Atomic-Resolution Electron Holography in Solids with Localized Sources

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We demonstrate three-dimensional reconstructions of the relative positions of atoms in a crystal by holography with atomically localized incoherent electron sources. Example reconstructions are shown using experimentally measured reflection electron Kikuchi-scattering angular distributions.

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Electron holography was conceived by Gabor as a means to overcome the barrier of electron-optical lens aberrations which prevented the achievement of atomic resolution.[1] The production of electron beams of high coherence and low-energy spread with field-emission sources has made it feasible to generate holographic images of samples by illumination with an external coherent electron source.[2] An alternative approach to holography, in which the electron reference waves are created in the sample itself, was demonstrated by Bartell and Ritz[3] and used to image the electron cloud of rare-gas atoms.

A conjecture by Szöke[4] that localized electron sources could produce three-dimensional holographic images of solids with atomic resolution has resulted in intensive efforts to demonstrate this effect. Computer models using multiple-scattering calculations to simulate an electron hologram have been constructed by Barton for the case of photoelectron diffraction[5] and by Saldin and de Andres[6] in the case of diffuse low-energy electron diffraction (diffuse LEED). These simulations have indicated that a sequence of phased two-dimensional Fourier transforms of an electron-diffraction pattern could, in principle, produce images of the atomic positions in a solid, even in the presence of strong multiple-scattering effects.

We report here the first experimental reconstruction of a three-dimensional crystal structure with atomic resolution by electron holography. These results prove that diffraction patterns produced by localized electron sources, such as those from Auger,[7] photoelectron,[8] and Kikuchi electrons,[9] can be interpreted as holograms. In addition, these results demonstrate an extension of electron holography to conditions in which electron backscattering is weak, and diffraction is dominated by forward scattering. It is now possible to construct real-space images of atoms by direct transform methods applied to chemically specific electron-diffraction patterns.

Atomic resolution is possible with incoherent electron sources by the method of Fraunhofer holography.[10] The hologram is produced when a "reference" electron wave is emitted from the vicinity of an atom core and interferes with an "object" wave produced by scattering of the reference wave from neighboring atoms. Since the distances between the objects (the nearby atoms) are small compared to the distance to the detector, the resulting interference pattern may be regarded as a lensless Fraunhofer hologram. A property of this type of hologram is that its form depends only on the relative positions of the objects, and not on the distance from the objects to the hologram. The result is that the individual holograms from each electron source are perfectly superimposed upon each other and reconstruction of the relative positions of atoms in the vicinity of the source is possible, provided that the source atoms are orientationally ordered in equivalent local environments. Both the hologram and its subsequent reconstruction, therefore, represent averages over all source atoms and their near neighbors.

The theoretical connection between electron diffraction and holography has been recently established for both photoemission and diffuse LEED. Barton[5] has shown that when an atomic adsorbate on a crystal surface is a source of photoelectrons, the resulting computer-simulated diffraction pattern formed by the interference between the direct waves and those backscattered from the nearby atoms in the substrate may be reinterpreted as a hologram, from which an atomic-resolution image may be reconstructed by computer. It was later pointed out by Saldin and de Andres[6] that the diffuse elastic-electron-scattering pattern from a disordered lattice gas of adsorbate atoms should also contain interference fringes which could be interpreted as a hologram.

These results suggest that other localized incoherent electron sources may also form similar holograms. There is an experimentally well-established similarity between the final-state diffraction patterns produced by high-energy photoelectrons, Auger electrons, and quasi-elastic (Kikuchi) electrons, for example.[9,11,12] Each of these three types of diffraction patterns satisfy the requirements of lensless Fraunhofer holography. In the case of photoelectrons and Auger electrons, the core hole associated with the primary excitation is responsible for the source localization. It has also been established that reflected electron Kikuchi patterns may be understood by a similar model of atomically localized sources of incoherent quasielastically scattered electrons.[13]

Perhaps the most obvious effect observed in the diffraction of electrons with kinetic energies above about 500 eV is the appearance of the so-called "forward-focusing" peaks observed at emission angles corresponding to projections of atomic rows in the crystal. This effect is due to the predominantly forward-scattering nature of atomic form factors at such electron energies.[14] The forward-focusing diffraction pattern formed by Kikuchi electrons bears a striking resemblance to the patterns produced by Auger or photoelectrons of similar kinetic energy.[7,8]

The aim of this Letter is to show that forward-focusing patterns are repositories of a previously unsuspected source of three-dimensional atomic structural information accessible by their reinterpretation as Fraunhofer holograms. The forward-focusing diffraction peaks are confined to specific regions of the hologram, and they provide information about bond angles in the sample. However, the important holographic information of three-dimensional local atomic environment resides in intensity modulations distributed throughout the 2π sr hemisphere of the diffraction pattern. We present here, in detail, results based on a measurement of the Kikuchi pattern of 1075-eV electrons reflected from a Cu(001) surface. We have obtained similar results using Auger and photoelectron diffraction patterns[8] with electrons of similar energies from a Cu(111) surface.

