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## Co/Au multilayers: LEED and photoemission studies

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**Abstract.** The crystallization of Co and Au(111) has been shown to be possible on molybdenite substrates at 650 K as proved by good LEED patterns. The growth is, however, affected by islanding. UPS evidences a structure attributed to the Co and Au. The same kind of measurement has been performed for Au/Co/Au and Co/Au/Co/Au sandwiches with identical issues.

### 1. Introduction

A lot of work has been devoted to the epitaxy of ultra-thin magnetic materials and multilayers alternating magnetic and non-magnetic materials, owing to their peculiar properties [1]: namely perpendicular anisotropy, as is the case for Co/Au up to a critical thickness of 1.2 nm [2] or the giant magnetoresistance of Fe/Cr superlattices [3] for instance. Co/Au bilayers also show interesting magnetoresistance properties [4, 5].

The realization and study of such layers is very useful to understand the origin of ferromagnetism in 3d metal layers. For this, one can rely on the Stoner criterion in the form  $J/W \geq 1$  where  $J$  is the exchange integral and  $W$  the band width [6]. Calculations have shown that the magnetic moment onset depends critically on the band filling and on the crystallographic parameters [6]. In particular, a decrease of the hopping integral can enhance the magnetic moment [7]. In contrast, the magnetism can be destroyed when the interaction with the substrate is strong as is the case for Fe/GaAs except if a gold layer is intercalated [8].

For Co/Au, the weak interaction between Co and Au and the extensive (14%) lattice mismatch permits the prediction of an enhanced magnetic moment. Nevertheless the surface and interface conditions (roughness and interdiffusion) may interfere, so the crystallization must be well controlled.

Low energy electron diffraction (LEED) was used to control the crystallization. Ultra violet photoemission spectroscopy (UPS) monitors not only the mode of growth but also the evolution of the electronic states. Moreover, the comparison of photoemission spectra with the spin-resolved band structure calculations proved that UPS was able to evidence the magnetic exchange splitting (ES) of the bulk Co valence bands [9].

Our study was brought about by the aim to crystallize Co on Au(111) by using the epitaxy of Au on MoS<sub>2</sub> as a starting Au(111) surface [10].

### 2. Experimental details

The experimental set up is composed of a UPS chamber including LEED, combined with a preparation chamber of epitaxy.

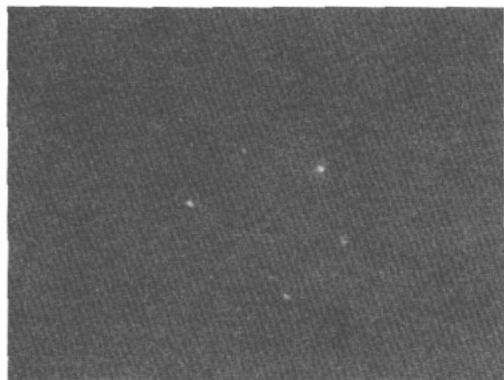
UPS uses the 21.2 eV radiation emitted by a helium discharge lamp as excitation. The photoelectrons were analysed in angle-resolved mode ( $1^\circ$ – $2^\circ$ ) along the normal to the surface of the sample using an adjustable hemispherical analyser with a resolution of about 150 meV.

The gold was deposited with a tungsten wire and Co was deposited by passing an electric current through a Co wire. The thicknesses deposited were controlled by a quartz balance. The epitaxy of Au(111) is easy on MoS<sub>2</sub>, which can be air cleaved or vacuum cleaved, the difference between the two methods being the initial size of the islands [11]. We chose the first method and deposited a 10 nm thick Au layer on a MoS<sub>2</sub> sample carefully outgassed under vacuum. The crystallization was improved by annealing at 500 K after the deposition. The Au(111) monolayer (ML) is 0.236 nm thick ( $1.4 \times 10^{15}$  atoms cm<sup>-2</sup>) while the Co(111)ML is 0.203 nm thick ( $1.8 \times 10^{15}$  atoms cm<sup>-2</sup>). For a Co ML on Au(111) the lattice expansion would be 14% (from a 0.251 nm to a 0.288 nm side of the hexagonal lattice). On the whole we successively fabricated the Co/Au, the Au/Co/Au and finally the Co/Au/Co/Au layers.

