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Growth of antiferromagnetically ordered Cr monolayer on Ag(100)

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ABSTRACT

Two-dimensional antiferromagnetism observed on flat Cr monolayers deposited on Ag(100) has been investigated under various growth conditions. Cr monolayer domains are found to grow within the temperature range of 373–453 K while multilayer growth mode is prevalent at room temperature (RT). In agreement with theoretical predictions, a $c(2 \times 2)$ antiferromagnetic configuration is observed on Cr monolayer and is confirmed by the presence of magnetic superstructural spots in Low Energy Electron Diffraction (LEED) and antiferromagnetic Cr 3*d* bands in Angle-resolved Photoemission Spectroscopy (ARPES) studies. Antiferromagnetism of Cr monolayer film is found to depend strongly on various growth parameters such as growth temperature, annealing temperature, annealing duration, rate of growth *etc.* and are studied in detail here. The phenomenon of Cr–Ag intermixing at the interface is found to be the main factor for the complex growth of Cr on Ag(100) substrate. Optimum parameters for the growth of large monolayer Cr domains on Ag(100) substrate are presented here.

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1. Introduction

Two-dimensional (2D) magnetism of ultrathin transition metal films has drawn the attention of many earlier research works because of their exciting magnetic properties at reduced dimensions [1–9]. The Ising-like magnetic behaviors of these films have made them fascinating due to the presence of long range 2D magnetic order that exists at finite temperature contradicting the Heisenberg model. According to total energy calculations of Blugel et al. [5,6], transition metal Fe, Co and Ni favor ferromagnetic $p(1 \times 1)$ configuration whereas V, Mn and Cr favors antiferromagnetic $c(2 \times 2)$ configuration as overlayers on top of Pd(100). In spite of the interesting physics, access to the surface antiferromagnetism has been limited due to the experimental difficulties in detecting them. For example in case of Cr, the reduced coordination at surface affects its magnetic moment strongly due to its half-filled 3*d* orbital. A significant enhancement of the magnetic moment is observed at the surface of bulk Cr [7,8] as well as in Cr films of sub-monolayer thickness [9–11].

The order and strength of 2D magnetism in a monolayer Cr film strongly depends on its physical structure. When deposited on a noble metal substrate, the magnetic structure is governed by different effects like film thickness, interfacial strain due to lattice mismatch, intermixing/alloying at film–substrate interface *etc.* The electronic structure of the film is expected to be influenced by the hybridization between transition metal *d*-bands and noble metal states. However, it is found that the 2D electronic property of a free monolayer is essentially retained when the monolayer is deposited onto the noble metal substrate as there is very little overlap (hybridization) between the noble metal *d* bands and Cr *d* states. Growth of Cr has been studied on different noble metal substrates with Cr forming interfacial alloy while deposited on Au(100) at room temperature [12–14] while no alloying is reported in case of Cr film deposited on Ag(100) [15,16] or on Cu(100) [10,17,18]. Growth of Cr on Ag(100) is interesting as it is a model system with very good lattice matching and almost no bulk miscibility between Cr and Ag above their melting points. However, growth of Cr on Ag(100) surface is dominated by interplay between the surface free energy and substrate temperature which controls the mobility of the overlayer Cr atoms. As Cr has a surface free energy (2400 mJ/m^2) almost double of Ag (1250 mJ/m^2), growth of Cr on Ag(100) is expected to result in 3D island formation as per Bauer's criterion [19] to lower the surface free energy. Thus for room temperature (RT) deposition, Cr atoms occupy second layer (bi-layer) sites before completion of the first layer [14,15] as expected. Hence multilayer domains are formed at the sub-monolayer thickness range. Scanning Tunneling Microscopy (STM) measurements [20,21] have confirmed growth of 3D Cr islands up to three layer height under deposition at RT exposing significant amount of uncovered Ag surface.

Interestingly, it was observed by Krembel et al. [22] that it was possible to grow flat monolayer of Cr on Ag(100) when deposited at

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higher substrate temperatures (430–450 K). Further, they also observed a weak but distinct $c(2 \times 2)$ superstructure spots attributed to the 2D antiferromagnetic order in the LEED pattern, which was a significant step towards experimentally studying 2D antiferromagnetism. Even though many further experiments were performed by the group of G. Gewinner, attempts by other groups to observe the magnetic $c(2 \times 2)$ superstructure spots were not successful. STM studies on the growth of Cr sub-monolayers at elevated temperatures have observed [20,21] Ag segregation/agglomeration, however have failed to observe the magnetic $c(2 \times 2)$ superstructure LEED spots and concluded that flat Cr film on Ag(100) is never formed. Monolayer growth of Cr was noted from the Inverse Photoemission studies [23] with enhanced magnetic moments, however failed to observe the $c(2 \times 2)$ LEED superstructure spots. An *in situ* surface X-ray diffraction study by Steadman et al. [24] has shown the sensitivity of Cr growth on Ag(100) at different temperatures. They observed the growth of flat Cr monolayer on Ag(100) at higher substrate temperatures, however could not confirm their magnetic ordering.

Here, we have undertaken a detailed growth study of Cr monolayer on Ag(100) substrate at elevated temperatures and confirm their antiferromagnetic ordering. Detailed knowledge of different parameters of the growth conditions is very important in obtaining reproducible experimental results and for better understanding the properties of the grown film. Thus, we have carried out the Cr monolayer growth at different substrate temperatures, direct/step-wise growth conditions, different growth rates and at different sample/substrate annealing temperatures which are *in situ* characterized using LEED and photoemission studies. We have optimized the best growth conditions for obtaining flat Cr monolayer on Ag(100) with highest degree of antiferromagnetic ordering along with large domain sizes. Different aspects of the growth, formation of $c(2 \times 2)$ monolayer domains, their structural stability with temperature and possible interfacial alloy formation are discussed in detail.

