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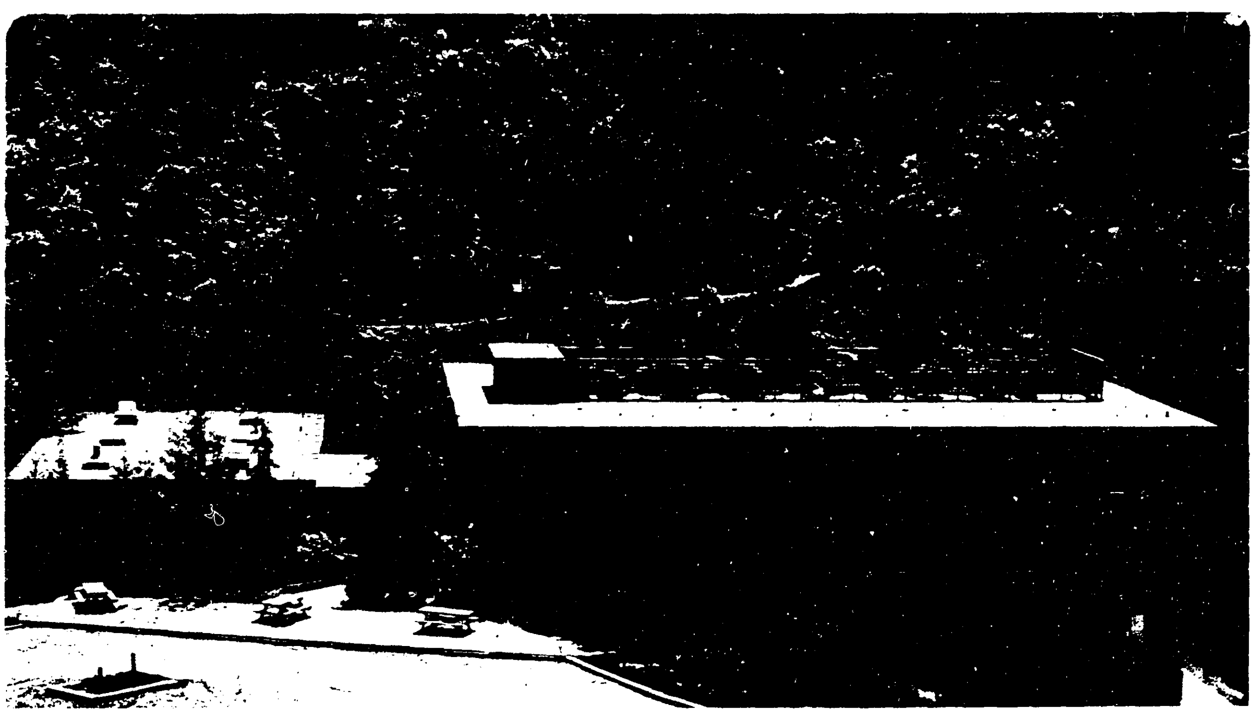

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PHOTOELECTRON DIFFRACTION AND HOLOGRAPHY: SOME NEW DIRECTIONS+

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ABSTRACT

Photoelectron diffraction has by now become a versatile and powerful technique for studying surface structures, with special capabilities for resolving chemical and magnetic states of atoms and deriving direct structural information from both forward scattering along bond directions and back-scattering path length differences. Further fitting experiment to theory can lead to structural accuracies in the ±0.03 Å range. Holographic inversions of such diffraction data also show considerable promise for deriving local three-dimensional structures around a given emitter with accuracies of ±0.2-0.3 Å. Resolving the photoelectron spin in some way and using circularly polarized radiation for excitation provide added dimensions for the study of magnetic systems and chiral experimental geometries. Synchrotron radiation with the highest brightness and energy resolution, as well as variable polarization, is crucial to the full exploitation of these techniques.

INTRODUCTION

Photoelectrons emitted from core levels represent localized sources of outgoing waves which can then scatter from nearby atoms to produce diffraction patterns. We will here consider several new directions for using such diffraction patterns to determine surface atomic positions, as well as surface magnetic structures [1-5]. The analysis of such data in a more recently suggested holographic manner so as to directly image atoms in three dimensions [6,7] will also be considered. The special benefits that synchrotron radiation brings to such studies will also be pointed out.

