Holographic LEED

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We propose a new electron holographic scheme for the three-dimensional imaging of the atomic environment of disordered adsorbate atoms on a crystal surface, in which the object acts as its own beam splitter. The technique may be regarded as a direct method in low-energy electron diffraction, or even, in a sense, as a form of "lensless electron microscopy."

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The possibility of the holographic reinterpretation of several common forms of diffraction patterns, formed by the superposition of amplitudes from scattering paths differing in length by the order of typical interatomic distances, has been appreciated recently.[1] A practical method of reconstructing atomic-resolution images from a calculated photoelectron diffraction (PHD) pattern has also been demonstrated.[2]

A sufficiently bright and tunable source of x rays for such a PHD experiment is currently found only at a central radiation facility such as a synchrotron. An experiment closer to the spirit of Gabor's original idea,[3] and one which could be performed in a moderately equipped laboratory, would involve only an electron source. We point out, in this Letter, that a holographic interpretation of a diffuse low-energy electron diffraction (diffuse LEED or DLEED) pattern, formed by the backscattering of low-energy electrons from a crystal surface containing a disordered layer of adsorbate atoms, may offer a very practical route to atomic resolution.

The intensity of a DLEED pattern is generated by electrons which scatter off the nonperiodic parts of the surface, i.e., the adsorbate atoms. These atoms may be regarded as gateways through which electrons *must* pass on their journeys from electron gun to the detector screen. In the low-coverage limit, such as in the early stages of the adsorption of gas atoms on a clean crystal surface, the adsorbates interact primarily with the substrate atoms, rather than with each other, and form a lattice gas, having identical local substrate environments, but devoid of long-range order amongst themselves. The short-range order of their adsorption sites results in the wavelets emerging from the adsorbates being identical, apart from an origin-shift phase factor. If the wavelets were superposed to determine the total amplitude scattered in any particular direction, it is easy to see that (except in the directions of the substrate's Bragg beams) due to the randomness of the origin-shift phases, the adsorbates *behave* like a collection of incoherent scatterers, and that the form of a DLEED pattern (apart from a uniform scaling factor) is the same as that from a single adsorbate.[4] (The origin-shift phases become equal in the directions of the substrate's Bragg beams, but since, in the experiment we propose, the Bragg intensities are deliberately removed from the data to be processed, this fact is of no concern to us.)

The assumption of lattice-gas adsorption might be expected to break down after the coverage exceeds a certain value. The magnitude of this limiting coverage must depend on the relative sizes of the relevant interatomic forces and will vary amongst different adsorbate and substrate combinations. For the moment, we wish merely to point out that DLEED intensities may be (and have been) measured under conditions for which the adsorbates *do* form a lattice gas. The evidence is presented in Fig. 1, which shows experimental and calculated DLEED intensities for a Ni(100) surface exposed to oxygen at 80 K. The calculations were performed with the computer program of Saldin and Pendry,[5] which assumes lattice-gas disorder. The remarkable agreement seen between experiment and theory was found for the optimal structure in a recent systematic search.[6]

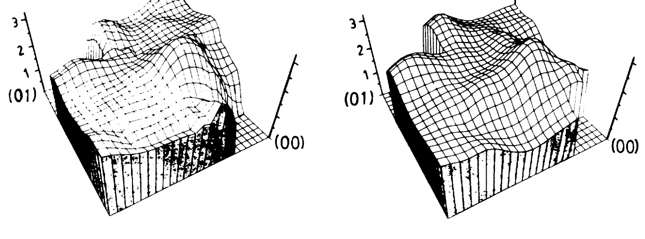

FIG. 1. Experimental (I_e) and best-fit theoretical (I_t) DLEED intensities for 64-eV electrons from a disordered O/Ni(100) surface (details in Ref. 6) with intensities around the (00) and (01) Bragg spots removed. Experimental data reproduced courtesy of Professor K. Heinz.

Under such conditions, we need analyze only the form of the diffuse wave field produced by a single adsorbate atom. An electron emerging from such an atom may travel either directly to the detector, or else do so after scattering off nearby substrate atoms. Since a DLEED pattern is formed by the interferences between these two components, if we identify the former as the reference wave, R, and the latter as the object wave, O, we may regard a DLEED pattern as a hologram. Whereas in the PHD experiment[2] electrons are ejected from the adsorbate atoms due to the absorption of x-ray photons, in DLEED they are directed into the sample by an external electron gun, and the adsorbates serve as beam splitters.

