Nuclear Instruments and Methods in Physics Research B 269 (2011) 1175–1178

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Negative ions, energy loss, and electron emission during grazing scattering of fast H and He atoms from a clean and oxidized NiAl(1 1 0) surface

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ARTICLE INFO

Article history:
Received 22 September 2010
Received in revised form 17 November 2010
Available online 23 November 2010

Keywords:
Ion scattering
Metal surface
Thin oxide film
Electron emission
Energy loss

ABSTRACT

Negative ion fractions, projectile energy loss, and the emission of electrons is studied for grazing scattering of hydrogen and helium atoms/ions from a clean and oxidized NiAl(1 1 0) surface. Making use of translation energy spectroscopy and the coincident detection of the number of emitted electrons we have studied the electronic interaction mechanisms for the change from a clean metal target to an insulator surface via the preparation of a well defined ultrathin alumina film on top of the metal substrate. We find that already for a monolayer thick oxide film the characteristic different features of electronic processes for the surface of an insulator crystal are present.

1. Introduction

Electronic excitations induced by impact of fast atoms and ions on a surface show pronounced differences for metal and insulator targets [1]. Recent studies on this topic have shown that details on the interaction mechanisms are fairly intricate and closely related to specific features for the two different types of materials [2,3]. The use of translation energy spectroscopy for scattered projectiles in coincidence with the detection of emitted electrons [4] has provided insights into the microscopic scenario for electronic excitations during the collision [5]. In those studies it was found out that electronic excitation and emission phenomena during ion impact on a metal surface are dominated for grazing angle impact by binary encounters with conduction electrons [6]. For incidence under larger angles electron promotion in collisions with small impact parameters will also play an important role [7].

Following these concepts, a substantial energy loss owing to electronic excitations of conduction electrons is present for metal targets, and electron emission shows a defined threshold, since the energy transfer to electrons has to be sufficiently larger in order to overcome the work function of the target [8]. For insulators with a wide band gap such a mechanism will be inefficient so that another process has to be present in order to explain the experimentally observed substantial total electron emission yields. It was shown that the formation of transient negative ions, i.e. capture of electrons from anion sites of ionic crystals, plays a key role for electronic excitation and emission processes for insulators [9,10]. As a consequence, electronic energy loss and electron emission induced by ion scattering from metal and insulator surfaces show clearly different features.

In this work we report on studies where fast H and He atoms were scattered under a grazing angle of incidence from a NiAl(1 1 0) surface which has the electronic properties of a clean metal surface. In addition, this surface can be oxidized in a controlled manner resulting in a monolayer thick alumina film [11]. This film has a well defined but intricate structure which was cleared up recently in a combined effort of STM studies and DFT calculations [12]. Such oxide surfaces play an important role as support for catalytic active materials or in electronic devices, where ultra-thin films of a variety of oxides have attracted considerable interest, since studies based on established surface analytical tools can be performed in a controlled manner by keeping the electronic structure to a major extent unchanged [13]. One may also profit from specific differences concerning electronic states compared to bulk material [14]. We will show below that the change from the clean NiAl(1 1 0) surface to an ultrathin alumina film grown on the same substrate leads to a pronounced difference in charge transfer as well as electronic excitation and emission processes.

2. Experiment and results

In our experiments we have scattered H and He atoms with kinetic energies E_o in the keV domain from the surface of a NiAl(1 1 0)-crystal under grazing angles of incidence Φ_{in} ranging from about 0.2° to 3°. The target surface was prepared via cycles of grazing sputtering with 100 keV Ar^+ ions under $\Phi_{in} \approx 2°$ and subsequent annealing at about 1000 K for some minutes. NiAl

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doi:10.1016/j.nimb.2010.11.051

has a CsCl structure with a lattice constant of $a = 2.88$ Å. The topmost layer of the (1 1 0) face is formed by the superposition of rectangular unit cells formed by Ni and Al atoms. The base pressure in our UHV setup was in the mid 10^{-11} mbar range where the pressure gradient to the accelerator beam line with the gas target was maintained by two differential pumping stages. The energy loss of scattered atoms was recorded by means of a time-of-flight (TOF) method where the incident ion beams were chopped by electric fields and thereafter neutralized in a gas target operated with Kr (for H^+) or He (for He^+). Scattered projectiles were recorded about 80 cm behind the target with a micro-channel plate which provides the start signal for the TOF setup. Electrons are detected by a surface barrier detector biased to about 25 kV where the pulse heights for detection of accelerated electrons can be directly related to the emission of a specific number of electrons for impact of single atoms [15,16].

The ultra thin alumina film was produced by a dose of 1200 L O_2 (5×10^{-6} mbar for about 15 min) at a target temperature of 540 K and subsequent annealing at 1200 K for 5 min. Following this recipe [17] a well ordered and smooth oxide layer can be produced as demonstrated by the polar angular distributions for 30 keV H atoms shown in Fig. 1 recorded with a channeltron detector (entrance aperture 0.5 mm) about 60 cm behind the target which could be translated through the scattered beam by means of step motor driven precision linear feed through. The data for clean NiAl (full circles) and for the alumina film (open circles) reveal well defined angular distributions which reflects a well ordered surface with a low concentration of defects [18]. The slightly enhanced broadening of the distribution for scattering from the oxide film indicates an enhanced surface corrugation of the alumina film. This is attributed to the complex structure of the film with large surface unit cells [12,19], but might be partly caused also by a small amount of imperfections of the film surface.

