Comparison between Calculated and Measured Photoelectron Diffraction Patterns for Cu (001)

G.P. Cousland[a], A.E. Smith[a], J.Riley[b], S.Homolya[c], A. Stampfl[d], J. King-Lacroix[d]

[a] *School of Physics, Monash University, Clayton, Victoria 3800.*
[b] *Department of Physics, La Trobe University, Bundoora, Victoria 3086.*
[c] *CSIRO Division of Materials and Manufacturing Technology, Clayton South, Victoria 3169.*
[d] *Bragg Institute, Australian Nuclear Science and Technology Organisation, Menai, NSW 2234.*

We compare the results of X-ray photoelectron diffraction experiments with simulations obtained using the EDAC multiple scattering computer simulation package. Comparisons are presented for Cu (111) at photon energies of ~ 600eV. With an intention to study Cu_3Mn, our initial work considers experimental and simulated data for Cu (001) at photon energies from 100 to 380eV.

1. Introduction

The detailed knowledge of surface structure and induced structural changes due to adsorbates, impurities and preparation techniques is important for the understanding of crystalline growth in basic science and technology applications [1]. We are investigating the interfaces of antiferromagnetic or ferromagnetic material with spin glasses beginning with Cu_3Mn. X-ray photoelectron diffraction (XPD) [2] has been developed as a way of determining surface, near-surface and interfacial structural information [3,4]. This paper presents initial results for Cu (001) using low energy X-ray photoelectron diffraction from a synchrotron source. Previous laboratory-based experiments have been carried out for this surface but with much higher photon energies and employing only a simple analysis [5].

Using synchrotron sources, lower photon energies can be obtained [6], and in the range below ~ 300eV forward focusing is far less apparent and the angular momentum of the emitted photoelectron cannot be ignored in any theoretical modeling undertaken [7]. This paper compares XPD experimental results within this low photon energy range with those obtained by the Electron Diffraction in Atomic Clusters (EDAC) computer simulation package [8]. Our intention is to extend the present visual comparisons and refine prediction of the structural and chemical properties by using a suitable R-factor analysis.

2. Simulation software

The EDAC program is a Multiple Scattering (MS) real-space simulation package that can be used to simulate Low Energy Electron Diffraction (LEED), Auger Electron Diffraction (AED) and X-ray Photoelectron Diffraction (XPD) experiments [8]. Multiple scattering algorithms are needed compared with their single scattering counterparts in order to accurately simulate experimental interference patterns [9]. EDAC can be obtained as an executable program that can be run on Linux (32 or 64 bit) or Microsoft (32 bit) platforms. There is scope for including phenomenological parameters such as the inelastic mean free path of the photoelectron and the inner potential of the material. A number of run-time variables, including atomic phase shifts and approximate muffin tin potentials, are calculated within the program.

The operator is given scope to include features that help replicate the experimental setup being simulated, including the orientation of the sample relative to the incident beam and how, and through what angles the sample is moved. The direction and polarisation of the incident beam as well as the energy and spin of the emitted photoelectron can be determined by setting arguments to pre-determined commands that are input to the program at runtime.

EDAC is implemented in C++ and the program runs as a single thread. It thus runs the scan associated with one input file on only one processor during runtime. The output of a photoelectron diffraction experiment requires emitting atoms on a number of consecutive levels into the surface, and the program runs more economically when there is only one emitting atom per input file. This means that the most efficient way to proceed is to calculate output data for a number of levels simultaneously on different computer processors. So far we have been able to run the simulations on a current model Intel dual processor computer, but it is our intention to run the work on a cluster system.

Once created, the data is combined linearly and can be displayed as a diffractogram, a scan for fixed polar (θ) or azimuthal (φ) angle, or an energy scan at a fixed angle. Most of this work shows simulations of 2π scans where $0^\circ < \theta < 89^\circ$ and $0^\circ < \varphi < 360^\circ$.

3. Results

Initial experiments were conducted on Cu (111) at photon energies of 600eV and above to compare with simulated results using EDAC. The scans are produced by a toroidal electron spectrometer [10] designed and built by the Centre for Materials and Surface Science (CMSS) at the La Trobe University Department of Physics, and now installed and operational at the Berlin synchrotron facility BESSY II. It allows the *simultaneous* analysis of the kinetic energy of electrons leaving the sample at all polar angles. This dramatically reduces measurement time in comparison to a conventional angle resolved photoelectron spectrometer, which can only measure photoelectrons for one combination polar and azimuth angles at a time. The CMSS spectrometer allows for the rotation of the sample for obtaining data at different azimuth angles. By combining a set of scans taken at different azimuth angles, a full hemisphere scan can be constructed. The resulting diffractogram is shown in Fig 1.

