Correlative TEM and XRD study of the role of Au on the solid state dewetting behavior of Au/Ni bilayers on α-Al_2O_3

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In this study we investigated the influence of Au on the structure and properties of binary Au-Ni nanoparticles (NPs) equilibrated on (0001) oriented sapphire single crystals. The NPs were fabricated by solid-state dewetting of ~40nm thick Au/Ni bilayers above the miscibility gap and subsequent quenching of the specimen by rapid thermal annealing. Since solid-state dewetting is of high technical importance in the field of microelectronics, in-depth studies of similar sample systems were conducted recently. This includes investigations regarding the orientational relationship between dewetted Al films on sapphire [1], the interface reconstruction at equilibrated Ni-sapphire interfaces [2] and the dewetting behavior and morphology of Au/Ni bilayers containing 39% of Au [3]. With this respect, our work contributes by giving insight into the role of Au on the structural as well as mechanical properties of Au-Ni NPs by a systematic variation of the Au content. Here, our methodological approach is to combine TEM and EDXS with XRD in order to obtain complementary information about the morphology, orientation and in-plane as well as out-of-plane strain of the NPs with respect to the sapphire substrate. Furthermore, the impact of alloying and interface structure on the mechanical properties of the NPs was tested by *in situ* SEM compression experiments.

The XRD data for three different Au/Ni compositions is displayed in Figure 1. From the θ/θ-diffractograms a dominant < 111 > texture of the NPs is revealed. In addition, an enhanced Au content manifests in a linear shift of the {111} Bragg Peak to smaller scattering angles, thus in a linear lattice expansion. However, the slope is larger than predicted by Vegard's law. Pole figures for the < 111 > Bragg Peak of the NPs reveal sharp intensity maxima which indicate an epitaxial orientation relationship with $<11\bar{2}0>$ NP || $<11\bar{2}0>$ α-Al_2O_3 in-plane orientation associated with two twin variants. For pure Ni NPs and for low Au concentrations the two twin variants occur with equal probability as can be seen from the sixfold symmetry of the pole figures. In contrast, for the highest Au concentration of 28 at% one twin variant dominates resulting in a threefold symmetry.

For a more detailed investigation of the NP-substrate interface, cross sectional TEM lamellae were prepared by FIB machining. A bright field TEM image, SAED pattern, and HRTEM image of the substrate-NP interface are shown for 28 at% Au in Figure 2. SAED and HRTEM confirm the orientation relationship obtained from XRD and reveal that the interface between the particle and the α-Al_2O_3 substrate is incoherent. EDXS mappings confirm that Au and Ni are uniformly distributed across the particle as expected for a solid solution. Slight segregation of Au towards the NP surface occurs as revealed by the EDXS concentration line profiles. Current investigations by complementary XRD and TEM techniques focus on the presence of strain in the NPs resulting from the epitaxial relationship and quenching from the dewetting temperature.

Finally, the mechanical properties of the NPs were tested with an *in situ* SEM manipulation device [4]. The evaluation of the force-displacement data shows a clear solid solution hardening effect of the alloyed NPs with increasing Au content.

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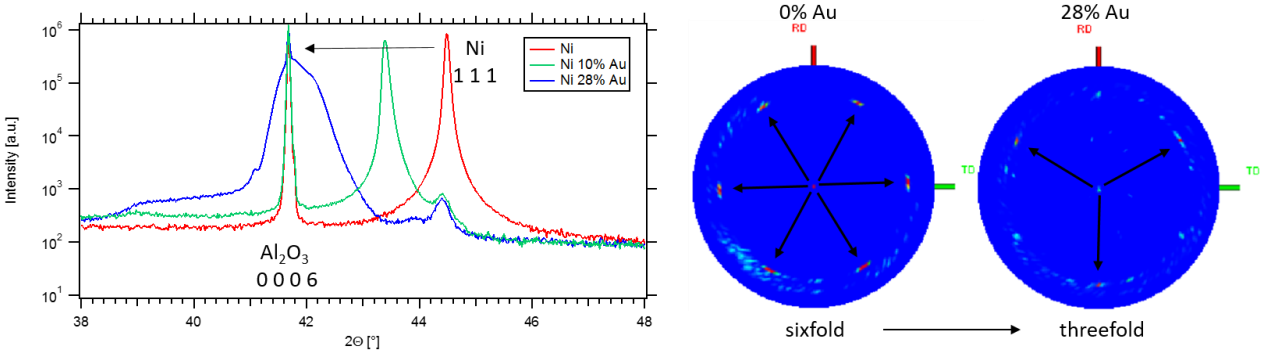