PHOTOELECTRON DIFFRACTION--BASIC CONCEPTS

Photoelectron diffraction patterns are by now well known and much studied, and have lead to the increasing use of this technique for surface structure studies [1-5]. The fundamental measurement is illustrated in Fig. 1. A photoelectron is emitted from a core level, and its intensity is measured as a function of its direction or its energy above a single-crystal sample, yielding what can be termed scanned-angle or scanned-energy data, respectively. In terms of the electron wave vector $\underline{k}$, this is equivalent to measuring intensity as a function of its direction $\hat{k} = \underline{k}/|\underline{k}|$ or its magnitude $k = |\underline{k}|$. Intensity variations are produced by the interference of the unscattered or direct wave component ϕ_o and the various scattered-wave components ϕ_j.

over initial and final magnetic quantum numbers and interference between the two final-state channels l_{final} = l+1 and l-1 that are allowed by the dipole selection rules [8a,8d,9].

Expanding the square in Eq. 3a now yields

$$I(\underline{k}) \propto |F_o|^2 + \Sigma_j[F_o^*F_j\exp\{-i\underline{k}\cdot\underline{r}_j\} + F_oF_j^*\exp\{i\underline{k}\cdot\underline{r}_j\}]$$
$$+ \Sigma_j\Sigma_k[F_j^*F_k\exp\{i\underline{k}\cdot(\underline{r}_j-\underline{r}_k)\} + F_jF_k^*\exp\{-i\underline{k}\cdot(\underline{r}_j-\underline{r}_k)\}] \quad . \qquad (4)$$

$|F_o|^2$ is thus simply proportional to $I_o(\underline{k})$: the intensity in the absence of any scattering. A normalized intensity function $\chi(\underline{k})$ can now be calculated, very much as in the analysis of extended x-ray absorption fine structure (EXAFS), with one choice being [7a]:

$$\chi(\underline{k}) = [I(\underline{k}) - I_o(\underline{k})]/I_o(\underline{k})^{1/2} \quad , \qquad (5)$$

and this yields

$$\chi(\underline{k}) \propto (|F_o|)^{-1}\Sigma_j[F_o(\underline{k})^*F_j(\underline{k})\exp\{-i\underline{k}\cdot\underline{r}_j\} + F_o(\underline{k})F_j(\underline{k})^*\exp\{i\underline{k}\cdot\underline{r}_j\}]$$
$$+(|F_o|)^{-1}\Sigma_j\Sigma_k[F_j(\underline{k})^*F_k(\underline{k})\exp\{i\underline{k}\cdot(\underline{r}_j-\underline{r}_k)\} + F_j(\underline{k})F_k(\underline{k})^*\exp\{-i\underline{k}\cdot(\underline{r}_j-\underline{r}_k)\}], \qquad (6)$$

where the r_j or r_k dependence of F_j or F_k, respectively, in spherical-wave scattering have not been indicated explicitly. This form is useful in considering holographic analyses of diffraction.

Another common approximation is to assume that the scattered waves ϕ_j and ϕ_k are small in amplitude with respect to ϕ_o, so that the cross terms $\phi_o^*\phi_j$ and $\phi_o\phi_j^*$ in Eq. 1 dominate the structural information. This directly leads via Eqs. 3 and 6 to

$$\chi(\underline{k}) \propto 2\Sigma_j(\hat{\varepsilon}\cdot\hat{r}_j/r_j)|f_j(\theta_j)|W_j\exp(-L_j/2\Lambda_e)$$
$$\cdot\cos[kr_j(1-\cos\theta_j)+\Psi_j(\theta_j,r_j)] \quad . \qquad (7)$$

This form directly shows that Fourier transforms of scanned-energy data along some direction $\hat{k}$ and over some interval Δk

$$F_{\Delta k}(\hat{k},r) \propto \int_{\Delta k} \chi(\underline{k})\exp[-ikr]dk \quad , \qquad (8)$$

should be useful for deriving path length differences $r = r_j(1-\cos\theta_j)$, a result that has been discussed and used in a number of previous studies [4,10].

There are several important characteristics of such photoelectron diffraction patterns, as summarized below. More detailed discussions with illustrative examples appear elsewhere [1-5].

-**Measurement of intensities**: In general, core peak intensities must be two interfering channels of 1 ± 1. Thus, theoretical modeling can be the most accurate for photoelectron diffraction and holography. Varying both the polarization and energy of the exciting photon also can be used to emphasize different scatterers or aspects of the emission or scattering process.

-Simple forward scattering: In measurements at photoelectron kinetic energies of about 500 eV or higher, the scattering amplitude $|f_j(\theta_j, r_j)|$ is highly peaked in the forward direction (i.e. near $\theta_j = 0$). Many studies have by now shown that such forward scattering or forward focussing peaks can be directly used to determine bond directions for adsorbed molecules [1,3] and low-index directions for single crystals and epitaxial overlayers [1,2]. As an illustration of the sensitivity of such high-energy patterns to different surface structures, Fig. 2 shows the full 2π intensity distributions above three different surfaces, in stereographic projection: fcc Ni(001) [20], hcp Ru(0001) [21], and the textured surface of highly oriented pyrolytic graphite with a preferred (0001) orientation [22]. Such forward scattering peaks have also been found to be sensitive to surface pre-melting phenomena [23]. The higher kinetic energies required for this kind of measurement have led to its being performed primarily with laboratory x-ray sources in the 1.2-1.5 keV range, but higher brightness synchrotron radiation sources in the 500-1500 eV range would be equally useful for this work.

