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The formation of sharp NiO(1 0 0)–cobalt interfaces

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Abstract

An atomically sharp interface between an antiferromagnetic oxide and a ferromagnetic metal may be obtained by the deposition of an epitaxial oxide buffer nanolayer in between. The buffer layer consists of the oxide of the ferromagnetic metal. The concept has been demonstrated on the NiO(1 0 0)–Co system, where the inclusion of a 1–2 ML CoO(1 0 0) interlayer inhibits the interfacial redox reaction which takes place between NiO and Co metal in the absence of the buffer layer.

Keywords: Nickel oxide; Cobalt; Cobalt oxide; XPS; LEED; STM

The combination of a ferromagnetic (FM) metal and an antiferromagnetic (AFM) oxide in a bilayer thin film system is of fundamental scientific interest and of considerable practical relevance due to its application in devices based on the exchange bias effect [1]. The physical origin of this effect, although it is known since 50 years [2] and already used in device technology, is still a matter of much debate. This is largely caused by the fact that the key element crucial for the effect, the FM–AFM interface, where the spin interaction responsible for the exchange bias occurs, is structurally and chemically complex and difficult to characterise experimentally. The FM–AFM materials of interest in exchange bias systems, such as the ferromagnetic metals iron and cobalt and the antiferromagnetic oxides NiO, MnO, or CoO, form so-called reactive interfaces between them: that is, the FM metal films cause the chemical reduction of the AFM oxide at the interface. This redox reaction leads to reduced metal cations of the oxide, to oxidised metal atoms of the metal layer, and to interfacial oxygen diffusion resulting in a complex intermixed region that may extend over several atomic layers. The interfacial reaction and diffusion modify the magnetic spin structure, which is sensitive to the chemical state, to the geometry and to the structural order at the interface, and this modification is difficult to control. For example, structural order at the FM–AFM interface is a desirable property which can be improved by elevated temperature treatment, but the latter also enhances the interfacial reactivity and thereby the width of the mixed interdiffused interfacial region [3,4].

Sharp FM–AFM interfaces are desirable for improved performance of the magnetoresistive properties in applied exchange biased systems as well as for theoretical simulations of the effect, since most models of the exchange bias coupling assume atomically abrupt interfaces. Here we show that a sharp interface between an antiferromagnetic oxide and a ferromagnetic metal overlayer can be obtained by the insertion of a magnetically active buffer nanolayer in between them. The system of study is cobalt on epitaxially grown NiO(1 0 0), which is a system of both practical relevance and scientific interest [5,6]. We show that, while Co metal overlayers react with NiO(1 0 0) surfaces to yield reduced NiO_{1-x} and oxidised CoO_x mixed interfaces, by controlled deposition of an ultrathin CoO buffer layer in between the NiO substrate and the Co metal overlayer the interfacial reaction is suppressed. The CoO buffer nanolayer (here 2 ML only) can be grown epitaxially with (1 0 0) orientation on the NiO(1 0 0) surface thus maintaining the substrate order for the subsequent deposition of the metal overlayer.

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Epitaxially ordered 10–20 ML thick NiO(1 0 0) films have been prepared under ultrahigh vacuum (UHV) conditions by reactive evaporation of Ni in 2×10^{-6} mbar O_2 onto a clean Pd(1 0 0) single crystal substrate at room temperature (RT). The deposition step was followed by annealing in 2×10^{-7} mbar O_2 at 850 K to give smooth, well ordered NiO surfaces with a root mean square roughness of the order of 0.2–0.3 nm, i.e. 1–2 monoatomic steps only [7]. Co and CoO overlayers were grown by evaporation of Co metal in UHV or in 2×10^{-6} mbar O_2, respectively, onto the NiO(1 0 0) surfaces at 100 K or at RT. X-ray photoelectron spectroscopy (XPS) with use of synchrotron radiation, low-energy electron diffraction (LEED), and scanning tunneling microscopy (STM) have been used to monitor the chemical state of the Ni and Co constituents and the surface order and morphology of the bi-(tri-)layer systems, respectively. The experiments with synchrotron radiation were performed at beamline I311 [8] in the Swedish synchrotron radiation laboratory MAX-Lab, Lund, whereas the STM measurements were carried out with a variable-temperature STM in our home laboratories in Graz [7]. A linear background has been subtracted from the XPS spectra displayed.