Figure 1 Left: θ/θ-diffractogram of pure Ni NPs and a Au content of 10 and 28 at%, respectively. Right: Pole figure of the 1 1 1 Ni Bragg-Peak for the specimen containing 0 at% and 28 at% Au in a logarithmic scale, respectively.

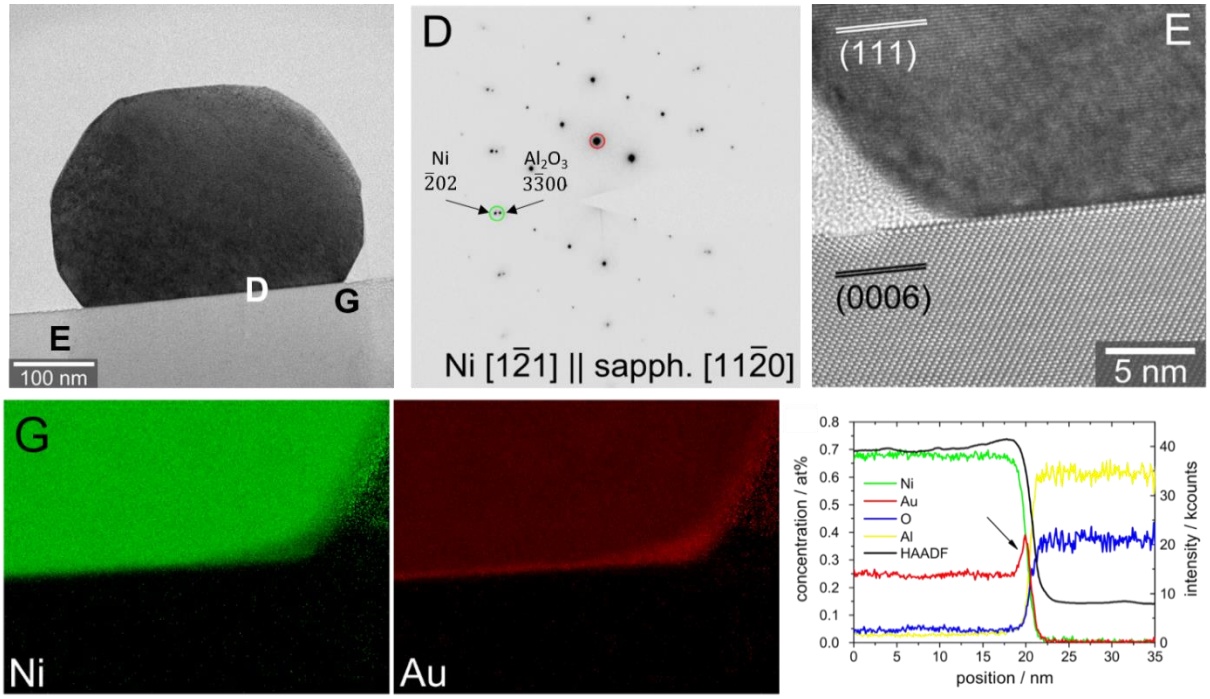

Figure 2 Top left: TEM bright field image of an Ni-28at%Au NP cross section prepared with focussed ion beam in <121> viewing direction (left). Selected area electron diffraction patterns of the position marked in the image (top middle) and high resolution TEM image of the particle substrate interface (top right). Bottom: Energy dispersive x-ray spectroscopy images of the Ni and Au content and the extracted line profiles across the interface, respectively.

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