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Publication Date
1987-10-01
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October 1987
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FORWARD FOCUSING OF DIFFRACTED AND EMITTED ELECTRONS AS A SURFACE STRUCTURAL TOOL

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Electrons with medium and high energies, \( E' \geq 500 \text{eV} \), undergo marked forward focusing by atoms in their path, analogous to optical focusing by lenses. This effect can be used to determine interatomic directions in surfaces and from there the surface structure. We have modeled the situation with full multiple-scattering calculations and successfully compared them to a variety of experimental data.

Techniques in which forward focusing has been observed experimentally include: angle-resolved x-ray photoelectron emission spectroscopy (ARXPS)\(^1\), Auger electron spectroscopy (ARAES)\(^{1a,2,3}\), and inelastic "Kikuchi-like" electron emission\(^{2,4}\). Marked focusing is observed as an enhancement along interatomic axes pointing out of the crystal, allowing the direct determination of surface structure. The above experiments were mainly carried out for metal surfaces, some of which include layers of foreign atoms (e.g. Cu layers in or on Ni(100)\(^1\) or O layers in or on Mg(0001)\(^4\)), while some experiments concern semiconductor surfaces\(^2\).

Our theoretical approach builds upon the "Taylor series, magnetic quantum number expansion" (TS-MQNE)\(^5\) theory of angle-resolved photoelectron emission fine structure (ARPEFS). This method provides an efficient treatment of electron-atom scattering. It is especially efficient at higher energies, relative to conventional LEED-style calculations. It includes all multiple scattering to convergence within a cluster of converged size; as such it carries the name "near-field expansion in clusters" (NFEC)\(^6\). The theory also includes specific methods to model electron scattering by linear chains of atoms\(^7\).

Some previous theoretical modeling of forward focusing effects only included single scattering of emitted electrons\(^{1c,2a,2c,2d,e,8a}\), while the importance of multiple scattering has been pointed out\(^8b\). We have now shown that the single-scattering approximation already includes most major features of the multiple-scattering result in a qualitative way. But multiple scattering is far from negligible and affects both peak positions and heights, through which surface structure is determined. We also have verified that additional non-focusing peaks occur: they are secondary.
interference maxima away from focusing directions. Model calculations are necessary to identify these and to avoid interpreting them as representing interatomic directions.

The focusing peak directions are affected by changes in the direction of interatomic axes, so that, for instance, interlayer spacings can be determined. In our experience, atomic positions could be determined to somewhat better than 0.1Å by this approach, at least if one only uses visual comparison of theoretical and experimental curves.

We have investigated three variants of this method, differing in the origin of the electrons. First, we considered medium-energy electron diffraction (MEED) in the 1000eV regime. For an ordered surface, one obtains only Bragg beams on a detecting screen (as in any LEED pattern from an ordered surface). This in general prevents detecting focusing peaks along interatomic directions, since they do not usually coincide with diffracted beam directions. However, with disordered surfaces a "screenfull" of diffuse MEED intensities can be obtained at any fixed setting of crystal, source and detector. In our calculations for this situation, we found marked focusing effects, but they are mixed with strong interference effects due to the coherence of the incident electron beam. These interference effects make diffuse MEED less attractive for structural determination than the applications described next.

In angle-resolved Auger and inelastic "Kikuchi" electron emission, each emission event is incoherent with respect to the others and may be assumed to take place at one atomic site, just as with XPS. By inelastic "Kikuchi" emission, we mean electrons that have undergone an energy loss, to a phonon for example, and thereby lost memory of their direction of propagation; the outgoing inelastic electrons then undergo elastic scattering that has some analogy with "Kikuchi" effects at high energies.

The "Kikuchi" variant of this technique appears to yield very similar experimental results as the Auger variant. We have therefore treated both identically in the theory. In particular, we assume in both cases that the
electrons start out as isotropic s-waves leaving atomic sites. The s-wave assumption was prompted by the experimental findings and is not to be regarded as definitive: other partial-wave components are undoubtedly present, but would tend to affect the overall envelope of the results rather than the individual peak positions. For Auger electrons, there is in addition chemical sensitivity; i.e., the emission process can be limited to one atomic element.

Egelhoff\textsuperscript{1a} has measured angle-resolved Auger electron emission from a single Cu layer buried at a variable depth within a Ni(100) surface. The Cu Auger electrons travel out through the overlying Ni layers and are focused to an extent that varies with the thickness of the Ni layer. Our calculations have reproduced these experimental data very satisfactorily and can, for instance, discriminate between different depths at which the Cu layer is buried. In addition, it appears that uneven depths have to be assumed; i.e., the overlying Ni layer may not be perfectly flat.

An analogous experiment by Egelhoff\textsuperscript{1a} considers Cu Auger electrons from a Cu layer of variable thickness deposited on Ni(100). Again, good agreement is found between our corresponding calculations and the data.

Auger and inelastic "Kikuchi" experiments were performed by Hilferink et al.\textsuperscript{3} for pure Ni(100). The results of the two kinds of measurement are mutually very similar, as noted above. Here again, we obtain quite good agreement between theory and experiment, further supporting the assumption of s-wave emission for both Auger and inelastic "Kikuchi" emission.

Finally, we have analyzed Auger data by Cronacher and Müller\textsuperscript{4}, taken for oxygen on Mg(0001). Here O Auger emission was measured after exposure of clean Mg(0001) to oxygen. Our calculations considered several adsorption and absorption sites (as did the experimental group with single-scattering calculations\textsuperscript{9}). Based on our comparisons, the most favored oxygen site appears to be an octahedral interstitial position between the 2nd and 3rd metal layers (with some expansion of the metal-metal layers spacing). However, we cannot exclude the octahedral interstitial site between the 1st and 2nd layers and the hollow sites on top of the surface (in each position,
very acceptable metal-oxygen distances are found). It is possible that all of
these sites are simultaneously populated, but better experimental data would
be needed to resolve this issue.

In conclusion, it appears that forward focusing is an effective tool in
surface structure determination, especially with the use of XPS, Auger and
even inelastic "Kikuchi" electrons of medium to high energy. Effective use of
this approach has already been exhibited by Egelhoff\(^1\) and Chambers et al.\(^2\) in
elegant studies. It is not clear whether atomic locations could be determined
with a higher accuracy than with LEED, for example. But it remains possible
that excellent accuracy could be attained with full multiple-scattering
calculations and non-visual theory-experiment comparisons as afforded by
R-factors. The most obvious strength of forward focusing is in determining
the site of surface atoms, in a way reminiscent of Rutherford backscattering:
one can relatively easily distinguish between different adsorption,
interstitial and substitutional sites.

Acknowledgments

This work was supported in part by the Army Research Office, and in
part by the Director, Office of Energy Research, Office of Basic Energy
Sciences, Materials Sciences Division of the US Department of Energy under
Contract No. DE-AC03-76SF00093. One of us (J.J.Barton) gratefully
acknowledges an IBM Postdoctoral Fellowship.
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