Fig. 1 displays XPS spectra which demonstrate that Co metal overlayers undergo redox reactions at the interface to NiO(1 0 0) surfaces. Fig. 1a shows Co $2p_{3/2}$ core level spectra of 0.75 ML of Co deposited on NiO(1 0 0) at 100 K, and after heating that surface to 550 K. The as-deposited Co overlayer displays a Co 2p XPS spectrum with a main peak at 778.5 eV and a broad tail towards higher binding energies. The structure at 778.5 eV is characteristic of metallic Co [9], but the tail towards higher binding energy is indicative of the beginning of the interfacial oxidation reaction already at the low temperature. Heating to 550 K leads to a significant change of the spectral profile, with the CoO_x structure at 780 eV now clearly apparent and also with the shake-up satellite (S) at around 787 eV visible, which is typical for the oxide and thus signals the oxidation of the cobalt atoms. The corresponding Ni $2p_{3/2}$ core level spectra of the NiO surface are shown in Fig. 1b. The Ni $2p_{3/2}$ core level spectrum of the pristine NiO(1 0 0) surface, grown epitaxially on Pd(1 0 0), is due to the direct emission from the Ni^{2+} species at 854.7 eV and to the oxide shake-up processes (S) at around 856.5 eV. After deposition of 0.75 ML Co at 100 K the Ni 2p core level signal remains, apart from some attenuation, almost unchanged. The weak XPS intensity at a binding energy of 852.8 eV, however, indicates the presence of some reduced Ni^0 species, thus corroborating the onset of the interfacial reaction at 100 K as derived from the analysis of the Co 2p spectra. The Ni 2p spectrum recorded after heating to 550 K shows the significant increase of the Ni^0 signal as a result of the reduction of NiO at the interface. The overall increase of the Ni 2p intensity after annealing reflects a morphology change of the Co overlayer and the intermixing of the NiO_{1-x} and CoO_x phases. The redox reaction processes at the Co–NiO interface are thus clearly established by the XPS core level spectra.

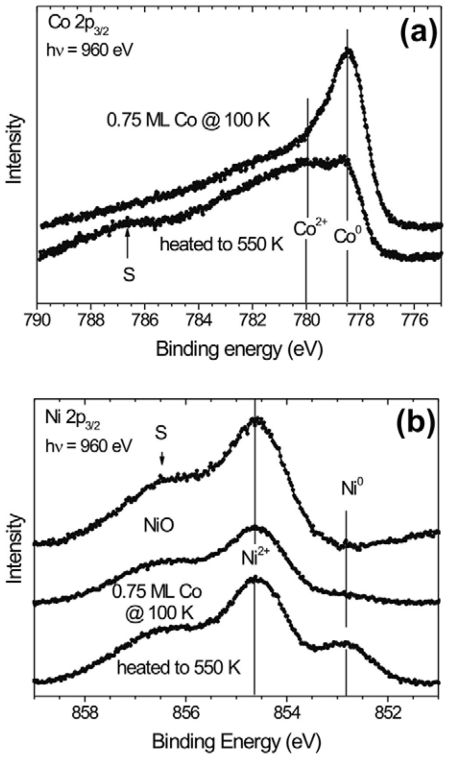

Fig. 1. Co $2p_{3/2}$ (a) and Ni $2p_{3/2}$ (b) XPS spectra of the pristine 20 ML NiO(1 0 0) surface, grown epitaxially on Pd(1 0 0), after the deposition of 0.75 ML Co metal on to the NiO(1 0 0) at 100 K, and after heating to 550 K. For the labelling of spectral features see the text.

Next, we demonstrate that a CoO(1 0 0) overlayer can be grown epitaxially on top of NiO(100). Fig. 2a and b contain Co 2p and Ni 2p core level spectra of NiO covered by 1 and 2 ML of CoO – the latter prepared by reactive evaporation of Co in oxygen atmosphere onto NiO(1 0 0) at RT followed by annealing in 1×10^{-6} mbar of O_2 at 600 K. The Co $2p_{3/2}$ spectra display the main Co^{2+} derived emission at ~780 eV binding energy and a charge transfer shake-up satellite at around 786 eV (S), which are both characteristic of CoO [9]. The intensity of the Ni 2p signal from the bare NiO(1 0 0) surface (top curve of Fig. 2b) becomes strongly attenuated upon deposition of 1 ML CoO, and is almost completely suppressed after a coverage of 2 ML CoO. The inset of Fig. 2b displays the decrease of the Ni 2p peak area with the CoO coverage. This decrease is exponential, which is a strong indication that the CoO overlayer covers the entire surface and grows in a layer-by-layer mode (note that the kinetic energy of the Ni 2p photoelectrons is of the order of 100 eV, thus ensuring a maximum surface sensitivity of the spectra). The LEED picture of Fig. 2c, displaying the (1 × 1) pattern of CoO(1 0 0), gives evidence of the epitaxial order of the CoO overlayer. Epitaxial growth of CoO on NiO(1 0 0) is actually not too surprising in view of the close lattice match of the two oxides (2% lattice mismatch). The STM image of Fig. 2d reveals the smooth morphology of the CoO(1 0 0) overlayer, with large terraces separated by rather straight