-Back scattering: In measurements at lower photoelectron kinetic energies of less than about 300 eV, there is also a significant degree of back scattering, and this can be used in several ways to extract structural information concerning atoms that are "behind" the emitter as viewed by the detector [1,4,5,10,24]. Synchrotron radiation is again necessary to insure sufficiently low kinetic energies in such studies.

- Single scattering and multiple scattering analysis: In a number of prior studies, it has been found that a simple single scattering model such as that outlined above is able to predict most of the structure in diffraction patterns, and thus it also can be useful for deriving some structural information. However, multiple scattering effects can be strong in both forward scattering along high-density rows of atoms (where events of order up to the number of scatterers between emitter and scatterer may have to be considered [8d]) and back scattering at lower energies (where events up to third order are found to be essential for predicting all diffraction features [8d,e]). This is illustrated in Fig. 3, where experimental and calculated full diffraction patterns above a Ni(001) surface are shown [20]. The experimental pattern away from low-index directions is reasonably well predicted by single scattering theory, but both the intensity and width of the low-index forward scattering peaks are much overestimated in this simple model. Multiple scattering theory by contrast predicts all aspects of the diffraction pattern very well, even though only five emitter layers were included in this simulation.

-Path-length differences: Another direct form of structural information that can be obtained by virtue of the strong single scattering character occurring near the origin. A total number of data points of about 1000 is thus needed for such a structure estimate. A more rigorously derivable method for summing such Fourier transforms of scanned-energy data so as to derive atomic positions in three dimensions will be discussed under photoelectron holography below.

-<u>Accurate surface structures</u>: In a growing number of studies to date, it has been possible also to determine more detailed surface structures by fitting experimental diffraction patterns of either the scanned-angle or scanned-energy type to theoretical simulations for various possible trial geometries [1,2a,4,5,8b,24]. Direct structural information from forward scattering or back-scattering path length differences can often be used to eliminate various possible structures and arrive at a very good guess for the final trial-and-error search. Theoretical calculations have been carried out at both the single scattering [1,2a] and more accurate multiple scattering [1,4,5,8] levels. With careful analysis of such fits, e.g., via R factors, accuracies in the approximately ± 0.03Å range have been obtained. However, further work is needed to speed up such structure searches and the multiple scattering calculations needed for the highest ultimate accuracy. Finally, more rapid data acquisition methods are also called for; these will benefit from next-generation higher-brightness synchrotron sources as well.

PHOTOELECTRON HOLOGRAPHY

More recently, it has been suggested by Szöke [6] that such photoelectron diffraction patterns can be treated as holograms, with the unscattered wave ϕ_o being identified as the reference wave of the hologram, and the scattered waves ϕ_j being identified as the object waves. A diffraction pattern that is somehow measured over a relatively large number of points in $\underline{k}$ space which may involve varying both direction and energy is then converted into a direct three-dimensional image of the atoms surrounding a given atom using a Fourier-transform-like integral. The hologram is in this interpretation just the intensity $I(\underline{k})$, or more conveniently the normalized function $\chi(\underline{k})$. The holographic analysis of diffraction data is in a much more developmental stage, but several encouraging experimental studies have been carried out to date [12,26-31].

The first holographic imaging procedure to be demonstrated quantitatively is due to Barton [7a]. It makes use of scanned-angle data at a single energy, for which the Helmholtz-Kirchoff theorem from optics is used to calculate the atomic image $U(\underline{r})$ (actually the source wavefield) from:

$$U(x,y,z) \propto \left| \iint_S \chi(\underline{k}) \exp[i\underline{k}\cdot\underline{r}] d\sigma_k \right| , \qquad (9)$$

where the integral on the <u>direction</u> of $\underline{k}$ is over the spherical surface on which the hologram is measured. Note that $\chi(\underline{k})$ has here been multiplied by the complex conjugate of the direction-dependent part of the phase factor due to path length difference $\exp[-i\underline{k}\cdot\underline{r}]$, and that the generalized scattered-wave strength F_j during the integration, which yields a new image function U':

$$U'(x,y,z) \propto \left| \iint \{\chi(\underline{k})\exp[ik_z z]/F_j(\underline{k},\underline{r})\}\exp[i(k_x x + k_y y)]dk_x dk_y \right| . \quad (11)$$

