Tuning Two-Dimensional Phase Formation Through Epitaxial Strain and Growth Conditions: Silica and Silicate on Ni_xPd_{1-x}(111) Alloy Substrates

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Supplementary Materials



Figure S1. Auger electron spectra taken during each step of preparation sequence: (a) After depositing 2 MLE SiO on Ni_{0.48}Pd_{0.52}(111) in UHV; (b) after annealing at 950 K in 4×10^{-8} Torr oxygen for 5 min; (c) after additional annealing at 950 K in 4×10^{-8} Torr oxygen for 20 min; (d) after additional annealing at 1000 K in 4×10^{-8} Torr oxygen for 10 min. No normalization was applied to the curves.

The influence of annealing in oxygen-deficient environments on the surface stoichiometry was systematically studied with AES. Figure S1 shows the Auger electron spectra taken during each step of the preparation sequence. No normalization was applied to the curves and all of the spectra were taken under the same conditions. Thus, the intensity of peaks can be used to track surface composition changes. After depositing 2 MLE SiO on Ni_{0.48}Pd_{0.52}(111) in UHV, both Si⁴⁺ and lower oxidation states were detected by AES. As a reference, the LVV peaks of Si²⁺ and Si⁴⁺ locate at 84 eV and 78 eV.^{1, 2} After annealing at 950

K in 4×10^{-8} Torr oxygen for 5 min, the Si peak was fully oxidized to Si⁴⁺ and the existence of 2D VDW silica and Ni silicate were detected by LEED, STM and RAIRS. The intensity of the O peak increased due to oxidation. Additional annealing at 950 K in 4×10^{-8} Torr oxygen for 20 min lead to the decomposition of 2D VDW silica and formation of Ni silicate based on the RAIRS data; however, no loss of Si could be detected. The increase of the O peak may due to Ni oxidation to form Ni silicate which partially covers the surface. The decrease of the Pd peak and increase of Ni peak after the first two annealing steps was due to Ni surface enrichment during the annealing in oxygen environment, which has been discussed in a previous paper.³ Further annealing at 1000 K in 4×10^{-8} Torr oxygen for 10 min did not change the structure of the 2D overlayer or the intensities of the Si, O, Pd and Ni AES peaks.

- 1. B. Carrière and J. P. Deville, *Surf. Sci.*, 1979, **80**, 278-286.
- 2. T. Schroeder, M. Adelt, B. Richter, M. Naschitzki, M. Bäumer and H.-J. Freund, *Microelectron. Reliab.*, 2000, **40**, 841-844.
- 3. C. Zhou, X. Liang, G. S. Hutchings, Z. S. Fishman, J.-H. Jhang, M. Li, U. D. Schwarz, S. Ismail-Beigi and E. I. Altman, *Chem. Mater.*, 2019, **31**, 851-861.