2. Experimental details

Well polished single crystal Ag(100) was treated by repeated cycles of Ar ion sputtering (600 eV, 1 μA) for 15 min followed by annealing at 823 K for 30 min to make the surface crystalline quality better until a sharp LEED pattern is observed with $p(1 \times 1)$ symmetry. Cr was evaporated in the preparation chamber from a high purity Cr rod (99.95%) kept in an electron beam evaporator at a rate of 0.2 Å/min calibrated by a water cooled quartz thickness monitor. Chamber pressure was maintained to below 2×10^{-10} mbar during Cr evaporation. Cr films were grown on top of Ag(100) crystal kept at various temperatures from RT to 473 K and the substrate temperature was brought down to RT immediately after Cr deposition is over. LEED measurements were performed using a four-grid LEED apparatus (OCI Vacuum Microengineering) to determine the crystalline quality of the deposited film as well as crystallographic symmetry directions. A high-sensitive 12-bit camera was used to collect the LEED images with a software controlling the gain/exposure settings of the camera as well as all the other controls of the LEED instrument.

Angle-resolved Photoemission Spectroscopy (ARPES) and X-ray Photoemission Spectroscopy (XPS) measurements were performed in the analysis chamber with base pressure better than 8×10^{-11} mbar and attached to the preparation chamber. ARPES measurements were performed using a combination of VG SCIENTA-R4000WAL electron energy analyzer with 2D-CCD detector and a high flux GAMMADATA VUV He lamp attached with a VUV monochromator, which has been described in detail elsewhere [25]. We used He I_α (21.218 eV) resonance line to excite the photoelectrons from sample surface for the ARPES measurements while Al K_α monochromatic X-ray source (1486.6 eV) from VG Scienta is used for XPS measurements (not shown here). All ARPES and XPS measurements were performed at RT with total experimental energy resolutions better than 0.1 eV and 0.65 eV, respectively. Thickness of the deposited Cr films was estimated by the XPS intensity measurements and were in good agreement with the quartz crystal thickness monitor calibrations. Temperature dependent LEED measurements were performed while warming up the sample after cooling down to 100 K using liquid nitrogen. The experimental results were repeated many times and have confirmed the reproducibility of all the results.

3. Results and discussions

The bcc lattice parameter of Cr is 2.88 Å whereas the fcc Ag(100) substrate has a lattice parameter of its non-primitive unit cell 4.09 Å which is about $\sqrt{2}$ times the lattice parameter of Cr. Therefore Cr grows on Ag(100) pseudomorphically with a crystallographic rotation of 45° with 0.2% lattice mismatch. Still due to the higher surface free energy of overlayers, a multilayer/cluster growth occurs at RT reducing the effective surface area of Cr film. However at elevated temperatures (430–450 K), the deposited Cr atoms attain sufficient mobility to spread over the Ag(100) surface leading to the formation of large terraces of flat Cr monolayer [26]. In agreement with the total energy calculation of Blugel et al. [5], the flat Cr monolayer on Ag(100) shows $c(2 \times 2)$ antiferromagnetic order as observed by previous studies [15] and confirmed by our LEED studies here. For even higher temperature growth, agglomeration of the Cr atoms takes place as observed by STM measurements [21].

Fig. 1(a) and (b) shows the LEED pattern of 1 ML Cr film deposited at RT with primary electron energy (E_p) of 40 eV and 23 eV, respectively. The first-order LEED spots [1,0] are distinctly visible in Fig. 1(a) and the corresponding first surface brillouin zone (SBZ) is depicted over the LEED pattern indicating the symmetry points. The LEED pattern of 1 ML Cr film deposited at 428 K is shown in Fig. 1(c) at 40 eV E_p where faint $c(2 \times 2)$ superstructure spots are observed (marked by arrows). These extra spots with weak intensity have coordinates [1/2, 1/2] and are called half-integer or half-order spots. Intensity of these half-order spots get enhanced at lower beam energies and reaches a maximum at 23 eV as shown in Fig. 1(d), though at this beam energy the integer-order spots does not enter the field-of-view of the LEED screen. These half-order spots are faintly visible up to about 60 eV above which they are not detectable with our setup. From Fig. 1 it is clear that only films grown at 428 K show the four-fold $c(2 \times 2)$ half-order spots while the RT grown films does not show any such half-order spots. Intensities of these half-order spots are only about 2% of the first-order integer spots at the respective energies of their intensity maximum (*i.e.* at 23 and 55 eV) and normalized to the respective electron beam current at room temperature. These low intensity half-order spots could only be detected using the high-sensitive peltier-cooled CCD camera attached to the LEED instrument. We have performed the temperature dependence of these half-order spots and observe that the intensity decreases with increasing sample temperature. In Fig. 1(e), we show the line profiles drawn along the boxed region of LEED spots in Fig. 1(d) for different sample temperatures (RT, 380 K and 460 K) where the intensity is found to vanish completely above 455 K. These half-order spots are similar to the observations of Hanf et al. [22,27] for Cr/Ag(100) system as well as to the case of NiO [28,29] where it is well established that such half-order spots originate due to the magnetic exchange interactions between the incident low energy electrons and the antiferromagnetically ordered surface Cr/Ni

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