This has been termed the scattered-wave-included Fourier transform (SWIFT) method. In practice, this procedure has to date generally involved simply dividing by a plane-wave or spherical-wave scattering factor, which may then have to be adjusted with position in space so as to allow for the different types of scatterers present [27a-c,34]. The latter adjustment thus requires some advance knowledge of the structure, or an iterative approach. F_j also can in principle allow for the anisotropy in the outgoing reference wave, as is implicit in the factor $\hat{\varepsilon}\cdot\hat{r}_j$ in Eq. 3c; this more general type of correction has been applied for the first time to experimental data from $CoSi_2(111)$ by Zhou et al. [28].

The overlap of real and twin images is a problem shared with optical holography, but it is potentially more serious in images of surface structures, since the surface inherently breaks the inversion symmetry along its normal, and thus the twins of substrate atoms may overlap the regions in space occupied by adsorbate or overlayer atoms. One solution to this problem is to note that, for some cases, the region of the hologram most strongly affected by some atom at $\underline{r}$ is well localized in a solid-angle region centered on $\underline{r}$; this was first demonstrated in theoretical simulations by Saldin et al. [35]. Analyzing only this portion of the hologram then may lead to an image in which the twin from another atom at $-\underline{r}$ is suppressed, as suggested by Saiki et al. for scanned-angle data from cases dominated by forward scattering [36]. For back-scattering cases at lower energies, Tong et al. [34b] have also proposed analyzing scanned-energy data over only small windows in direction in order to emphasize a single scatterer behind the emitter.

In Fig. 5, we show the effects of simultaneously using these last two image improvement procedures, again for the case of c(2x2)S/Ni(001) [27c]. Only the right half of the hologram has been analyzed to focus on the position of the nearest neighbor along +x, and the SWIFT procedure has been applied in doing the image formation. The agreement between experiment and theory is again excellent, and the peak positions have improved to within about 0.3 Å of the known structure. This example thus suggests that even single-energy holographic images for adsorbate overlayers or thin epitaxial layers can be obtained with sufficient accuracy to be used for ruling out many possible structures and providing excellent starting points for more accurate final trial-and-error refinements. Other single-energy, SWIFT-corrected results for bulk $CoSi_2$ at 700 eV are also encouraging [28]. However, previous studies on multilayer bulk specimens of Cu [26a], Si [27a], and Ni [20] at higher single energies suggest that the presence of inequivalent emitters in several layers can lead to strong image distortions along forward scattering directions.

interatomic distances that are to be studied. The behavior of these artifacts is illustrated in Fig. 7, where images in the xy plane of c(2x2)S/Ni(001) are shown for different numbers of energies spanning the range from 862 to 1324 eV. Only the right half of the hologram has been analyzed (as in Fig. 5) to emphasize the real image due to the nearest-neighbor along +x. In going from 1 to 3 to 5 to 7 to 13 energies, we see a gradual suppression of twin-related features in the left half of the image, as expected. But anomalous features remain in circles at multiples of $\pi/\delta k$ away from the origin and these are fully moved out of the region of interest only in the last panel with 13 energies. Thus, such criteria on the choice of δk are crucial if image artifacts are to be suppressed.

Tong and co-workers [37] have also proposed a similar holographic approach for analyzing scanned-energy data so as to simultaneously correct f‹ · scattered-wave anisotropies and eliminate twin and multiple-scattering effects. This method makes use of a number of scanned-energy diffraction curves that are then Fourier transformed, summed, and used to determine the real-image positions of certain atoms. What is being done in this procedure is to Fourier transform a $\chi(\underline{k}_q)$ obtained along the direction $\hat{k}_q$ over small steps in k_q first and then to carry out a phased sum over several larger steps in direction, as shown below:

$$U''(x,y,z) \propto \left|\Sigma_q \exp[i\underline{k}_q\cdot\underline{r}]\int_{\Delta k_q}\chi(\underline{k}_q)\exp[-ik_q r]dk_q\right| . \qquad (13)$$

Corrections for scattering amplitudes and/or phase shifts can also be included in this integral, in the same spirit as indicated in Eq. 12. Encouraging atomic images have been obtained using this approach for ($\sqrt{3}$ x $\sqrt{3}$)Al on Si(111) by Wu et al. [30]. This equation is similar to Eq. 8b, except that the sum on directions now has a phase that is more clearly related to a three-dimensional holographic transform.