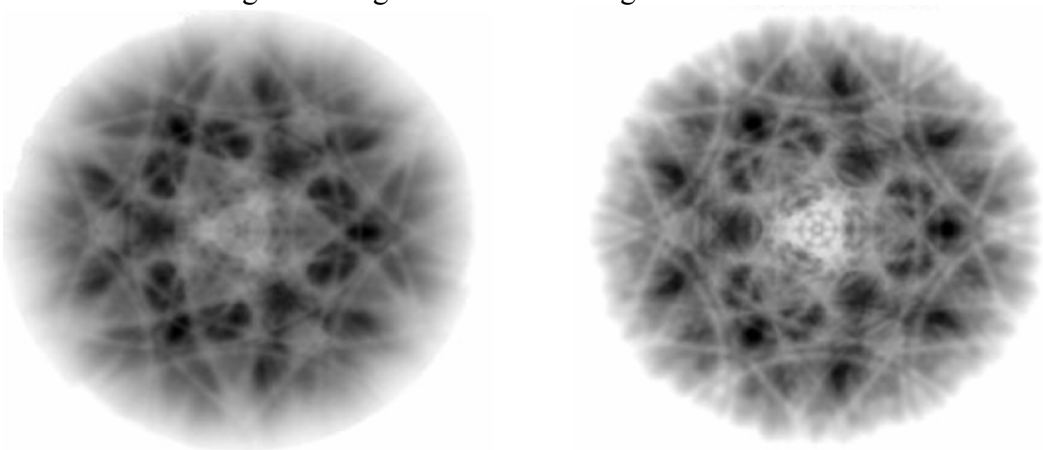

Fig 1 Stereographically projected image of Cu (111) data for an incident photon energy of 600eV for experimental (left) and simulated (right) diffractograms. Note the bright peaks indicating forward focusing along low index axes and Kikuchi bands indicating Bragg diffraction of the photoelectron wavefunction from planes of atoms.

Our group intends to investigate magnetic exchange properties at the interface of antiferromagnetic and ferromagnetic materials with spin glass surfaces. The initial material chosen is Cu_3Mn, where it is envisaged that we will acquire azimuth-scanned synchrotron X-ray photoelectron diffraction measurements using the Cu and Mn 3p core levels from a Cu_3Mn (001) surface. Owing to the growth properties of the CuMn alloy [11], as a starting point we are looking at Cu (001) surfaces. We have carried out photoelectron diffraction simulations using EDAC and have completed low energy 2π scans of Cu and Cu_3Mn and a Mn layer on a Cu substrate.

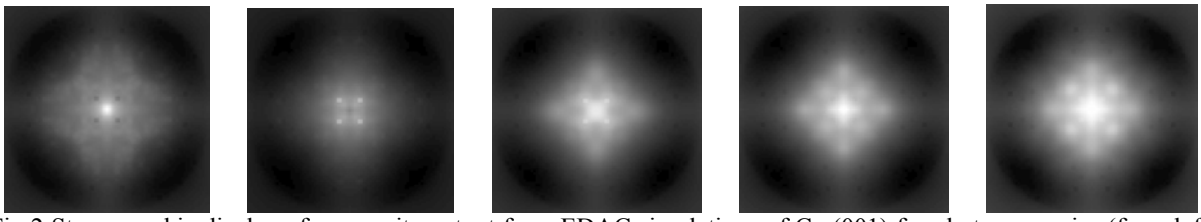

Fig 2 Stereographic display of composite output from EDAC simulations of Cu (001) for photon energies (from left to right) of 100, 150, 180, 280 and 380eV. The images demonstrate forward focusing at photon energies of 280eV and above. Note the minimum in the [001] direction for the 2π diffractogram at a photon energy of 150eV.

Some preliminary simulations using EDAC are shown in Fig 2. At low photon energies there is not the same degree of forward focusing that there is at higher energies. This is evidenced by the bright intensities for photon energies of 280eV and above. We find it interesting that at a photon energy of 150eV, there is little forward focusing along the [001] direction. In single scattering simulations such effects have been explained by destructive interference between the forward focusing [001] amplitude and some higher-angle scattering amplitude [12]. An alternative explanation is the character of the source wave as in Auger Electron Diffraction [7].

Fig 3 is a superimposed azimuthal scan at a polar angle of 45º for simulated data and a scan of Cu(001) taken at the Taiwan synchrotron. The simulated result has rotational symmetry but not full mirror symmetry about the principle axis. This can be explained as a polarization effect of the X-rays with respect to the face centred atoms on the (100) and (010) faces of the unit cell.

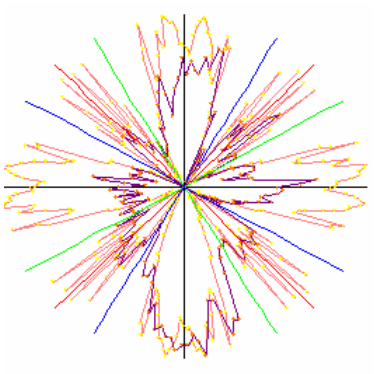

Fig 3: Initial result showing an azimuthal scan of Cu (001) for a photon energy of 150eV and a polar angle of 45º. The scan is a comparison of experimental results taken at the Taiwan synchrotron (in purple) compared with simulation results using EDAC (yellow). The 4-fold symmetry of the crystal can be noted from the simulation. This corresponds to forward focusing along the principle axes.

Fig 3 shows only some correlation in the comparison for azimuthal scans between preliminary experimental data and simulated results. This is even though considerable work went into processing the experimental data, including background subtraction, before this plot was displayed.

4. Conclusion

We have simulated X-ray photoelectron diffraction in the low energy range using EDAC, which has previously been demonstrated for high energy simulations to be efficient multiple scattering simulation software. Although some of the experimental data does not correlate well with the simulation results, this might be attributable to problems with the data acquisition or the normalisation process.

Considerable variations in the simulated intensity in the forward direction have been observed with changes of energy for scans taken at very low photon energies, 100 to 380 eV range for Cu (001). This would be somewhat unexpected within the multiple scattering model at higher energies, where forward focusing is a prominent feature, but at lower energies it seems that diffraction is more sensitive to the nature of the photoelectron.

Our group is investigating physical structure and chemical properties of the surfaces of materials, including alloys and substrates with overlayers. Our intention is to study the properties at the interface of antiferromagnetic or ferromagnetic material and a spin glass. The material chosen for this is Cu_3Mn where site element specificity will be determined by XPD. Owing to the growth properties of this alloy we are initially studying Cu (001) using low energy XPD from a synchrotron source. Multiple scattering XPD simulations have shown large variations in the forwarded scattered intensity with incident energy. As experimental data becomes available we will employ R-factor analysis so as to further refine our structural and chemical information.

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