### 3. Results

#### 3.1. Co/Au(111)

The photoemission spectra presented in figure 2 were obtained from increasing deposition (0.1 nm to 0.7 nm) of Co on Au(111) maintained at 650 K for a good crystallization (figure 1). The Au(111) spectrum (10 nm thick Au deposit on MoS<sub>2</sub>) is comparable to the best ones obtained for the Au(111) face [12] exhibiting all its characteristic features, indicated in figure 2 with the notation of [12]. The labels 2.3 and 5.6 represent the 5d<sub>3/2</sub> and 5d<sub>5/2</sub> states, SS is a Shockley surface state, *D*<sub>1</sub> and *D*<sub>2</sub> are surface resonances while the origin of S seems unclear [12]. At 21.2 eV and normal analysis the emission arises from the vicinity of the  $\Gamma$  point along the  $\Lambda$  direction of the Brillouin zone [13]. The spectrum which shows a marked (111) fingerprint however, has a local character as testified by the obtaining of a similar one for non-crystallized Au/Al(111) [14]. The overall crystallinity is born out here by the good LEED patterns.



**Figure 1.** LEED patterns obtained with a beam energy of 94 eV after deposition of 0.7 nm of Co on Au(111) at 650 K.

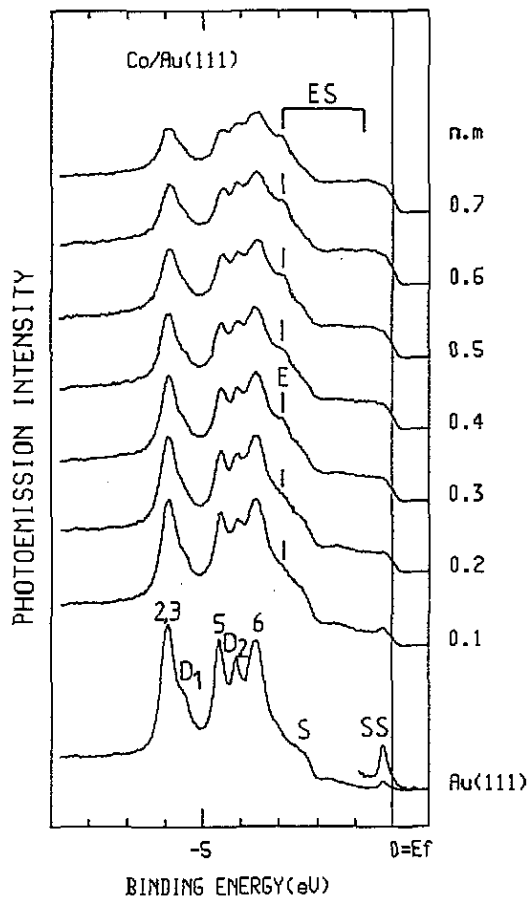


Figure 2. Evolution of the Au(111) photoemission spectra with Co deposits from 0.1 to 0.7 nm at 650 K.