Comparison of Eqs. 12a and 13 makes it clear that the approaches of Barton and Tong are fundamentally equivalent, in that they just interchange the order of integration and summation, with the same overall phase factor of $\exp[-ikr]\exp[i\underline{k}\cdot\underline{r}] = \exp[-ikr(1-\cos\theta)]$. However, the first emphasizes finer steps in $\hat{k}$ and the other finer steps in k. Thus, if both are carried out over equivalent ranges of Δk_x, Δk_y, and Δk_z, one would expect corresponding resolutions in the coordinates x, y, and z, provided that the $\underline{k}$ steps are sufficiently small in all directions to avoid spurious features due to the non-cancellation of twin and multiple scattering features (cf. Fig. 7). If applied correctly, both methods should be equally capable of suppressing twin and multiple scattering effects. For a given image accuracy and scope in $\underline{r}$ space, it is also expected that these two approaches would require about the same size of data set: something like 3,000-5,000 intensities with allowance for surface symmetry.

In fact, these two methods of summing/integrating over intensities are really just the two limits of a continuous range of choices in sampling a given volume of $\underline{k}$ space, as illustrated in Fig. 8 [38]. Here are shown the holographic images for a simple pyramidal cluster of Cu polarized radiation can cause preferential excitation of spin-up or spin-down electrons, even if there were equal populations of the two types in the initial spin-orbit-split core states. In either case, the degree of dichroic asymmetry can be measured as a function of $\underline{k}$ via

$$A^{CD}(\underline{k}) = [I^{RCP}(\underline{k}) - I^{LCP}(\underline{k})]/[I^{RCP}(\underline{k}) + I^{LCP}(\underline{k})] \quad , \qquad (14)$$

where I^{RCP} and I^{LCP} are the intensities measured with right and left polarized light, respectively. Very few measurements of the $\underline{k}$ dependence of A^{CD} in core-level emission have been made to date, but we illustrate the types of effects expected with two examples.

Bansmann and co-workers [39a] have studied normal CD in C 1s emission from CO adsorbed on Pd(111) in a chiral experimental geometry. Some of their experimental data as a function of electron emission angle θ are shown in Fig. 9, together with theoretical calculations based on several models. The effects are quite pronounced, being as large as $\pm$75% variations in A^{CD}. The three theoretical curves all agree reasonably well with the data: two are based upon treating an isolated CO molecule only, and one includes the effect of the Pd substrate. Two of these curves (------ and - --- -) have been calculated by Westphal et al. [39b] in a multiple-scattering diffraction picture of the outgoing wave, thus emphasizing the fact that <u>it is only through photoelectron scattering and diffraction from neighboring atoms that normal circular dichroism can manifest itself in core-level emission</u>. Diffraction theory including the effects of the underlying Pd atoms (- --- -) shows that the substrate could produce noticeable effects on such data, especially at lower energies for which back scattering is more important, but there are as yet no conclusive experimental data indicating such effects. The future measurement of circular dichroism in core emission with synchrotron radiation from insertion devices designed to produce high-brightness circularly-polarized radiation, coupled with analysis in terms of more accurate cluster-based multiple-scattering calculations [8b-d], thus represents a very interesting new direction of study in photoelectron diffraction.

MCD in core-level emission has so far been studied only for a few cases, and then only with a fixed emission direction. In Fig. 10, we show the first data of this type due to Baumgarten et al. [40a] for Fe $2p_{1/2,3/2}$ emission from Fe(110). In the lower part of (a) are shown two partial spectra obtained with the sample magnetization parallel to- and anti parallel to- the direction of helicity of circularly-polarized radiation; this is equivalent to changing from right to left polarization in the frame of the sample. The full spectrum in (a) represents an average over the two magnetizations. In (b), A^{CD} is plotted, and it is clear that significant effects of the order of a few percent are seen, even if they are much smaller than those found for normal CD in Fig. 9 Similar results have been obtained by Waddill et al. [40b] for Fe 2p emission from thin overlayers of Fe on Cu(001), again for a fixed direction of emission. Both sets of data have been qualitatively explained in terms of preferential excitation of photoelectrons of one spin or another in the $2p_{1/2}$ and $2p_{3/2}$ peaks, which is simply a difference of two normal images, and

$$\Delta'(x,y,z,\uparrow-\downarrow,\uparrow) = |F_\sigma(x,y,z,\uparrow,\uparrow) - F_\sigma(x,y,z,\downarrow,\uparrow)|, \tag{16}$$

in which F_σ is the (complex) Fourier transform integral within U and the absolute value is taken after calculating the difference. The second spin argument here is the orientation of the scatterer, here chosen to be up. Through its sign, Δ can be shown to be sensitive to the orientation of the scatterer, whereas the always-positive Δ' can be shown to measure more directly the strength of the spin-dependent exchange scattering.