In our experiments on charge transfer we have concentrated on the formation of negative hydrogen ions, since former experiments have demonstrated that H^- fractions are very sensitive to the electronic structure of the target surface [20]. As a consequence, for metal and insulator surfaces H^- fractions can differ by more than one order of magnitude [21]. In Fig. 2 we show normalized H^- fractions $n(H^-)/(n(H^-) + n(H^o))$ for impact of H atoms on clean NiAl(1 1 0) (full circles) and on an alumina film (triangles) under a grazing angle of incidence $\Phi_{in} = 1°$ as function of projectile velocity. The fractions show in both cases a kinematic resonance where for the scattering from the surface of the oxide film the fractions for negative ions are substantially higher than for the metal target.

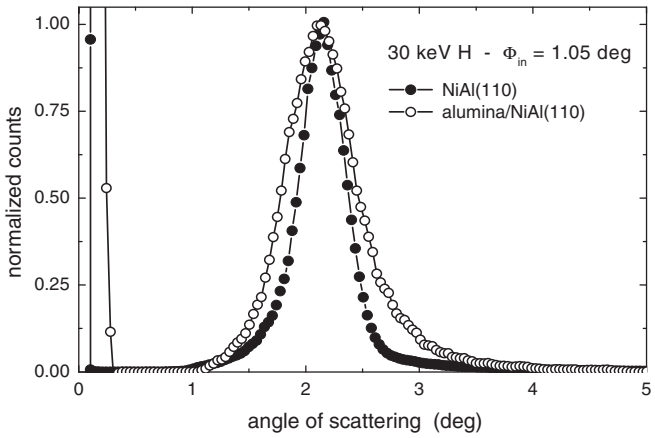

Fig. 1. Polar angular distributions for scattering of 30 keV H atoms under $\Phi_{in} = 1.05°$ from NiAl(1 1 0) (full circles) and alumina/NiAl(1 1 0) (open circles). Intense signal at left side stems from residual beam that has passed above target surface without scattering and serves as reference for angle of scattering.

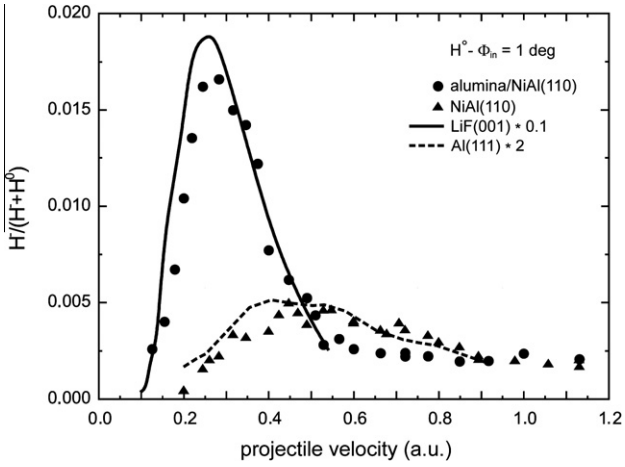

Fig. 2. Normalized negative ion fractions as function of projectile velocity for scattering of H atoms under $\Phi_{in} = 1°$ from clean NiAl(1 1 0) (triangles) and alumina/NiAl(1 1 0) (full circles). Curves represent data for LiF(0 0 1) surface divided by factor of 10 (solid curve) and for Al(1 1 1) surface multiplied by factor of 2 (dashed curve).

This is in accord with the observations for scattering from bulk crystals as indicated by the black and dashed curves which represent scaled data for scattering from a LiF(0 0 1) surface (negative ion fractions reduced by a factor of 10) [22] and an Al(1 1 1) surface (negative ion fractions enhanced by factor of 2) [23].

The data for clean metal surfaces can be described by a charge transfer model where the kinematically assisted electron capture and loss can be illustrated by a shifted Fermi sphere in momentum space [2,20,24]. Furthermore, capture and loss show exponentially decaying transition rates with the distance from the surface so that effective charge transfer is dominated by the equilibrium population at a defined distance from the surface. Since binding energies of negative hydrogen ions close to a surface of about 1 eV are much smaller than typical work functions of metal surfaces (4–5 eV), electron loss dominates charge transfer so that fractions of negative hydrogen atoms amount to some permille only.

For insulator surfaces, the band gap suppresses electron loss, but for capture valence electrons have generally higher binding energies than for metals. In scattering from ionic crystals high fractions of negative ions have been observed [22,25] which were explained by the confluence of valence band and affinity levels mediated by the Madelung interaction of the surrounding point charge lattice [10]. For the alumina film ionic bonds might play a less important role than for alkali halides which explains the larger negative ion fractions observed for LiF compared to the thin oxide film here, whereas the structure of the kinematic resonance is similar for the two insulators. Our experiments on projectile energy loss outlined below will demonstrate that the band gaps in both cases are fully developed so that the different results for bulk LiF and the thin alumina film can hardly be attributed to imperfections of the electronic structure of the thin film compared to an insulator.

The electronic excitations of the target surface are investigated via the energy loss of scattered atoms. In Fig. 3 we show energy loss spectra for 2.5 keV He atoms scattered from clean NiAl(1 1 0) under $\Phi_{in} = 2.7°$. The spectra are recorded in coincidence with the pulse heights of the biased surface barrier detector where the noise level of the detector is related to events without emission of an electron [3]. For comparison, the energy spectrum of the incident projectile beam (direct) is also given. The data shows a defined energy loss of about three percent which stem from interactions of the projectiles with the electron gas at the selvedge