Consequently, the wave field forming the DLEED pattern may be written as

$$\psi = R + O \,. \tag{1}$$

Both R and O originate from a region of the order of interatomic distances, while the detector measures intensities on a backscattering hemisphere, macroscopic distances away. The spatial distribution of the DLEED intensities on this hemisphere may therefore be uniformly mapped onto an angular distribution. In this limit we may write the following:

$$R = F_0(k,\hat{\mathbf{k}})e^{ik\hat{\mathbf{k}}\cdot\mathbf{r}} \,, \tag{2}$$

and

$$O = \sum_{i\neq 0} F_i(k,\hat{\mathbf{k}})e^{ik\hat{\mathbf{k}}\cdot(\mathbf{r}-\mathbf{r}_i)} \,. \tag{3}$$

In these expressions F_i represents the angular variation of the amplitudes of the spherical waves, $\mathbf{k}$ the wave vector of the electrons, $\mathbf{r}_i$ the position of the ith atom (the origin of coordinates is assumed at the position of the adsorbate), and $\mathbf{r}$ a general position vector. Note that the presence of electron multiple scattering may alter the magnitudes of the F_i, but not the form of expressions (4) and (5).

The intensity detected on a hemispherical detector screen can be written as

$$I = R^*R + O^*O + R^*O + RO^* \,. \tag{4}$$

If we imagine this intensity distribution imprinted on a positive photographic film, and the film [which by proper development may take on the contrast value $\gamma = -2$ (Ref. 7)] illuminated by the conjugate of the reference beam, R^*, which would in this case be in the form of a converging spherical wave, the transmitted amplitude would be proportional to

$$|R|^2R^* + |O|^2R^* + |R|^2O^* + (R^*)^2O \,. \tag{5}$$

So long as the reference wave is uniform in both amplitude and phase, the first term above would be proportional to the conjugate of the reference wave and the fourth that of the object wave, i.e., in our case to spherical waves converging on the adsorbate and nearby substrate atoms, respectively. The third term is associated with the so-called twin image, well known in holography,[7] and the second term would be negligible if

$$|R| \gg |O| \,. \tag{6}$$

This condition is certainly satisfied in our case, since the object wave is derived from the reference wave, after extra backscattering. The condition that the phase of the reference wave be constant over the wave front is also satisfied, since the hologram is assumed a hemisphere centered on the adsorbate atom, but the uniformity of the amplitude of R over this hemisphere can only be regarded as approximately satisfied. Moreover, since it is probably impractical to reproduce the exact conjugate of this wave at the reconstruction stage, we follow Barton[2] and investigate reconstruction with an idealization of this wave, namely, a converging wave of uniform amplitude and phase over the hologram.

Returning to Eq. (5) for the DLEED distribution, we notice that the first term does not contain any information about the phase of the reference wave and is slowly varying over the backscattering hemisphere. Under condition (6), the second term would be expected to be negligible compared to all the others. Thus neither term would be expected to cause significant diffraction of the reconstructing wave. However, the last two terms must contain fringes due to the interferences between the reference and object waves. These will significantly alter the form of the reconstructing beam after propagation through the positive photographic film representing the hologram. In a region within a few interatomic spacings of the origin (i.e., the adsorbate atom) we may determine this wave field from the Helmholtz-Kirchoff integral:[2,8]

$$A(\mathbf{r}) = \int (R^*O + RO^*)e^{-ik\hat{\mathbf{k}}\cdot\mathbf{r}}d\hat{\mathbf{k}} \,. \tag{7}$$

Substituting expressions (2) and (3) above, it is easy to see that stationary phase conditions[2] result in the magnitude of $A(\mathbf{r})$ peaking at positions

$$\mathbf{r} = \pm\mathbf{r}_i \,, \tag{8}$$

the positive sign corresponding to the true positions of atoms in the substrate near the adsorbate, and the negative sign to the positions of their twin images.

In order to test our hypothesis, and to simulate a DLEED pattern in the lattice-gas limit, we calculated that from a disordered layer of O atoms located in the hollow sites at a height of 0.9 Å (Ref. 9) above the topmost substrate layer on a Ni(100) surface, using the program of Saldin and Pendry.[5] We then used the Helmholtz-Kirchoff integral (7) to evaluate from the resulting DLEED intensity distribution, $I(k,\hat{\mathbf{k}})$, the reconstructed amplitude:

$$A(\mathbf{r}) = \int\int I(k,\hat{\mathbf{k}})\exp[-ikz(1-\hat{k}_x^2-\hat{k}_y^2)^{1/2}] \times\exp[-ik(x\hat{k}_x+y\hat{k}_y)]d\hat{k}_x d\hat{k}_y \tag{9}$$