In Fig. 11, the image functions Δ and Δ' are plotted for the two different orientations of the scatterer: spin-up in parts (a.2)-(a.4) and spin-down in parts (b.2)-(b.4). The effects seen here are 10-15% of the magnitude of the peaks in the direct U images, and thus should be measurable, especially from higher-quality experimental data obtained with a next-generation synchrotron radiation source. As expected from their definitions, Δ and Δ' exhibit different behavior on flipping the scatterer spin: Δ changes in sign, whereas Δ' does not. Thus, it has been suggested that the locations of near-neighbor magnetic scatterers could be determined via Δ', and actual spin flips (e.g., as temperature is changed) could be detected via Δ [42]. In parts (c.1)-(c.4) and (d.1)-(d.4), the effect of adding a non-magnetic O^{2-} scatterer midway between the two Mn^{2+} ions, with the scatterer spin being down, is considered. Although the normal image function U shows a strong additional peak due to the non-magnetic scatterer, this peak is strongly suppressed in Δ', verifying that the latter should be useful for imaging only the magnetic scatterers in a system.

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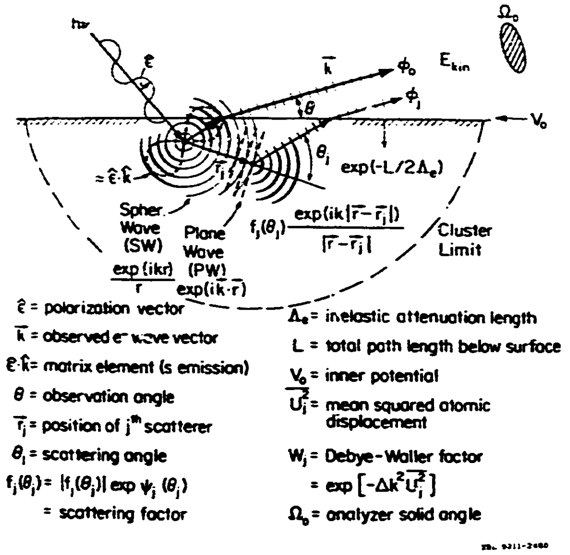
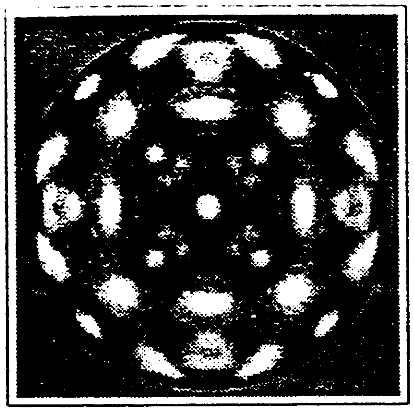
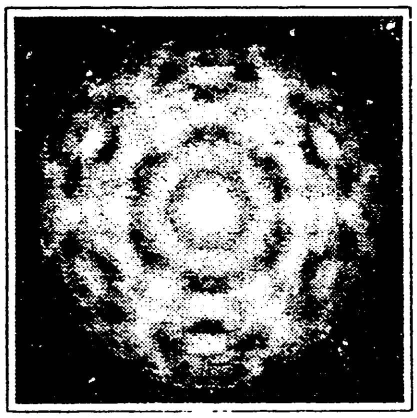


Fig. 1- The basic process involved in photoelectron diffraction, with important physical variables indicated. Only single scattering is indicated for simplicity. In a holographic interpretation of such measurements, the direct or unscattered wave ϕ_o is identified with the reference wave, and the scattered waves ϕ_j are identified with object(subject) waves.

Ni(001):Ni 2p at 636 eV



Ru(0001):Ru 3d at 1206 eV



Graphite (0001): C 1s at 946 eV

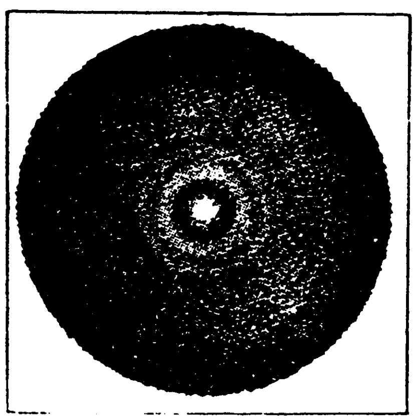


Fig. 2- Full 2π intensity distributions for higher-energy emission from several surfaces: fcc Ni(001) [ref. 20], hcp Ru(0001) [ref. 21], and highly oriented pyrolytic graphite in textured growth with preferred (0001) orientation [ref. 22].

