The Structural Determination of Au(1ML)/Fe(15ML)/Au(100) Using Angle-Resolved Photoemission Extended Fine Structure

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A great amount of attention has been given to investigating thin magnetic films and magnetic multilayers, especially systems involving iron and the noble metals[1-8]. In most of these studies the authors assume that the Fe layers will maintain bulk spacing even at interfaces. However, it is well known that for the clean metal the first and second layer spacing is contracted from the bulk value, and that absorbates can significantly expand this spacing[9-20]. Atomic structural details about these interfaces is important because the electronic states that are localized at the interface between the two different materials are critical in determining the magnetic properties of ultra-thin films and multilayers[21, 22]

In this study we use Angle-Resolved Photoemission Extended Fine Structure (ARPEFS) to investigate thin (*ca.* 10 and 15 monolayers) Fe films grown on a Au(100) single crystal. ARPEFS is a well established technique for determining the atomic structure of atomic and molecular adsorbates on metal surfaces[18, 19, 24-27]. The technique's advantages are its atomic selectivity due to the unique binding energies of core level electrons, the large oscillations, which in this study are ± 40%, and its inherent accuracy. In the past, structural determinations have only been done with ARPEFS signals from initial states with zero angular momentum because of the difficulties in treating non-s initial states in the scattering calculations. This study presents the first structure determination of a bimetalic system using the ARPEFS from non-s initial states. We report results from a new computer simulation and fitting procedure based on the Rehr and Albers formalism[28]. This program, developed by our group, uses second-order matrices (6x6) and up to eighth-order scattering to produce a convergent calculation at these

electron energies and inter-atomic distances[29]. The experiment was performed at the Advanced Light Source (ALS) on the bend magnet beamline 9.3.2, which covers the photon energy range of 30 eV to 1500 eV. The system studied and reported here is for 15 monolayers (ML) of Fe grown on the Au(100) single crystal. Breaks in the slope of the Fe 3p photoemission intensity *vs.* evaporation time curves confirm the layer-by-layer growth reported by other authors (Fig. 1). Comparison of the Fe and Au photoemission intensities as well as sputtering studies confirm that the Fe layers grow with one ML of Au atoms, acting as a surfactant, on the top of the growing Fe layers.
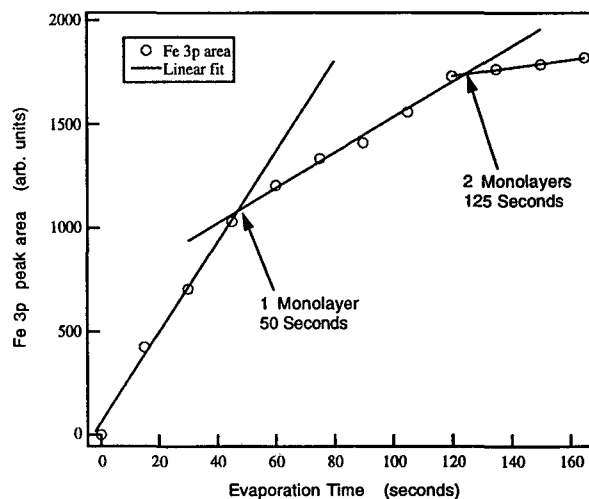

Figure 1. The Fe 3p photoemission intensity plotted against the Fe evaporation time. The breaks in the slope of the curve indicate layer-by-layer growth. One ML of Au atoms, acting as a surfactant, remains on top of the growing Fe layers. The LEED pattern is 1x1

We collected the ARPEFS intensity modulations, I(k), from the Au $4f_{7/2}$ core level along the normal and the 45° off-normal emission directions. This function I(k) has two components; a

slowly varying, atomic-like portion, $I_0(k)$, upon which is superimposed a rapidly oscillating beat pattern that arises from the interference between the primary wave propagating directly to the detector and waves which scatter elastically off nearby atomic potentials before reaching the detector. diffraction curves, $\chi(k)$ are obtained by removing the slowly varying $I_0(k)$ portion from $I(k)$.

$$\chi(k) = \frac{I(k) - I_0(k)}{I_0(k)}$$
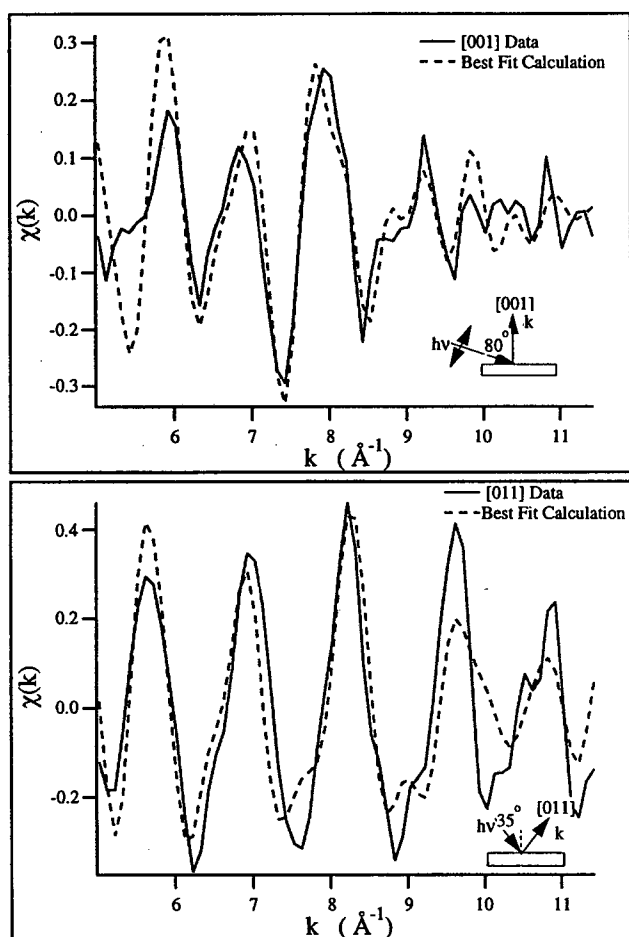
Figure 2. shows the experiemntal $\chi(k)$ curves.