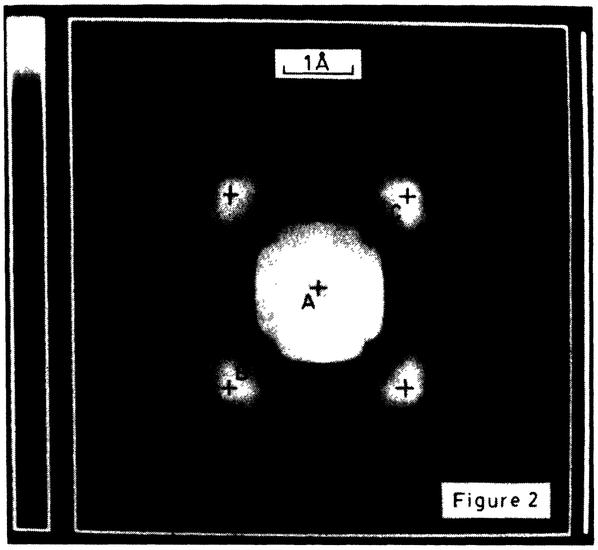

FIG. 2. An atomic-resolution holographic image from electrons of energy $E=435$ eV of the plane of the outermost substrate layer. The cross A marks the projection of the position of the adsorbate atom, and those denoted by B and C two of the nearest-neighbor atoms in the outermost substrate layer. Inset: The nonlinear intensity scale used.

by means of a fast-Fourier-transform routine.[10] In the above expression, x and y are Cartesian coordinates in the plane of the surface, and z is parallel to the surface normal.

The spatial distribution of the resulting reconstructed intensity $|A|^2$ for electrons of energy $E=435$ eV in the plane of the outermost substrate layer (corresponding to $z=-0.9$ Å) is illustrated in Fig. 2. Apart from the strong central peak associated with (largely undiffracted) components of the wave converging near the projection, A' of the adsorbate atom, intensity maxima are seen close to the positions (e.g., B and C) representing the nearest substrate atoms. Since the terms, such as the first two of Eq. (5), which we neglected in our argument leading up to Eq. (9), may be expected to give rise to random spurious features, we repeated the calculations for $E=424$ and 462 eV, and averaged the three resulting images. A marked improvement in image quality is observed, as shown in Fig. 3(a). Since (9) may be used to calculate a three-dimensional distribution of intensities, it is of interest to examine the distribution of reconstructed intensities in a plane perpendicular to the surface and passing through two of the substrate atoms nearest the adsorbate. This energy-averaged image is shown in Fig. 3(b). The position, D, of the second-layer Ni atom directly below the adsorbate appears to be clearly imaged. The features above the adsorbate position, A, are the twin images, referred to above. The resolution of the atomic images is seen to be close to the diffraction limit, of the order of the wavelength of the electrons (~0.5 Å).

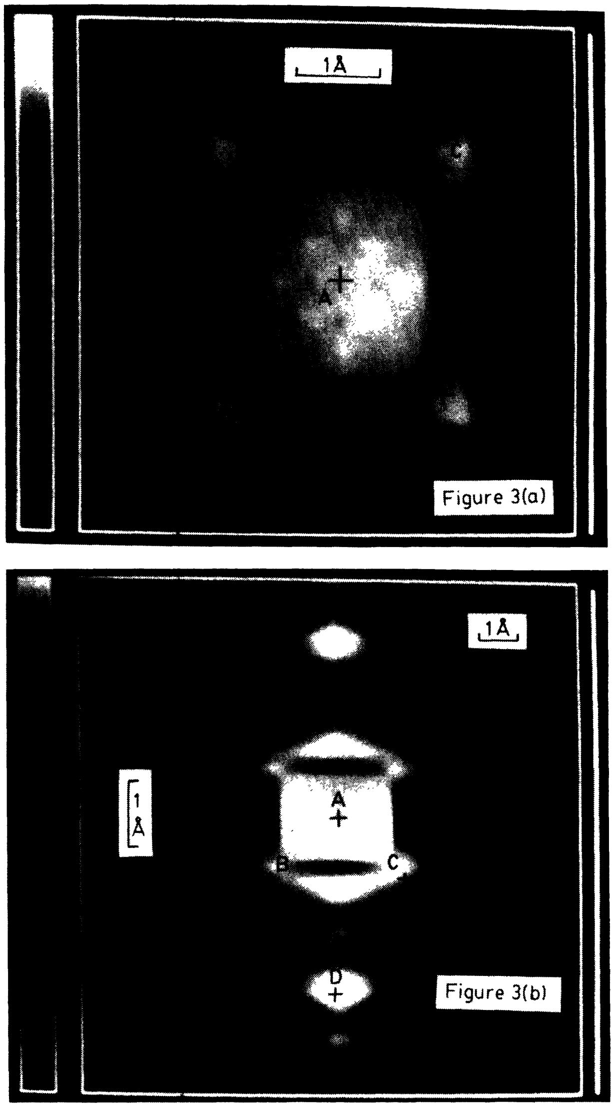

FIG. 3. Sections through the three-dimensional holographic image obtained by averaging those for $E=424$, 435, and 462 eV. (a) The same plane as that of Fig. 2; (b) the plane perpendicular to the surface and passing through the adsorbate A and substrate atoms B and C. The atom in the second substrate layer directly below the adsorbate is denoted by D. The features above A are the twin images.