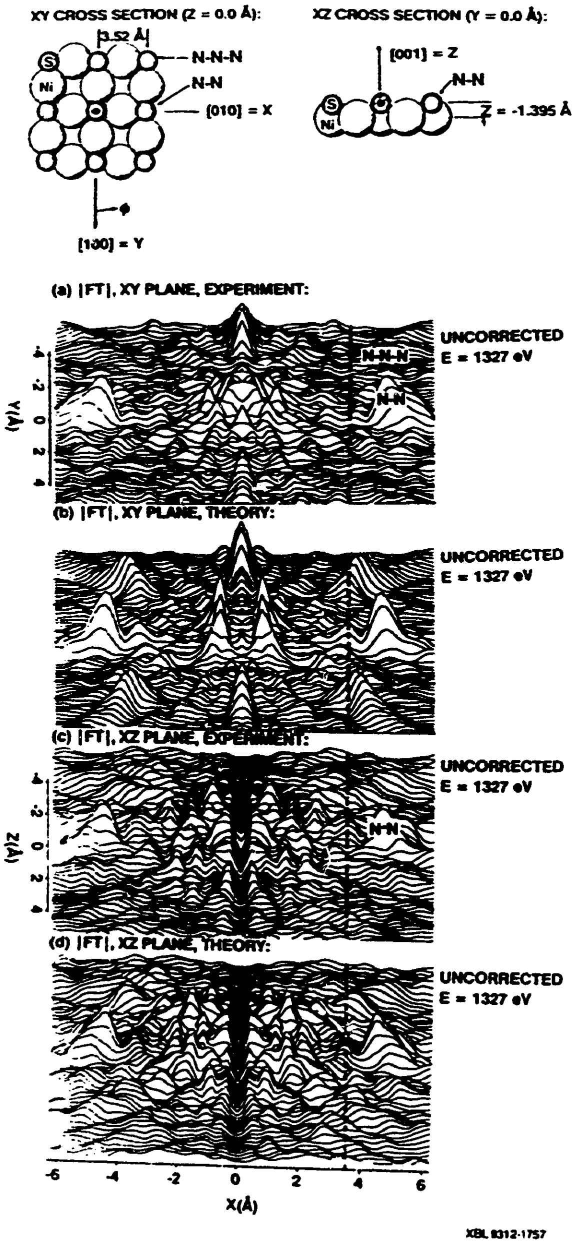


Fig. 4- The geometry of c(2x2)S/Ni(001) is shown together with Fourier transform holographic images from Eq. 10, as based upon S 2p emission at 1327 eV. The hologram analyzed has cylindrical symmetry about the z axis, and extends from 10° to 50° above the surface. Images are shown in both the xy (=sulfur) and xz planes. No scattered-wave correction has been made, and results are shown for both experiment ((a) and (c)) and single-scattering theory ((b) and (d)). The positions of nearest-neighbor (N-N) and next-nearest-neighbor (N-N-N) S atoms are indicated. The vertical dashed line indicates the known positions of these atoms. [From Thevuthasan et al. ref. 27c]

