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Trivalent Sm on Cu(111) at and above Room Temperature

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Introduction

Intermetallic compounds based on lanthanides have attracted much attention from both physical and chemical aspects such as magnetism, valence fluctuations, superconductivity and catalytic activities. Particularly, samarium is of particular interest due to its dramatic change of the valency (i.e. the population of 4f electrons) depending on its physical and chemical environment, which comes from the extremely small energy difference between its two electron configurations.

So far, a number of studies have been performed for the systems of Sm adsorbed on various elements. Although the growth manner of Sm adlayer on Cu(111) has been precisely studied by LEED previously[1], its electronic properties still remain unclear.

In this study, ultraviolet photoelectron spectroscopy (UPS) measurements were performed for the Sm over-layer on Cu(111) at room temperature, indicating that the electronic state of Sm adsorbed on Cu(111) is obviously different from the case of on Cu(100)[2,3]. In addition, the Sm valency of various phases formed on Cu(111) above room temperature is also presented.

Experimental

The UPS experiments were carried out with angle-resolved electron spectrometer (VG, ADES-400) at BL-7B. Samarium (purity: 99.9%) was evaporated by resistive heating with tantalum filament. Cu(111) (purity: 99.999%) was pre-polished in air and cleaned in UHV (base pressure: 4 x 10^{-10} Torr) with repeated cycles of Ar^+ sputtering and annealing up to 900 K.

Results and Discussion

The valence-band photoelectron spectra of Sm adlayer on Cu(111), around 1 ML at room temperature (Fig.1), obviously shows an absence of divalent species in the binding energy region below the Cu 3d peaks. This indicates that Sm is completely trivalent on Cu(111) surface. This result presents a striking contrast to the spectra of the Sm/Cu(100) (at RT)[3], which shows clear divalent multiplets.

Upon annealing the sample of over 1 ML coverage at 600 K and 800 K, (2 x 2) dominant and (1.46 x 1.46) dominant LEED patterns were observed, respectively. The (1.46 x 1.46) structure is assigned to be the Sm simple overlayer and the (2 x 2) structure is associated with the Sm-Cu alloy surface[1]. For both superstructures, only trivalent species was detected as shown in the spectra.

Our STM studies for Sm on Cu(111), mono-atomic-height clusters were observed on surface even from quite low coverage region, which indicates strong aggregating tendency of adsorbed Sm, while on Cu(100), no Sm induced structure was observed, which implies formation of ‘lattice-gas’ of Sm adatoms. These facts suggest the coordination number dependant valence transition of Sm; the low coordinated ‘lattice-gas’ Sm is divalent and the Sm cluster induces trivalence. It is also consistent with previous studies for Sm adsorbed on various substrates, suggesting that high coordination induces the trivalent state against the divalent one which is favored by gas-phase Sm[4].

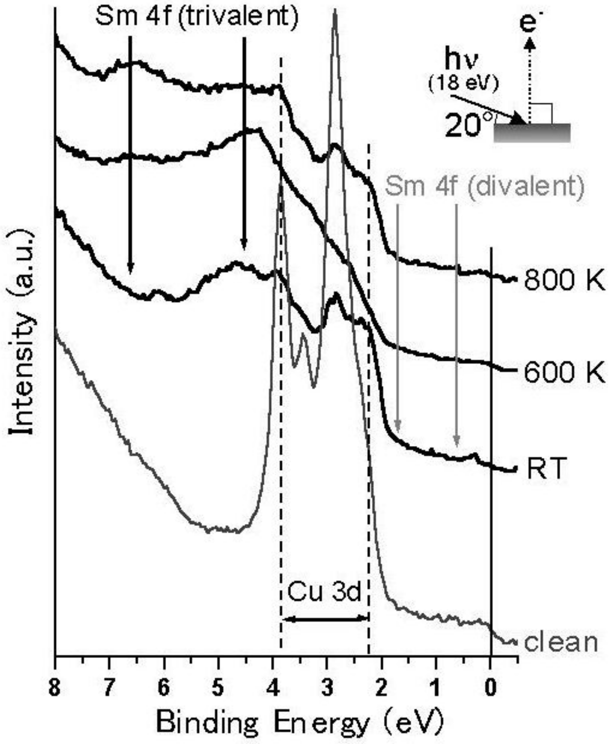

Fig.1: Valence band spectra from Sm adlayers on the Cu(111) with annealing temperatures. Photon energy = 18 eV, 20^o incidence from surface, normal emission. Arrows denote peak positions of trivalent and divalent Sm4f.

References

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