Available online at www.sciencedirect.com
SciVerse ScienceDirect
Acta Materialia 61 (2013) 4984–4995
Acta MATERIALIA
www.elsevier.com/locate/actamat

Mechanical behavior of nanoscale Cu/PdSi multilayers

I. Knorr [a,*], N.M. Cordero [b,c,1], E.T. Lilleodden [b,d], C.A. Volkert [a,b,*]

[a] Institute for Materials Physics, Georg-August-University Göttingen, Friedrich-Hund-Platz 1, D-37077 Göttingen, Germany
[b] Institute for Applied Materials, Karlsruhe Institute of Technology, Hermann-von-Helmholtz-Platz 1, D-76344 Eggenstein-Leopoldshafen, Germany
[c] MINES ParisTech, Centre des Matériaux, CNRS UMR 7633, BP 87, 91003 Evry Cedex, France
[d] Institute of Materials Research, Materials Mechanics, Helmholtz-Zentrum Geesthacht, Max-Planck-Straße 1, D-21502 Geesthacht, Germany

Received 22 March 2013; accepted 10 April 2013
Available online 3 June 2013

Abstract

Berkovich nanoindentation and uniaxial microcompression tests have been performed on sputter-deposited crystalline Cu/amorphous $Pd_{0.77}Si_{0.23}$ multilayered films with individual layer thicknesses ranging from 10 to 120 nm. Elastic moduli, strengths and deformation morphologies have been compared for all samples to identify trends with layer thicknesses and volume fractions. The multilayer films have strengths on the order of 2 GPa, from which Cu layer strengths on the order of 2 GPa can be inferred. The high strength is attributed to extraordinarily high strain hardening in the polycrystalline Cu layers through the inhibition of dislocation annihilation or transmission at the crystalline/amorphous interfaces. Cross-sectional microscopy shows uniform deformation within the layers, the absence of delamination at the interfaces, and folding and rotation of layers to form interlayer shear bands. Shear bands form where shear stresses are present parallel to the interfaces and involve tensile plastic strains as large as 85% without rupture of the layers. The homogeneous deformation and high strains to failure are attributed to load sharing between the amorphous and polycrystalline layers and the inhibition of strain localization within the layers.

Keywords: Multilayer thin film; Nanoindentation; Microcompression; Mechanical behavior; Strength

1. Introduction

Metallic glasses (MG) offer high strength [1], good wear and abrasion resistance, as well as good corrosion resistance, making them promising candidates for many technological applications [2]. Nanocrystalline metals also show high strengths due to their small grain sizes, and are promising candidates for various applications, especially as thin film laminates [3–13]. However, both materials suffer from limited ductility. Metallic glasses fail via shear band formation after almost no global plasticity. Nanocrystalline metals show reduced ductility compared with their large grain counterparts because of inhibited dislocation-mediated plasticity [14]. Interestingly, combining both materials in a layered system with individual nanoscale layer thicknesses offers promising composite properties, which can include high strength as well as high strains to failure [15–17], also under tension [15]. These superior composite properties are attributed to the inhibition of shear bands in nanoscale and mechanically constrained MG samples [18–21], and to the mutual elastic and plastic constraints at strong interfaces [15,22].

The aim of this work is to study the effects of individual layer thicknesses on elastic modulus, strength, strains to failure and deformation mechanisms of multilayers consisting of nanocrystalline Cu layers and $Pd_{0.77}Si_{0.23}$ metallic glass layers. In particular, the effect of the amorphous/crystalline interfaces on the transfer of local shear strains, such as shear transformation zones or impinging dislocations,

* Corresponding authors. Address: Institute for Materials Physics, Georg-August-University Göttingen, Friedrich-Hund-Platz 1, D-37077 Göttingen, Germany. Tel.: +49 551 39 5011; fax: +49 551 39 5000.
E-mail addresses: iknorr@ump.gwdg.de (I. Knorr), volkert@ump.gwdg.de (C.A. Volkert).
[1] Present address: Saint-Gobain Recherche, 39 quai Lucien Lefranc, BP 135, F-93303 Aubervilliers, France.

1359-6454/$36.00 © 2013 Acta Materialia Inc. Published by Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.actamat.2013.04.047

and on the inhibition of strain localization by load sharing will be investigated.