Under the Co deposition the attenuation of the intensity of the Au valence bands (weaker than exponential) shows that the growth does not occur layer by layer but islanding. Recent STM measurements evidence bilayer islands that grow laterally with increasing coverage [15]. The values given for the thicknesses of the deposits are therefore only average values. SS disappears completely around 0.4 nm implying a uniform coverage at this step while the slower disappearance of the  $D_2$  resonance due to its larger spatial extent indicates a weak perturbation of the interfacial Au layers. The density of states which progressively appears below the Fermi level ( $E_F$ ) (from 0 to  $-2$  eV with a maximum around  $-0.8$  eV) arises from the Co 3d states. Likewise the feature labelled E has an unclear origin and its attribution to the ES of Co can be questioned. In the Co(0001) photoemission spectrum [9] it has been shown that the structure at  $-2$  eV (similar to the E one in our case) is separated from the principal peak at  $-0.75$  eV by the ES (1.25 eV) of the lower  $\Delta_5$  band ( $\Gamma'_{25}$  at  $\Gamma$ ). The corresponding value in our case should be  $ES = 2.1$  eV and it is right to ask whether such a high value is plausible. Recent photoemission experiments on Co/Cu(100) [16] evidence an  $ES = 1.95$  eV for a Co ML (as seen in the difference curve) and this was moreover strongly supported by spin-resolved photoemission measurements beyond the ML coverage. Calculations by the same authors [16] give the following values for the ES of Co:

1.55 eV for the bulk, 1.76 eV for a Co ML on Cu(100) and 1.95 eV for an unsupported Co ML with the Cu(100) lattice parameter.

A larger ES value can be expected for Co/Au(111) compared to Co/Cu(100) due to the effect of the substrate, for instance through to the extension of the lattice parameter. For this last point to occur the growth must be pseudomorphic. In fact, as the thickness increases a gradual relaxation has been evidenced for Co/Au(111) and the residual mismatch would be 5% for 1 nm as measured by RHEED [4]. Values of strain which aimed to be more representative of the final relaxed structures were carried out by x-ray scattering measurements [19] and gave an in-plane extension of 1.75% for a 1 nm thick Co layer on Au(111) while the value for Co/Cu(100) was  $-\frac{1}{2}\%$  (compressive). With regard to the coordination number, increasing its value reduces the magnetic moment [6, 7, 16]. Thus magnetic moment calculations [18] for unsupported ML of 3d metals put in evidence an enhancement of  $0.32 \mu_B$  for a (100)Co ML and only  $0.15 \mu_B$  for a (111)Co ML with respect to the calculated bulk value of  $1.69 \mu_B$ .

Although the magnetic moment is proportional both to the exchange splitting and to the density of states at the Fermi level it is interesting to examine the results of some magnetic moment calculations for 3d metal overlayers on Ag [20]: for an unsupported Co ML a  $2.2 \mu_B$  value has been obtained which reduces to 2.03 for an overlayer, i.e. when the interaction with the substrate is taken into account. For a Co overlayer on Pd a  $2.12 \mu_B$  value is obtained [20–22]. In this case, it has been argued that the 3d–3d and the 3d–sp hybridization can be reduced by structural effects like lattice mismatch so that the moment become insensitive to environment and can approach the atomic-like value [21]. In both cases Co/Ag [20] and Co/Pd [22] the ES is 2 eV from the local density of states for ferromagnetic calculations.

Finally, an enhancement of the ES up to 2.1 eV could be plausible for Co/Au(111). However in our case, the E feature which appears in the ML range cannot be attributed unambiguously to the ES of Co since it remains at a fixed binding energy with increasing coverage instead of decreasing towards the bulk value, as we have checked for thicknesses beyond those of figure 2. The attribution of the E feature to quantum states (from Au 5d states falling in the gap between 3d and 4s states of Co for instance) is also difficult to verify due to the lack of variation with the thickness [23, 24]. We have also ascertained that the E feature does not appear for layers grown at room temperature. So a plausible explanation of this feature can be found in an interaction between Co and Au due to intermixing. For alloys there is a shift of the  $5d_{5/2}$  state of Au away from the Fermi level on alloying (dilution effect) [25–27] which is not observed here, but there is also an interaction between the Au 5d states and the d states of the host which play a subtle but crucial role in the electronic structure of dilute Au alloys [27]. Two-dimensional interfacial alloying has been put in evidence (even with components which do not form alloys in the bulk [28, 29]) and form interfacial states arising from the interaction of the d states of the adsorbate with the d states of the substrate. Our case is more complex due to the 3D mode of growth and even if the exact nature of the interaction is not known (probably between Au  $5d_{5/2}$  state and Co 3d states) our results indicate nevertheless an interdiffusion between Co and Au which is favoured by the fact that the deposits are made at 650 K.

