

Diffusion-induced recrystallization in nickel/palladium multilayers

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Received 30 August 2010; received in revised form 16 November 2010; accepted 17 November 2010

Available online 27 December 2010

Abstract

In size-mismatched thin film interdiffusion couples diffusion-induced recrystallization (DIR) occurs rather than conventional Fickian atomic transport. Grains formed in this process have characteristic composition levels that are so far not understood. In this work, DIR is studied in sputter-deposited Ni/Pd films. By pre-alloying one side of the diffusion couple, the mismatch, and thus the driving force, are varied. After heat treatment, transmission electron microscopy, energy-dispersive X-ray spectroscopy and X-ray diffractometry demonstrate recrystallization. Characteristic concentration levels are derived from X-ray diffraction data. Remarkably, the concentration inside newly formed grains shifts coherently to the concentration inside the parent layers. We demonstrate that the observed concentration levels are in agreement with a recently published thermomechanical model.

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Keywords: Metallic thin films; Diffusion-induced recrystallization; X-ray diffraction (XRD); Ideal shear strength; NiPd

1. Introduction

Atomic transport in size-mismatched thin film layer structures represents a complex phenomenon. Particular intricacy arises from the grain structure of the related materials. Even epitaxially grown diffusion couples, initially single crystalline, may transform into a polycrystalline microstructure by diffusion-induced recrystallization (DIR). Newly formed high-angle grain boundaries (GBs) serve as short-circuit transport paths. Moreover, the boundaries themselves may migrate through the volume (diffusion-induced grain boundary migration (DIGM)) so that the bulk gets completely intermixed, although volume diffusion can be frozen by low experimental temperatures.

The described processes obviously affect the thermal stability of nanostructures. In view of this, it is remarkable that our physical understanding of the phenomena is nevertheless incomplete. There is strong evidence that DIGM is related to elastic mismatch strain ahead the moving GB [1]. Recently, Penrose developed a stringent mathematical

model that describes composition profiles and driving forces [2] at the moving boundary. He was able to demonstrate that steady-state solutions of the composition profile perpendicular to the moving front require a constant composition level behind the front.

The DIR process is far less well understood [3], although it was reported many decades ago [4,5]. In analogy to plastically deformed materials, recrystallization is not that surprising after sufficient stress and lattice defects have been built up by interdiffusion, although the event of nucleation is still not understood even in classical recrystallization. Most remarkable, however, is the fact that grains that are newly formed by DIR do not reveal a continuous variation of local concentration as one may expect from Fickian diffusion. Instead, clearly distinguished concentration levels dominate the region of interdiffusion. Furthermore, these preferred concentration levels appear to be characteristic for a given combination of materials. Explaining these compositions remains a challenge. It is important to note that an understanding of the concentration characteristics will presumably also help to predict whether DIR or conventional interdiffusion can be expected for a given combination of materials.

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For the frequently investigated case of zincification, Kajihara and Gust [6] tried to justify the observed composition of new grains by the principle of maximum chemical driving force. However, the agreement with experimental data was not fully convincing. More recently, DIR in solid-state diffusion couples has attracted attention. Hartung and Schmitz [7] investigated DIR phenomena in epitaxially grown Au/Cu thin film reaction couples. During annealing several subsequent generations of grains could be distinguished by different preferred composition levels. This effect was confirmed in independent experiments with sputter-deposited Au/Cu multilayers [8]. Similar phenomena were demonstrated in size-mismatched thin film couples of Ag/Pd [9], Ni/Cu [10,11] and, in the case of reactive diffusion, also of Sn/Ag [10]. The latter two diffusion couples have been discussed in the literature [10] and the observed composition levels were justified – similar to the early experiments with zincification – by a modification of the model of maximum chemical driving force. By contrast, in the above-mentioned study of Cu/Au multilayers [8], the composition of the Au side of the diffusion couple was systematically varied by pre-alloying with Cu. It was shown that the composition of new grains formed at the Au-rich side shift coherently with the composition of the parent layer. It is not possible to justify this coherent shift of composition by the principle of maximum chemical driving force [8].

