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The angle-resolved photoemission study for ultrathin NiO and CoO thin films on Ag(1 0 0) surfaces

C.-M. Cheng[a,*], C.-C. Wang[b], H.-T. Jeng[c,d], C.S. Hsue[c], B.Y. Hsu[a], D.-J. Huang[a,c], K.-D. Tsuei[a,c]

[a]*National Synchrotron Research Center, Hsinchu 300, Taiwan*
[b]*Department of Physics, National Taiwan University, Taipei 106, Taiwan*
[c]*Department of Physics, National Tsing-Hua University, Hsinchu 300, Taiwan*
[d]*Insititute of Physics, Acdemia Sinica, Taipei 115, Taiwan*

Abstract

We have performed a high resolution angle-resolved photoemission study on ultrathin NiO and CoO films on Ag(1 0 0) surfaces. The NiO and CoO thin films were grown on Ag(1 0 0) at room temperature and 180 °C, respectively, for an O_2 pressure of 7.5×10^{-7} Torr. The thickness of thin film was determined by a quartz monitor and RHEED. A clearest (2×1) low-energy electron diffraction (LEED) pattern was observed at a NiO coverage of 0.9 monolayer(ML), consistent with literature. LEED shows a clear (1×1) pattern indicating well-ordered domains for the 2 ML NiO and CoO thin films. Angler-integrated photoemission spectra (AIPES) show a finite density of states at the Fermi level for the 2 ML NiO thin film, suggesting a metallic character, while the 2 ML CoO thin film was insulating. Angle-resolved photoemission spectroscopy (ARPES) was used to detect the band dispersion of 2 ML NiO and CoO thin films. Very different band dispersions of the valence bands were observed. The insulating nature of bulk NiO and CoO is related to their antiferromagnetic order, but the unusual metallic behavior of ultra NiO thin film may affect its magnetic order. The detail comparison of electronic structure for 2 ML NiO and CoO thin films will be discussed.

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1. Introduction

The electronic structure of transition metal oxides thin films on metal substrates has been an important topic in recent years. Much interest focused on the potential of NiO thin films to be used in magnetic nanodevices technology. Monoxides such as NiO and CoO are also prototype compounds to study the strongly correlated electron systems. The geometric structure, electronic properties and magnetic order of these oxides with several atomic layers on metal surfaces are expected to be very different from the bulk oxides. The study of these relatively simple oxides on metal surfaces is significant to understand how to tailor these interesting properties of ultrathin films of more complex oxides and other strongly correlated electron systems [1,2].

2. Experiments and discussion

The experiment was carried out at the National Synchrotron Radiation Research Center in Hsinchu, Taiwan using BL21B1 U9-CGM Spectroscopy Beamline. The photoemission spectra were measured in a UHV chamber equipped with a Scienta SES-200 hemispherical analyzer with ±8° collecting angle. An angular resolution of 0.5° was chosen for the angle-resolved measurements. All spectra are collected with 51 eV photons and $HeII_\alpha$(40.8 eV) at $T = 40$ K and the overall energy

*Corresponding author. Tel.: +886 3 5780281x7331; fax: +886 3 5783813.
E-mail address: makalu@nsrrc.org.tw (C.-M. Cheng).

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resolution was 15 meV. The Ag(1 0 0) crystal was cleaned by the standard sputtering and annealing process. Well-ordered thin films were prepared by evaporating Ni and Co in an oxygen atmosphere onto Ag(1 0 0) [3], where the O_2 pressure was set at 7.5×10^{-7} Torr [3]. The substrate temperature was kept at 180 °C and RT during growth for CoO and NiO, respectively. The thickness of these thin films was determined by a quartz thickness monitor and RHEED. A clearest (2×1) low-energy electron diffraction (LEED) pattern for the 0.9 monolayer(ML) NiO thin film was also used to calibrate the thickness. The post annealing in an O_2 atmosphere helped the oxidization of residual Ni and Co and led to more ordered films as revealed by LEED. A 10 ML thick film was also grown to compare to the bulk spectra.

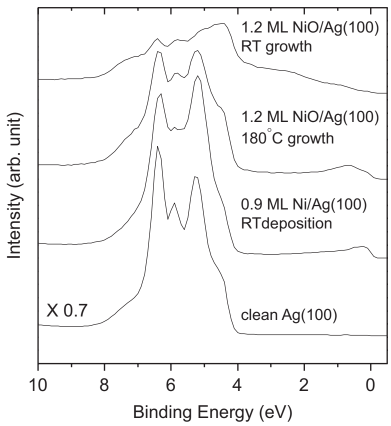

Fig. 1. AIPES of clean Ag(1 0 0), 0.9 ML NiO/Ag(1 0 0) as well as 1.2 ML NiO/Ag(1 0 0) grown at 180 °C and at RT with post annealing, all taken spectra at normal emission using $HeII_\alpha$ at RT.