### 3.2. Au(4.5 nm)/Co(0.7 nm)/Au(111)

The study of the deposit of Au on Co/Au(111) and so on is interesting for the realization of metallic multilayers. Figure 3 gives the photoemission spectra obtained for Au deposits (after annealing at 500 K) from 0.25 nm to 4.5 nm. From the increase of the intensity of the d bands, the mode of growth is seen here also to proceed by islanding. The E structure is strongly attenuated at 0.25 nm and disappears around 0.5 nm. The final spectrum looks

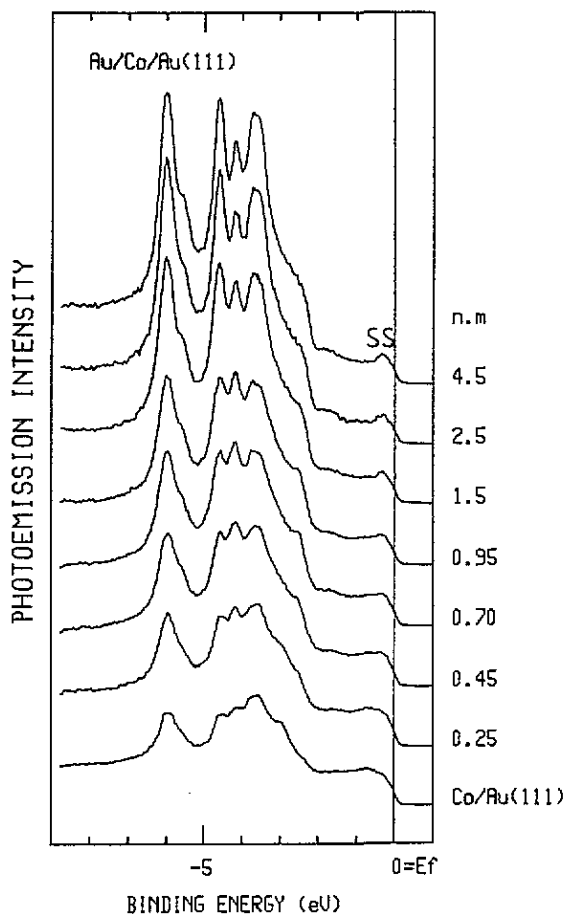


Figure 3. Evolution of the Co(0.7 nm)/Au(111) spectrum with Au deposits annealed at 500 K from 0.25 to 4.5 nm.

like that of Au(111) but with a smeared surface state perhaps due to defects. The good LEED pattern similar to that of figure 1 certifies the overall crystallization, and we find this encouraging.

### 3.3. Co(0.7 nm)/Au(4.5 nm)/Co(0.7 nm)/Au(111)

The photoemission spectra for the Co deposit are in the same sequence and the same temperature as previously done for Co/Au(111). The growth still proceeds by islanding and the evolution of the spectra is qualitatively the same as in figure 3. The E structure appears at the same thickness and at the same binding energy but with a weakened intensity. A LEED pattern similar to that of figure 1 is still present; there is however from the beginning of the deposits a regularly enhanced background.

## 4. Conclusion

We have shown the possibility of crystallization for Co on Au(111) using MoS<sub>2</sub> as a substrate, and also for alternated multilayers. However the thicknesses of the individual

layers are difficult to control due to islanding. We have evidenced an extra feature on the valence bands which can be ascribed to an interdiffusion between Au and Co enhanced by the temperature of deposits (650 K). This interdiffusion can have important consequences for the transport properties of magnetic multilayers.

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