Therefore, an alternative interpretation of the preferred compositions in the interdiffusion region has recently been suggested: the composition in the growing grains is adjusted so that maximum elastic stress is achieved in the diffusion zone ahead of migrating GBs [12]. Since the stress is limited by the ideal strength of the respective matrix, this mechanical property of the material becomes an important controlling factor in the DIR process.

In this paper, we refer the details of an experimental study on the concentration characteristics of DIR performed at Ni/Pd couples in order to check the validity of the suggested model [12]. Since only two preferred new grain compositions are observed, the interpretation of the experiments is clearer than in the above-mentioned more complex case of Cu/Au. With about 10%, the lattice mismatch between both face-centered cubic (fcc) metals is smaller than with Au/Cu, but nevertheless sufficient to expect the occurrence of DIR or DIGM. Furthermore, the phase diagram predicts complete miscibility over the whole range of composition which avoids any influence of potential intermetallic compounds. The annealing temperatures used are higher than the Curie temperature T_C of Ni and its alloys with Pd. Thus, also any pronounced magnetic effects can certainly be excluded.

In addition to diffusion experiments between the pure metals, couples are also studied, in which one side was pre-alloyed with considerable amounts of the respective second component, to shed light onto the nature of the finite concentration gap between initial layers and new grains. The observed concentration levels are quantitatively

compared with the suggested interpretation of maximum possible stress ahead of the migrating boundaries.

2. Experiments

Multilayers of the symmetrical sequence (100 nm Ni/200 nm Pd/200 nm Ni/100 nm Pd) were prepared by ion-beam sputter deposition on thermally oxidized silicon substrates in an ultra-high vacuum chamber of 10^{-6} Pa background pressure. To modify the initial mismatch in atomic size ($\eta = 9.9\%$), we also prepared couples with pre-alloyed Ni layers Ni_{0.95}Pd_{0.05} ($\eta = 9.1\%$) and Ni_{0.88}Pd_{0.12} ($\eta = 8.3\%$) and, as a fourth type of samples, with pre-alloyed Pd layers Ni_{0.08}Pd_{0.92} ($\eta = 9.4\%$) (see also Table 1).

Isothermal heat treatments were performed at 723 K in a salt bath. Subsequent to heat treatment, the samples were quenched in iced water. To protect the multilayers from direct contact with the liquid salt, samples were densely wrapped in thin Al foils. This method allows a quasi-instantaneous contact heating and cooling of the sample to enable short annealing times with a precision of 2 s. Electron-transparent cross-sections were prepared from selected samples by mechanical polishing, dimple grinding and ion-milling. They were investigated by transmission electron microscopy (TEM) and energy-dispersive X-ray spectroscopy (EDX) to analyze the microstructure. The TEM investigations were performed using a Zeiss Libra 200 kV FEG microscope equipped with a Noran EDX detector. Since microscopic analysis suffers from bad statistics, preferred compositions in the diffusion zone were accurately measured by X-ray diffractometry (XRD) utilizing the lattice parameter variation with local composition. In this way, a volume average over the diffusion zone is naturally achieved. For this diffractometry, a Bragg–Brentano ($\theta - 2\theta$) geometry and Cu K $_{\alpha}$ radiation ($\lambda = 0.1541$ nm) were used.

3. Experimental results

During sputter deposition, the Ni and Pd layers grow with a (111) wire texture, i.e. the densely packed layers are predominantly arranged parallel to the surface of the substrate. The microstructure of the deposited layers was checked by TEM. Columnar grains grow across the layers perpendicular to the interfaces. This is an evidence for epitaxial growth with semi-coherent interfaces.

During diffusion annealing, the microstructure transforms completely. EDX demonstrates that new grains formed at the interface are distinguished in composition.

Table 1
Composition of thin film diffusion couples, lattice constant of the Ni-rich and the Pd-rich alloys, and lattice mismatch in between.

	a_{Pd} (nm)	a_{Ni} (nm)	η (%)
Ni/Pd	0.389	0.352	9.9
Ni _{0.95} Pd _{0.05} /Pd	0.389	0.355	9.1
Ni _{0.87} Pd _{0.13} /Pd	0.389	0.358	8.3
Ni/Pd _{0.92} Ni _{0.08}	0.387	0.352	9.4

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