Fine tuning of spin-orbit splitting in Ag ultrathin films deposited on Au(111)

H. Cercellier[1], Y. Fagot-Revurat[1], B. Kierren[1], D. Malterre[1], D. Popovic[2], F. Reinert[2]

[1]*Laboratoire de Physique des Matériaux, UMR 7556, F-54506 Vandoeuvre-les-Nancy.*
[2]*Universität des Saarlandes, Fachrichtung 7.2-experimentalphysik, D-66041 Saarbrücken*

e-mail: fagot@lpm.u-nancy.fr

Introduction

Due to their surface localization, sp-derived Shockley states have been shown to be a unique spectroscopic probe of surface phenomena [1]. In particular, any perturbation of the surface like gas adsorption [2], lateral confinement [3], or surface reconstruction [4] should influence the free-electron like surface state parameters : band minimum, effective mass and lifetime. For exemple, by comparing STM and ARPES measurements, we have recently evidenced a significant change of the surface state energy due to the structural transition observed in the Ag/ Cu(111) interface [4-5].

The Au(111) surface band is known to present a k-dependent splitting Δk_{SO} attributed to a non zero spin-orbit interaction induced by the breakdown of inversion symmetry at the surface [6-7]. In a simple nearly free-electron theory (NFE), the spin-orbit interaction leads to spin-polarized surface bands, i.e. two electronic states with different energy and spin direction associated with the same parrallel wave vector $\mathbf{k}_{//}$. Such a surface effect has been also reported for W(110) d-type surface states [8] and each free-electron like parabola has been shown to be 100 % spin polarized [9-10] as expected theoretically [11]. Nevertheless, the NFE model fails to predict the correct experimental value of Δk_{SO} (10^{-6} times too weak) and a more realistic model should take into account the core potential so that this splitting should be proportionnal to the atomic spin-orbit coupling [12]. In this case, this effect should be stronger for heavy atoms and explain why ab-initio calculations are able to predict the correct value for Au(111), a small splitting for Ag(111) (order of the best resolution available) and a negligible one for Cu(111) surface bands [13]. Again, any perturbation of the surface should influence the spin-orbit interaction as already shown for rare gas adsorption [14]. In this contribution, we present a complete ARPES, AES and STM investigation of the growth of Ag on the Au(111) surface and its consequences on the electronic properties, focusing on the spin-orbit splitting [15].

Results and discussion

Ag deposition below room-T leads to a layer by layer growth and an abrupt Ag/Au interface. ARPES spectra obtained on Au(111) surface, 0.5 ML and 1 ML of Ag on Au(111) are presented in figure 1-a, 1-b and 1-c respectively. As already observed in the Ag/Cu(111) interface [16], two surface states are observed for coverages lower than 1 ML : one correponding to the non covered Au termination (same energy position than for free Au(111) surface state) and a second one shifted by ΔE=+170 mV toward the Fermi level corresponding to the Ag termination. In addition, the effective mass is enhanced from m^*=0.26 to $0.33m_0$ and the SOC reduced from Δk_{SO}=0.023 to 0.019 $Å^{-1}$. The band minimum, the effective mass, the Rashba parameter and the workfunction have been measured as a function of Ag coverage up to 10 ML. The spin-splitting varies slower than the workfunction and exhibits the same exponential behavior as observed for the other surface state parameters like E_0 and m^*.

Annealing the Ag film above room-T leads to the formation of an Ag-Au alloy with a well-ordered cristallographic structure and a well-defined corresponding surface state (fig. 2-a and 2-b). A detailed study of the surface state parameters as a function of annealing (up to 600 K) evidences an increase in the spin-splitting directly related to the Au concentration in the alloy (fig. 2-c and 2-d).

The exponential decrease of Δk_{SO} observed with increasing the Ag coverage and its increase observed with the Ag-Au alloy formation could be together explained by the atomic character of the spin-orbit interaction. Indeed, this splitting is directly proportionnal to the average number of Au atoms probed by the surface state wave function. We will present a quantitative analysis of ARPES and AES results : - the Au concentration profiles are calculated using a 1D diffusion model and allow us to reproduce the AES intensity curves measured as a function of annealing; - then, a simple one dimensionnal model [4, 17] is used to calculate the surface state parameters (the band minimum E_0 and the spin-splitting Δk_{SO}) both as function of the coverage and of the annealing.

Finally, a fine tuning of the surface state parameters is obtained in the Ag/Au(111) interface, including the 100 % spin polarized k-splitting ,just by adjusting the number of Au atoms probed by the Shockley state wave function at the interface.

