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ScienceDirect

Acta Materialia 64 (2014) 100–112

Acta MATERIALIA

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Interface structure of Nb films on single crystal MgO(100) and MgO(111) substrates

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Received 21 September 2012; received in revised form 3 November 2013; accepted 10 November 2013
Available online 20 December 2013

Abstract

This study systematically investigates the interface structure of Nb films grown on MgO substrates with different orientations ((100) and (111)) by experiments and simulations. X-ray diffraction, transmission electron microscopy (TEM) and high-resolution TEM (HRTEM) were used to characterize the structure of Nb films and the structure of interfaces between Nb films and MgO substrates. The results show that thin films exhibit different preferred planes on different orientations of MgO substrates. First-principles calculations were used to understand the interface configuration through a coherent interface model. The combination of experiments and simulations shows that the work of separation, together with substrate orientation and lattice mismatch, determines the interface structure between films and substrates.

Keywords: Interface structure; TEM and HRTEM; First principles; Lattice mismatch; Work of separation

1. Introduction

Studies on interfaces in composite materials have attracted much attention in modern materials science since these materials have practical applications in many fields, such as coatings, sensors, photovoltaic devices, microelectronics devices, superconductors and catalysts [1–6]. Recently, new applications of interfaces concerning the enhancement of radiation tolerance of materials by manipulating interfaces, which act as powerful sinks for point defects and assist the recombination process between interstitials and vacancies, have been reported [7–14]. The interfaces in thin film materials play key roles in controlling their epitaxial quality, mechanical, physical, chemical, structural and functional properties [15–17]. As one of the major types of interfaces, metal/oxide interfaces exhibit specific properties, including magnetic, catalytic and tribological properties, and have been widely studied both in experiments and in simulations [1,18]. A fundamental understanding of interface structure at the atomic scale is considered to be necessary for the further development of film properties. Atomic observations of many metal/oxide interfaces, such as Au/MgO, Ag/CdO, Nb/Al_2O_3, Cu/Al_2O_3, Pt/NiO and Cu/NiO, have been reported [1,19–22]. As a major component of a number of metal/oxide interfaces, single crystal MgO is widely used as a substrate

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1359-6454/$36.00
http://dx.doi.org/10.1016/j.actamat.2013.11.031

material because it has a simple sodium chloride structure and clean MgO surfaces are easy to obtain. Epitaxial growth of metals on single crystal MgO has frequently been reported. Lu and Cosandey studied the interface structure in metal/MgO(100) interfaces by electron microscopy [23,24]. They concluded that Cu/MgO and Pd/MgO(100) interfaces are partially coherent and the contrast variation in high-resolution transmission electron microscopy (HRTEM) images is due to interfacial dislocations. They defined the dislocation structure of (001) and (111) interfaces based on coincidence-site lattice (CSL) and displacement shift complete (DSC) lattice models [23–26]. Trampert et al. reported HRTEM studies of the Ag/MgO interface made by molecular beam epitaxy (MBE) growth and revealed the interface structure by experiments and simulations [27]. The Ag/MgO(100) couple exhibits a preferential orientation of the Ag(100) plane parallel to the (100) surface of the MgO substrate. HRTEM shows a square network of edge-type dislocations with [100] line directions and Burgers vectors of the type $b = 1/2$Ag[100] [27]. Comparative theoretical studies of Ag/MgO(100) and Ag/MgO(110) interfaces concluded that chemical bond formation is not important for either Ag/MgO(100) or Ag/MgO(110) perfect interfaces, and the physical adhesion associated with polarization and charge redistribution are the dominant effects [28]. Chen et al. studied the interfaces of Cu/MgO(111) and Pd/MgO(111) by HRTEM and concluded that their interfaces have a cube-on-cube orientation relationship. A periodic localized interfacial dislocation contrast in both Cu/MgO(111) and Pd/MgO(111) interfaces was observed along the MgO[110] direction [29].

While the interface structures of metal/MgO thin film couples have been extensively studied, most of the studies have focused on face-centered cubic (fcc)/MgO interfaces, with few studies being performed on the interfaces of body-centered cubic (bcc)/MgO. Ikuhara et al. studied the atomic structure of MgO/V/MgO multilayer films grown by MBE, and they reported on the orientation relationship and interfacial dislocations between bcc V and MgO(100) in multilayer films [30]. Another study focused on the atomic and electronic structure of V/MgO interfaces with the V film thickness of 5 nm [31]. The comparison between the experimental and theoretical details by molecular orbital calculation found the presence of a hybridized orbital of V3*d* with Mg3*p* [31]. However, the detailed atomic structure of the interface and the fundamental mechanisms for forming orientation relationships of the interface in bcc/MgO thin films have not been widely studied and are not well understood. Furthermore, to the best of our knowledge, very few studies on the atomic interface structure of bcc metals on single crystal MgO(111) have been reported. In the present paper, the interface structures of Nb films grown on MgO(100) and MgO(111) substrates are systematically investigated by the combination of experiments and simulations. The studies reveal fundamental mechanisms that control the orientation relationships of interfaces in both thin films.

2. Experimental

Nb thin films were deposited on single crystal MgO(100) and MgO(111) substrates by means of electron-beam evaporation. The substrate temperature was set at 950 °C, and the deposition rate was 5 Å s^{-1}. The thickness of Nb thin films was 180 nm. The purity of the Nb evaporation target was 99.999%. The pure single crystal MgO(100) and MgO(111) substrates have dimension of 10 mm × 10 mm × 0.5 mm. The misorientation of (100) in MgO(100) substrate and (111) in MgO(111) substrate is less than 0.5°. The surface of substrates was polished by chemical mechanical polishing (CMP) technology with minimum sub-surface damage and possessed a surface roughness less than 1 nm. The substrates were immediately loaded into a vacuum chamber for thin film deposition to avoid surface roughening and hydroxide formation after they were removed from a clean plastic bag packed under 1000 class clean room conditions.

X-ray diffraction (XRD) analysis was used to characterize the Nb thin film structure and the orientation relationship between the film and the substrate. TEM experiments were performed on an FEI Tecnai F30 transmission electron microscope operated at 300 kV with a field-emission gun, and images were recorded by a Gatan SC1000 ORIUS CCD camera with image size of 2048 × 2048 pixels. To identify interface structure at atomic resolution, HRTEM was carried out on cross-sectional TEM (XTEM) specimens in the same microscope. The point-to-point resolution of this microscope is 0.17 nm. Electron diffraction was used to identify the orientation relationships between Nb films and MgO substrates. HRTEM images were processed by using the image processing software of Gatan Digital Micrograph with fast Fourier transform (FFT) algorithm. Specimens with MgO(100) substrates were cut along the MgO(010) plane or MgO(001) plane and specimens with MgO(111) substrates were cut along the MgO($1\bar{1}0$) or MgO($11\bar{2}$) plane, which are all perpendicular to the interfaces, respectively, in order to observe interface structure easily at in-zone conditions in TEM. Samples were glued face to face with M-610 bond at room temperature. XTEM specimens were prepared by using a dimple grinder on a MultiPrep System followed by low energy ion milling with Ar ions at low angle on a Gatan PIPS System to reach electron transparent.

3. Results

3.1. General features of interface structure

Nb has a bcc structure with a lattice parameter of 0.3307 nm and MgO has a sodium chloride structure with a lattice parameter of 0.4217 nm. The binary phase