The auto-regressive linear-prediction based Fourier transform (ARLP-FT), shown in Fig. 3, transforms the diffraction data from momentum space to real space[30]. In ARPEFS, the positions of the strong backscattering peaks in ARLP-FTs from adsorbate/substrate systems can be predicted with very good accuracy using the single-scattering cluster model together with the concept of strong backscattering from atoms located within a cone around 180° from the emission direction. The ARLP-FT peaks correspond to path-length differences between that component of the wave which propagates directly to the detector and those components which are first elasticly scattered by the atomic potentials. Analysis of the ARLP-FT provides information about the adsorption site as well as the bonding distance of the gold atoms. Figure 3 shows the ARLP-FT for Fe/Au(100)
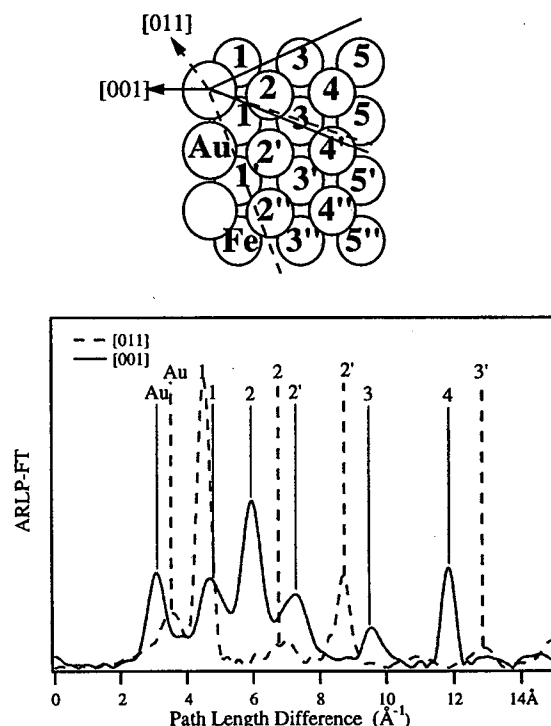

Figure 2. The Au $4f_{7/2}$ ARPEFS data from Au(1ML)/ Fe(15ML)/Au(100) collected along the [001] and [011] emission directions. Schematics of the experimental geometry are shown. Dashed lines are the best fit multiple scattering calculation results. The largest-amplitude oscillations in each curve arise from strong backscattering off the nearest-neighbor Fe atoms in the [001] and [011] directions, respectively. See Fourier transforms in Fig. 3.

Figure 3. ARLP-FTs of the ARPEFS [001] data (solid line) and the [011] data (dashed line). A model of the lattice with the backscattering cones for each emission direction indicates the scattering atoms coresponding to the FT peaks. Note the excellent agreement between peak positions and calculated values.

The observed peaks in the ARLP-FT are most consistant with a structural model where the Au atoms sit in the four-fold hollow site, 1.6 Å above the surface plane of the bulk Fe. The ARLP-FT shows peaks corresponding to scattering events from as far away as the fourth iron layer.The fact that we see such long PLD is an indication of the qualitiy of the Fe films and the sharpness of the Fe-Au interface. The very good agreement between the predicted and the observed peaks in the ARLP-FT and the presence of sharp ARLP-FT peaks due to scattering from the fourth Fe layer, shows the Fe lattice to be very like the Fe bulk.

Fitting the experimental diffraction curves to a multiple-scattering model yields more precise structural parameters than that given by the Fourier analysis alone. Chen *et al.* recently developed a new multiple-scattering code, based on the Rehr-Albers formalism, which can model initial states with arbitrary angular momentum and which is fast enough to allow practical fitting to be done[28, 29].This calculation requires both structural and non-structural parameters. We used the structural parameters determined by the Fourier analysis as the initial guesses in the fitting procedure. The non-structural parameters include the initial-state angular momentum, the atomic scattering phase-shifts, the crystal temperature, the inelastic mean free path, the emission and light polarization directions, the electron analyzer acceptance angle, and the inner potential.

The best fits determine the Au-Fe1 spacing to be 1.67 Å, and the Fe1-Fe2, and the Fe2-Fe3 spacing to be that of the bulk iron, 1.43 Å, within the experimental error limits. It is interesting to note that this value is a slight expansion of interlayer spacings realatiave to the clean metal[31].

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