As seen in Fig. 1 of Angler-integrated photoemission spectra (AIPES), for 0.9 ML Ni deposited on Ag substrate, the 3d band crosses the Fermi level. For the 1.2 ML NiO thin film grown at 180 °C, the Ni induced peak shifts below the Fermi level at about 0.7 eV but still has high intensity at the Fermi level. On the contrary, for the 1.2 ML NiO film grown at RT, the Fermi level intensity diminishes, similar to bulk NiO, and the intensity of the Ag 4d band is much more attenuated than that of the film grown at 180 °C. A

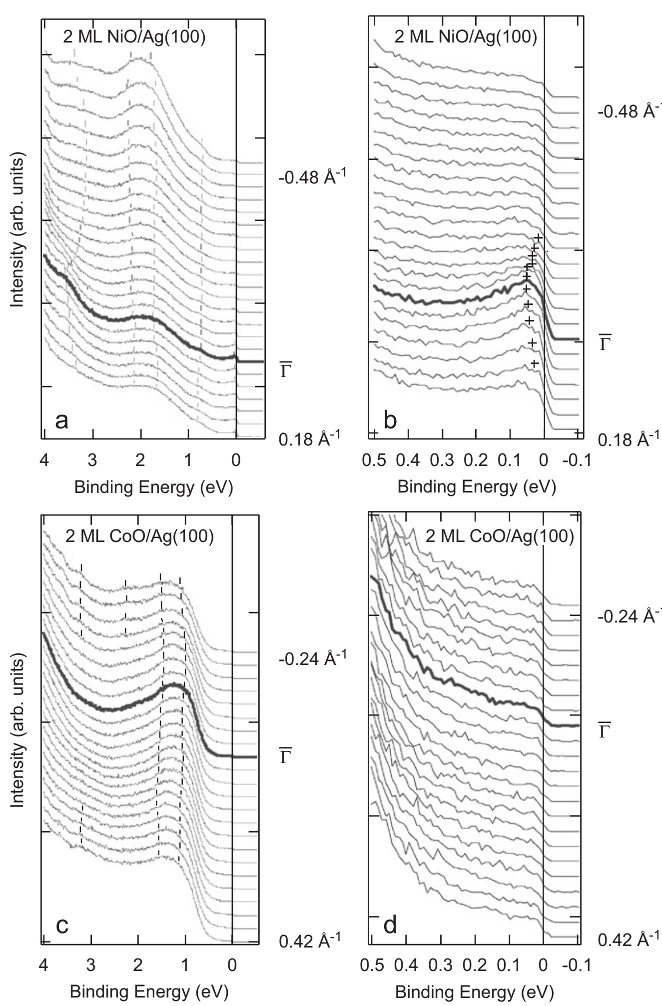

Fig. 2. ARPES spectra of (a) the valence band of 2 ML NiO/Ag(1 0 0) grown at RT also (b) the enlarged region near the Fermi level for (a), (c) valence band of 2 ML CoO/Ag(1 0 0) grown at 180 °C and (d) the enlarged region near the Fermi level for (c). The measurement is along $\bar{\Gamma}$–$\bar{M}$ direction, where $\bar{M}$ is referred to the (1×1) Ag surface Brillouin zone. All spectra were taken at 51 eV photo energy.

combined STM and photoemission study on Ni/Ag(1 0 0) showed that Ni clusters grow on Ag(1 0 0) surfaces and a large fraction of Ni may segregate into the subsurface region of Ag with increasing temperature of the substrate [4]. The stronger intensity of Ag 4d band and high intensity at the Fermi level for NiO film grown at 180 °C in Fig. 1 suggest less NiO on the Ag surface and more Ni in the subsurface with higher temperature of the substrate. We thus conclude that growth of ultrathin NiO films on Ag(1 0 0) at RT with less unreacted Ni is more suitable than at high temperature. Due to a bilayer growth in the O_2 atmosphere observed by STM [3,5], we study the electronic structure of 2 ML films of NiO and CoO. We have made a quantitative analysis on the contribution of the Fermi level intensity of the AIPES spectra of different thickness. We found that for 2 ML NiO thin film, there exist a finite density of state at the Fermi level, which indicates that this film contains a metallic character.

Angle-resolved photoemission spectroscopy (ARPES) was used to detect the electronic structure of the 2 ML NiO and CoO thin films on Ag(1 0 0) (Fig. 2), as well as 10 ML thin films. The results of 10 ML are consistent with bulk crystals [6,7]. A shallow band dispersive crossing the Fermi level was observed on a 2 ML NiO thin film, which indicated an metallic behavior, but there was no clear features near the Fermi level for a 2 ML CoO thin film. Because the insulating nature of bulk NiO and CoO is related to the antiferromagnetic order, the metallic character of the 2 ML NiO may affect the magnetic order. The XMLD result shows the antiferromagnetic order and strong linear dichroism effect for the 3 ML NiO thin film [1], different from the bulk crystal. A theoretical calculation may help elucidate the band dispersion and the metallic characters observed in our measurements, and the possible variation of the magnetic ordering in these thin films.

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