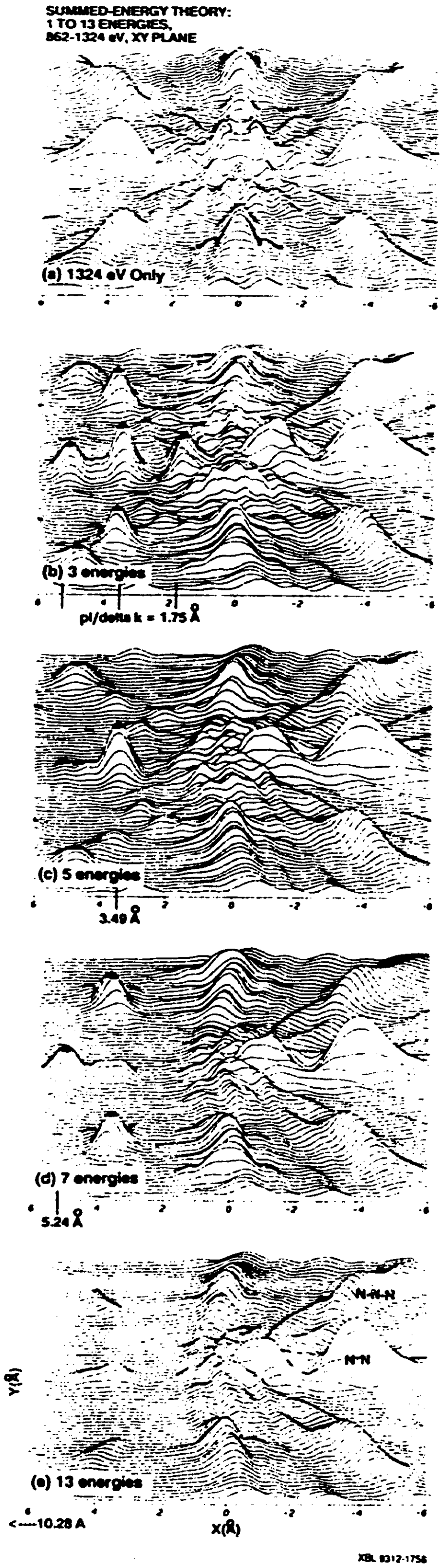


Fig. 7- Theoretical Fourier transform images for c(2x2)S/Ni(001) in the S plane obtained using only the right half of a hologram extending from 10° to 50° above the surface (as in Figs. 4 and 5). Data are shown for different numbers of energies in a phased sum according to Eq. 12b, but with no scattered-wave correction: (a) 1 energy, (b) = 3 energies, (c) = 5, (d) = 7, and (e) = 13. The multiples of $\pi/\delta k$ at which artifacts can remain on spherical surfaces surrounding the origin are also indicated; the shaded peaks all occur at such positions. [From Thevuthasan et al., ref. 32b]

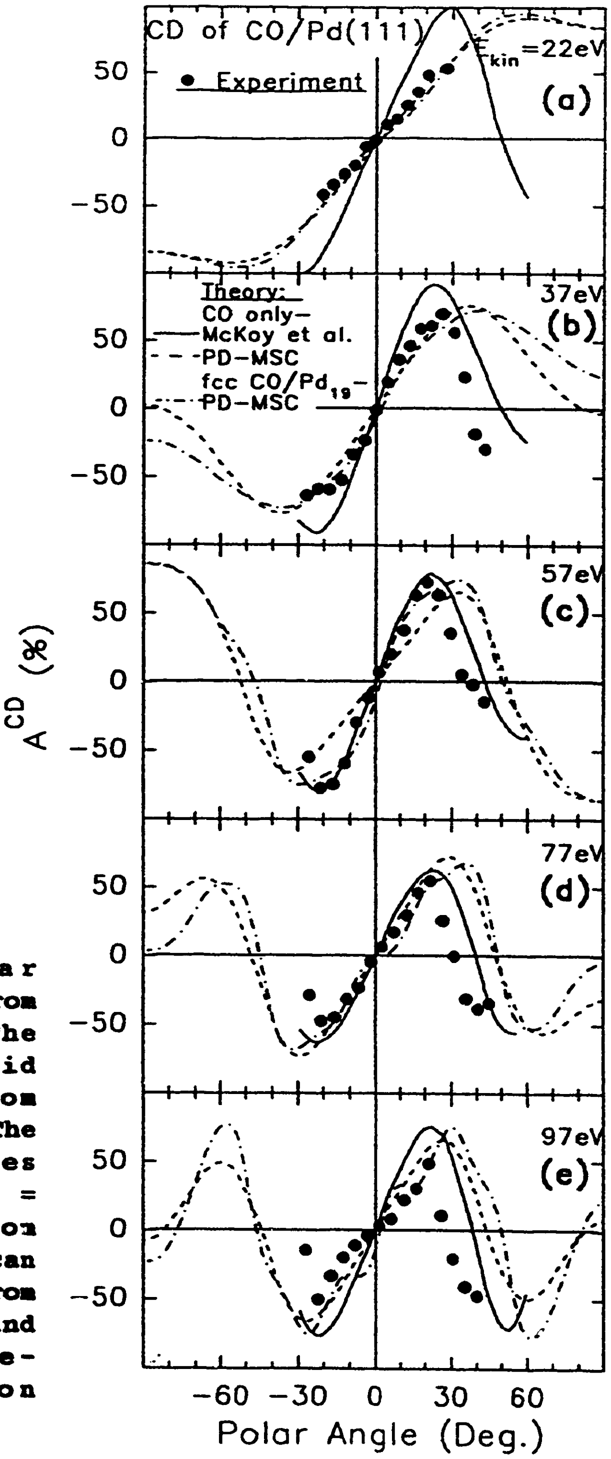


Fig. 9- Normal circular dichroism in C 1s emission from CO adsorbed on Pd(111). The experimental data and solid theoretical curve are from Bansmann et al. [ref. 39a]. The other two theoretical curves (------ = CO only and - --- - = CO in fcc sites on a 19-atom Pd(111) cluster, with the θ scan in the [10,-1] azimuth) are from Westphal et al. [ref. 39b] and are based upon multiple-scattering photoelectron diffraction calculations.

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