Available online at www.sciencedirect.com
SCIENCE DIRECT
Surface Science 599 (2005) 255–261
SURFACE SCIENCE
www.elsevier.com/locate/susc

Evolution of Fermi level position and Schottky barrier height at Ni/MgO(001) interface

Y.Y. Mi [a], S.J. Wang [b,*], Y.F. Dong [a], J.W. Chai [b], J.S. Pan [b], A.C.H. Huan [b,c], C.K. Ong [a]

[a] Department of Physics, National University of Singapore, Singapore 117542, Singapore
[b] Institute of Materials Research and Engineering, Materials Science and Characterization Laboratory, 3 Research Link, Singapore 117602, Singapore
[c] Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore 637616, Singapore

Received 26 May 2005; accepted for publication 11 October 2005
Available online 10 November 2005

Abstract

Fermi level evolution and Schottky barrier height (SBH) for Ni/MgO(001) interfaces have been studied using X-ray photoelectron technique. It was found that upward band bending occurred at the initial Ni growth stage and Ni bulk properties recovered after 6–8 Å thickness. The measured Schottky barrier heights are strongly interface structure-dependent, with the variation in the range of 3.1 eV for perfect interface to 1.6 eV for defect-rich interface. First-principles calculations on the evolution of SBH for the initial growth of Ni on perfect MgO(001) surface are combined with experimental results to investigate the underlying microscopic mechanism. Adatom-induced states (or interfacial bonding states), MIGS and defects states were used to rationalize the evolution of Fermi level and corresponding SBH for various interface structures. This work shows that SBH can be engineered by interface structure control, and is expected to shed light on the effect of interface structures on the formation mechanism of SBH at metal/oxide interface.

Keywords: Nickel; Magnesium oxides; Metal–oxide interfaces; Schottky barrier; Interfaces states; X-ray photoelectron spectroscopy

1. Introduction

The wide applications of metal/oxides systems, particularly in magnetic media devices and heterogeneous catalysts, have stimulated many experimental and theoretical studies on this system

* Corresponding author. Tel.: +65 6874 8184; fax: +65 6872 0785.
E-mail address: sj-wang@imre.a-star.edu.sg (S.J. Wang).

0039-6028/$ - see front matter © 2005 Elsevier B.V. All rights reserved.
doi:10.1016/j.susc.2005.10.010

[1–4]. In particular, more and more researchers notice the importance of interface structure for attaining desirable performances of metal–oxides contacts and start to study the interfacial properties of this system [5–9]. For example, the enhanced catalytic activation of nanoscale gold catalysts supported on various oxides was reported to be related to the interface structure [7]; the influence of the electronic structure of metal–oxide interfaces on the spin polarization opens new ways to optimize the magnetoresistance of tunneling junctions [8]. Although large efforts have been addressed to understand the underlying mechanisms, it is still far from achieving the complete and fundamental microscopic mechanisms describing the interface structure in metal–oxides system [6,10]. More theoretical and experimental studies are urgently required in this field.

MgO(0 0 1) is much suitable for both theoretical and experimental study. Its stoichiometry and rigid crystalline atomic structure can simplify the theoretical models [1,9]. Meanwhile, the wide band gap, high chemical and thermal stabilities of MgO promise its applications in magnetic tunneling junctions (MTJs) and heterogeneous catalysis [3,11]. Ni/MgO(0 0 1) is the case of transition metal on insulating ionic metal oxides. Several theoretical models have been proposed to explore the interface electronic structure of Ni/MgO(0 0 1) contacts [9,12,13], but existing experimental studies were still mainly restricted to the growth mechanism and magnetic properties [14–18]. More specific set of experimental results on interfacial electronic properties are required for the complete understanding of properties and application of this system. One fundamental parameter characterizing the metal/oxides interfacial electronic properties is Schottky barrier height (SBH), which is defined as the energy separation between the Fermi level of metal and the edge of the majority carrier band of oxide right at the interface [19]. It is believed that leakage characteristics of microelectronic devices are strongly influenced by SBH at metal/insulator junctions. Further study on Fermi level evolution and resulting SBH at metal/oxide contacts is not only necessary for industrial applications but also significant for understanding of the interface electronic structure of this system. To this end, we studied the Fermi level evolution and SBH of Ni/MgO(0 0 1) with different interface structures by monitoring the core-level and valence-band shift observed from X-ray photoelectron spectroscopy (XPS). Meanwhile, the evolution of SBH for the initial growth of Ni on perfect MgO(0 0 1) surface has also been studied by first-principles calculations based on density-functional theory (DFT), which aims to explore the microscopic mechanisms for the band alignment at the Ni/MgO(0 0 1) interfaces.

2. Experimental and theoretical methods

In our experiment, Ni metal was deposited from a high purity (+99.99%) rode in the Omicron EFM3 e-Beam evaporator with a base pressure of 5×10^{-10} mbar. This evaporator was mounted to UHV preparation chamber in X-ray photoelectron spectroscopy (XPS) system. Before the deposition of Ni, high-purity polished single crystal MgO(0 0 1) substrates were ultrasonically cleaned for 10 min in acetone solvent. After cleaning the substrates, they were introduced to UHV chamber for subsequent pretreatment: one sample was annealed at 673 K for 10 min in UHV chamber in order to effectively remove the carbon contamination on the surface of substrate. For the other two samples, surfaces were sputtered in situ by Ar^+ ion beam at 1 keV for different sputtering time (5 and 10 min), producing defect-rich surfaces on MgO(0 0 1) substrates. Nickel was deposited stepwise at room temperature, and the deposition rate of nickel was found to be 1 Å/min based on the average deposited thickness, which was calibrated by cross-sectional high resolution transmission electron microscopy (HRTEM). After the metal was deposited at each step, core-levels (Ni2p, Mg2p and O1s) and valence-band spectra were recorded immediately. The XPS measurement was performed in VG ESCA Lab XL-200i system. Photoelectrons were excited by the monochromatic AlKα source ($h\nu = 1486.6$ eV), and constant pass energy mode with 10 eV was used in the analyzer to increase the energy resolution. One experimental difficulty encountered on our samples is the peak broadening and shifting due to surface