These are results of extraordinary implications. Although LEED has been a very successful probe of surface structures,[11] it has been limited by the need to perform complex multiple-scattering calculations, and by the difficulty of implementing a true *direct* method[12] for determining a structure from the experimental data. The technique we propose here overcomes both these problems, since it is a true direct method, and because the concentration of the reconstructed intensities around atomic positions is independent of the details of the multiple scattering (only the intensities are affected, not the positions).

Experimental techniques of DLEED are now relatively routine[13-16] and we believe that the collection of data for electron energies corresponding to wavelengths small enough for atomic resolution (probably a few hundred eV) to be very feasible. The computational resources required for image reconstruction are only a minute fraction of that for a typical LEED structure determination, usually measured in hours of supercomputer central processing unit time. In contrast, it is possible to generate a holographic image like that displayed in Fig. 2 from a DLEED pattern in a matter of a few minutes on a personal computer. This opens up the possibility of the rapid processing of the diffraction patterns to yield real-time atomic-resolution images.

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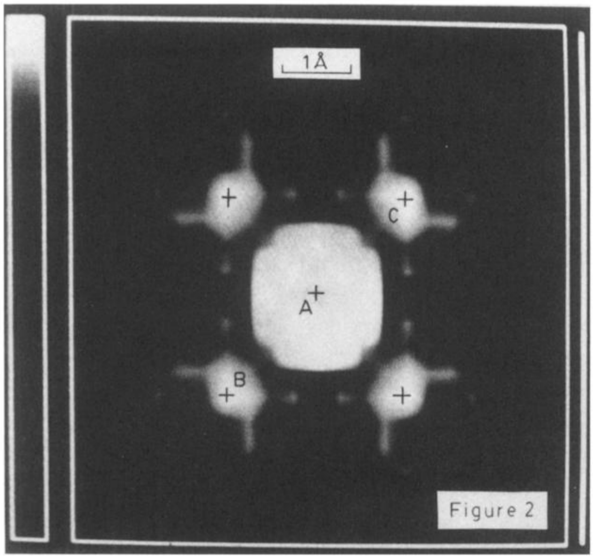

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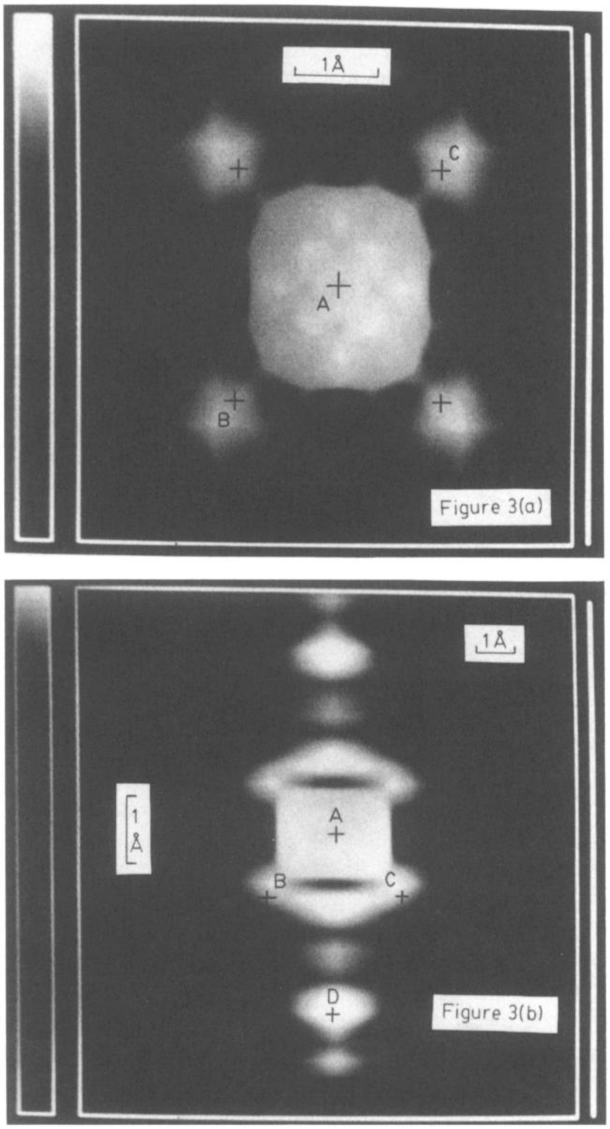

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