the same substrate temperature this value is rather badly reproducible. This indicates that the lead source stability was not satisfactory. Lead was melted in the crucible before every deposition. For technical reasons, the axis of the crucible was almost horizontal and the shape of the melted lead surface could be different after successive meltings. On the other hand, one can expect that this shape remained unchanged during recording of particular AES($t$) plots. Because the scatter of $h_1$ values is not large, we will suppose further that the first linear part corresponds to the formation of the first layer (1 ML) of lead, in spite of large scatter of $t_1$ values. This supposition agrees with the literature data.[1,3] For $T<180$ K the $h_1$ point cannot be found so clearly. In some AES($t$) plots for $T=150$ K three linear segments appeared and the first break was obtained for $h_{\mathrm{Ni}}/h_{\mathrm{Ni,0}}=0.63$ at $t_1=260$ sec. However, results for $T=150$ K were strongly irreproducible and AES($t$) plots similar to those obtained for higher temperatures were found as well. For $T>180$ K, the presence of the second linear part indicates that two-dimensional growth is continued after the first layer formation. However, this is not the Frank–van der Merwe (FM) growth. Namely, the substrate AES($t$) plot calculated for FM growth under the supposition that at $h_1$ point the first lead layer is completed do not fit the experimentally found AES($t$) plots [see curves 1 in Figs. 1(a), 1(b)]. A much better fit is obtained for AES($t$) plot calculated for the growth of flat islands (''wedding cakes'') composed of three atomic layers of lead [curves 3 in Figs. 1(a), 1(b)]. It should be added here that in a number of AES($t$) plots (not shown in Fig. 1) the experimental points fit the AES($t$) plot calculated for the growth of two-atomic layer islands or lie between AES($t$) plot calculated for the growth of three- and two-atomic layer islands. Thus, the growth mode seems to depend strongly on the conditions of lead deposition (rate of deposition, temperature, and morphology of the substrate). Of course, the wedding cake growth should be confirmed with the use, for example, the scanning tunnelling microscopy. Nevertheless, the FM growth, as well as the Stranski-Krastanov growth, seem to be excluded for 180 K$<T<$380 K. For $T<270$ K the substrate AES($t$) plots progressively decreases its slope after completion of the second linear part which corresponds to the start of three-dimensional (3D) growth. On the other hand, for 270$<T<$380 K, after the second linear part a clearly visible maximum appears [see Fig. 1(b)] and only after this maximum the part indicating 3D growth can be seen. The maximum can indicate a reconstruction of the wedding cake islands in which they become slimmer and higher. These results contradict the literature data[1,3] where the Stranski-Krastanov growth at room temperature was reported. In addition, the maximum mentioned above does not appear in the AES($t$) plots presented in Refs. 1,3. Namely, it is stated in Ref. 1 that ''After the monolayer break unsystematic variations in signal amplitudes always occured which is an indication of spontaneous clustering,'' but this statement concerns the clustering of the first lead layer while in our case the maximum appears after deposition of 3 ML of lead. It should be added that in[3] no traces of maximum or unsystematic variations are visible in the presented AES($t$) plots. It cannot be excluded that those discrepancies are connected with the continuous deposition of lead, without interruptions for recording of Auger signals, in our measurements. For deposition temperatures 410 K$<T<$480 K only the first linear segment corresponding to the first monolayer formation exists. After this, the three-dimensional growth AES($t$) plots are not linear (its slope increases with increasing deposition time) while the first segment of adsorbate AES($t$) plots remain linear [see Fig. 1(d)]. This nonlinear shape of the substrate AES($t$) plots and, simultaneously, the linear shape of the adsorbate AES($t$) plots have been reproducibly found in our experiments but we have no explanation for them. For 580 K$<T<$730 K the AES($t$) plots saturate after the first lead layer completion [see Fig. 1(e)]. For $T>730$ K the equilibrium coverage with