2. Experimental details

$Cu/Pd_{0.77}Si_{0.23}$ multilayer films were deposited on oxidized single-crystalline Si(1 0 0) substrates by Ar-ion beam sputtering from a 99.99% pure Cu target (Goodfellow) and a PdSi alloy target [23]. The total film thickness is approximately 3 μm while the individual layer thicknesses range from 20 to 120 nm for polycrystalline Cu and from 10 to 90 nm for amorphous PdSi. The top and bottom layers are Cu for most samples. Three different series of samples were produced (see Table 1): series 1, with a constant PdSi layer thickness of 10 nm; series 2, with a constant Cu layer thickness of 90 nm; and series 3, with equal layer thicknesses. The samples are designated by the Cu and PdSi nominal layer thicknesses in nanometers; for example, Cu40/PdSi10 is used to describe a 3 μm thick film composed of alternating layers of 40 nm of polycrystalline Cu and 10 nm of amorphous $Pd_{0.77}Si_{0.23}$.

Berkovich nanoindentation was performed using MTS XP and MTS G200 nanoindenters to evaluate indentation hardness and modulus. All data were corrected for load frame and indenter compliance. The tip area functions were calibrated on fused silica samples that have well-known Young's modulus and hardness. The continuous stiffness technique was applied (using a harmonic displacement amplitude of 1 nm and a frequency of 37 Hz, which was observed to have the lowest noise level) during loading with a constant indentation strain rate of $0.05\ s^{-1}$. The Young's modulus was taken as the mean value at a penetration depth of 100–200 nm, whereas the hardness was taken as the mean value at a penetration depth of 200–300 nm. A minimum of nine indents were made on each sample to a maximum indentation depth of 2 μm, as well as a minimum of three indents to a smaller indentation depth of 200 nm. After indentation, the 200 nm indents were imaged by scanning electron microscopy (SEM) and a pile-up-correction was performed. Furthermore, cross-sections of some indents were milled using a 30 keV focused Ga beam in a focused ion beam (FIB) microscope (FEI Nova Nanolab) to analyze the multilayer deformation under the indentation and especially in the pile-up region using either SEM or FIB imaging. Additionally, lamellae were prepared from some of the Berkovich indents using FIB and analyzed in a transmission electron microscope (CM30, 300 keV, twin geometry).

In addition to the nanoindentation experiments, mechanical characterization of the multilayers was also carried out using the microcompression method. Pillars with diameters of 1.5 μm were milled through the film thickness using FIB, and tested with the nanoindenter machine using a flat punch tip at a loading rate of 20–50 $\mu N\ s^{-1}$. The pillars were milled down to the Si substrate, giving an aspect ratio of approximately 2 for all samples. In comparison with previous milling procedures [24], an additional milling step was added in this study to reduce the shoulder at the base of the pillars. This allows straighter pillar sides at the cost of a trench in the Si around the pillar. Given the high strength and negligible ductility of Si, it is assumed that plastic deformation does not occur in the substrate despite the stress concentrations from the trench. The final pillar structure is a slightly tapered (2–3.5°), with a well-defined height equal to the original film thickness, and was self-similar for all Cu/PdSi samples. The engineering stress, $\sigma = F/A_t$, was calculated from the measured force F and the pillar cross-sectional area A_t at 10% of its height beneath the pillar's top, since deformation primarily occurs in this part of the pillar. In order to avoid possible effects from early plasticity, and also to avoid large contributions from strain hardening, a compromise plastic engineering strain of 5% was used to determine the flow strength. Moreover, at 5% plastic strain the shape of the pillars has not changed significantly, so that the engineering stress calculated from the top diameter of the pillar before compression is a good measure of the true flow strength. For determination of the engineering strain, the displacement was first corrected for the compliance of the underlying Si substrate (see Refs. [18,24] for more details), then divided by the film thickness. Although the accuracy of stress and strain assessment by microcompression experiments is a topic of controversial discussion, it has been shown that the determination of flow stresses at small plastic strains are little affected for the aspect ratio and taper values used in this study [25,26]. We ensured the comparability of the results for the different Cu/PdSi samples by using of self-similar pillars.

For determination of the Young's modulus, microcompression tests with multiple load–unload cycles were conducted. Young's modulus values were computed from the initial unloading stiffness S as evaluated with the Oliver–Pharr method [27] and according to the equation $E = S\ H_0/A_0$, where A_0 is the cross-sectional area at the pillar at half-height and H_0 is the initial height. The measured moduli showed an increase with applied plastic strain presumably due to an increase in cross-sectional area as well as to time-dependent (or strain-dependent) flow or creep of the pillars. A careful examination of the unloading–reloading segments reveals significant hysteresis, particularly at larger total strains, indicating that the multilayers continue to deform during unloading and lead to an artificial

Table 1
Overview of the layer thicknesses of the three series of multilayer films investigated in this study.

PdSi (nm)	Cu (nm)							
	20	40	50	60	70	80	90	120
10		S1	S1	S1	S1	S1		S1
20	S3						S2	
50			S3				S2	
90							S2, S3	

S1: Series 1, S2: Series 2, S3: Series 3.