References

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Figures

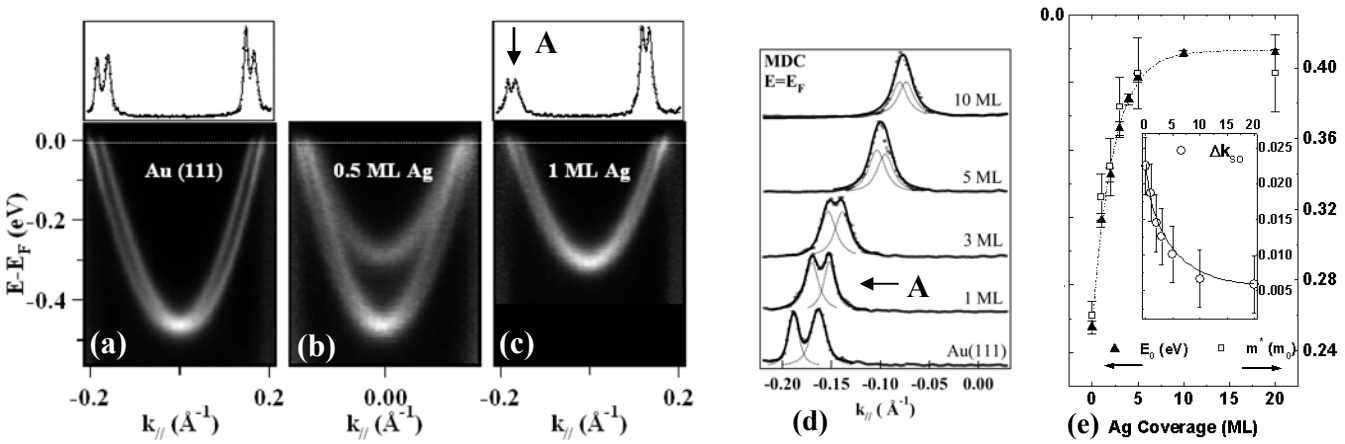

Figure 1: ARPES Intensity map I(E, $k_{//}$) measured on (a) Au(111), (b) 0.5 ML Ag/Au(111), and (c) 1 ML Ag/Au(111); (d) zoom on the momentum dispersion curves (MDC) taken at $E=E_F$ for different coverages; (e) E_0 (eV) , m* (m_0) and Δk_{SO} as a function of Ag coverage.

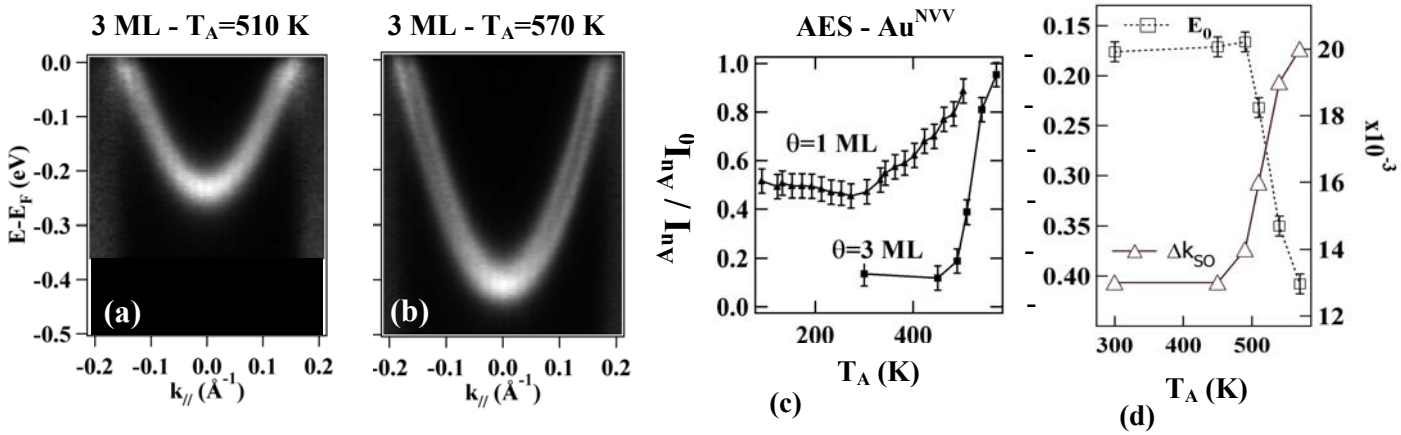

Figure 2 : (a) ARPES Intensity map I (E, $k_{//}$) measured on 3 ML Ag/Au(111) annealed at 510 K (b) and 570 K. (c) annealing effect on the Au NVV AES Au signal for 1 and 3 ML coverage. (d) E_0 (eV) and Δk_{SO} ($Å^{-1}$) as a function of annealing.