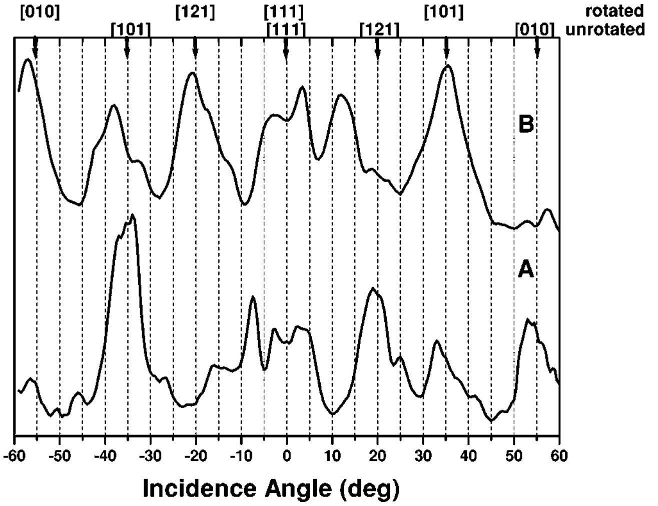

FIG. 2. DEPES polar profiles for the $[1\bar{2}1]$-$[\bar{1}2\bar{1}]$ azimuth of the Ni(111) sample, for electron energy of 1100 eV. (A) Clean Ni(111) face at $T=300$ K, (B) Ni(111) face covered with 10 ML at $T=150$ K.

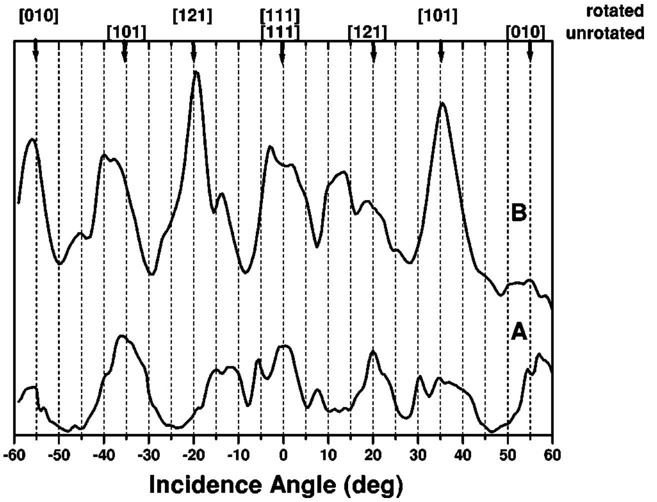

FIG. 3. DEPES polar profiles for the $[1\bar{2}1]$-$[\bar{1}2\bar{1}]$ azimuth of the Ni(111) sample, for electron energy of 1600 eV. (A) Clean Ni(111) face at $T=300$ K, (B) Ni(111) face covered with 10 ML at $T=150$ K.

lead becomes smaller than 1 ML and decreases with increasing temperature. This can be explained by the thermal desorption of the second lead layer for $T<730$ K and the first layer for $T>730$ K.

### B. DEPES

DEPES profiles for the clean Ni(111) face and for 10 ML of lead deposited on this face at $T=150$ K are shown as curves A and B, respectively, in Fig. 2 for electron energy $E=1100$ eV and in Fig. 3 for $E=1600$ eV. In curves A permanent maxima are clearly visible close to angles $-35°$, 19.5°, and 55°, where they are expected for the (111) face of fcc crystals. On the other hand, in profiles recorded for lead layers (in curves B) new permanent maxima are visible close to angles $-55°$, $-19.5°$, and 35° while the maxima present in curves A are practically invisible. This indicates that the lead layer is rotated by 60° with respect to the Ni(111) substrate. Such a rotation is a result of the formation of a twin interface which can be described as an *abcBACBAC* sequence.

## IV. CONCLUSIONS

(1) In accordance with the literature data, two dimensional growth of the first lead layer on the Ni(111) face is observed for $180\ \mathrm{K}<T<480$ K.

(2) For $180\ \mathrm{K}<T<380$ K, the shape of AES($t$) plots indicates the growth of flat two- and three-atomic lead islands on the first lead layer, instead of the Stranski-Krastanov growth reported in the literature. For $270\ \mathrm{K}<T<380$ K spontaneous reconstruction is suggested to take place in the layer containing the number of Pb atoms equivalent to 3–4 monolayers.

(3) For $T>480$ K, the nonlinear shape of the substrate AES($t$) plots is observed for coverages up to 1 ML while the adsorbate AES($t$) plots is linear.

(4) The DEPES results indicate that the twin interface *abcBACBAC* is formed in the Ni-Pb interface region.

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