Consider the near-surface atomic configuration of the (001) surface of a face-centered-cubic (fcc) metal such as copper (Fig. 1). Apart from the outermost layer, all atoms have nearest neighbors in identical relative positions. After an inelastic-scattering event, a Kikuchi electron may emerge from the surface in a particular direction either without further scattering or else after elastic scattering from its near neighbors. The electron current detected in a given direction is the result of a coherent superposition of the amplitudes of all possible scattering paths. Identifying the direct path from the inelastic scatterer to the detector with the reference wave, and paths involving subsequent elastic scattering from atom cores with object waves, the Kikuchi pattern (Fig. 2) may be reinterpreted as a hologram.

The Kikuchi pattern of Fig. 2 was acquired from a Cu(001) single crystal which was prepared using standard surface-science techniques. This pattern was generated from a digitized video image of a conventional reverse-view LEED system using an incident electron-beam kinetic energy of 1075 eV. At this energy, the purely elastic-backscattering intensity is very small, leading to the absence of LEED spots. However, lattice vibrations lead to a proportionally larger quasielastic-backscattering intensity, giving rise to the Kikuchi pattern. The figure shows a standard stereographic projection of the angular distribution of Kikuchi electrons, ranging from normal emission at the center of the figure to $\approx 50°$ polar angle at the edge. The pattern was fourfold-symmetry averaged.

In order to demonstrate that this pattern is a hologram, we have performed a holographic reconstruction by the computer methods previously described.[5,6] The spatial distribution of intensity in the hologram (Fig. 2)

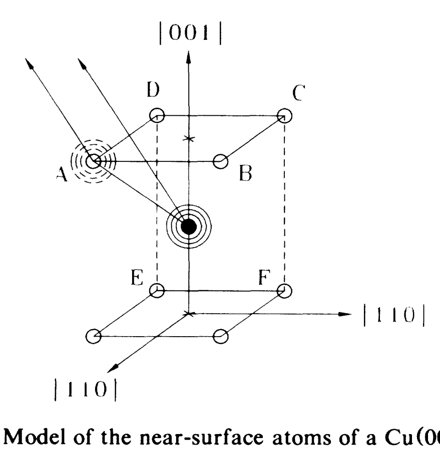

FIG. 1. Model of the near-surface atoms of a Cu(001) crystal. The source atom is shown at the origin emitting a direct (reference) wave. Object waves emanate from neighboring atoms to produce an interference pattern which can be recorded in the form of electron intensity as a function of emission angle.

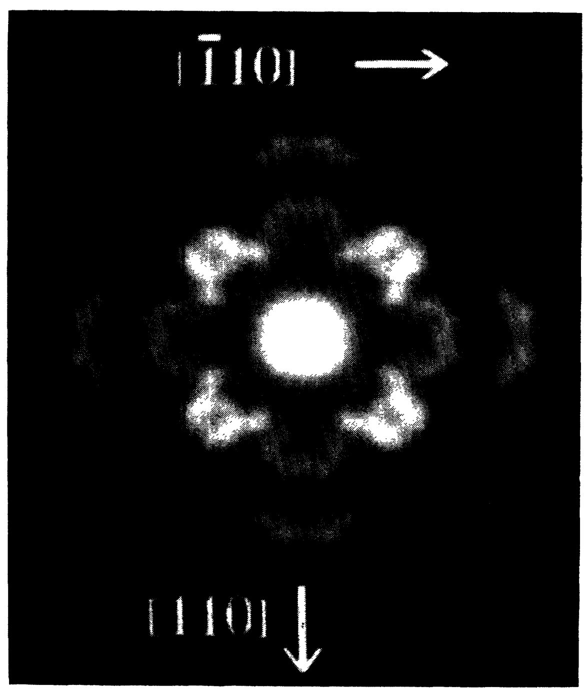

FIG. 2. Measured Kikuchi pattern hologram at 1075 eV from Cu(001). The electron interference pattern is displayed as a function of parallel momentum, with zero momentum (normal emission) at the center. The arrows show the orientation of the crystallographic axes.

is mapped into an angular distribution of electron scattering, $I(\hat{\mathbf{k}})$, where $\hat{\mathbf{k}}$ is a unit vector parallel to the electron momentum. It is common practice in photoelectron diffraction to remove a smoothly varying contribution, $I_0(\hat{\mathbf{k}})$, due to the reference wave and instrumental factors, and to work with an anisotropy function $\chi(\hat{\mathbf{k}})$, defined by $I(\hat{\mathbf{k}})=[1+\chi(\hat{\mathbf{k}})]I_0(\hat{\mathbf{k}})$. A transform of the anisotropy function will have significant intensity only in the vicinity of atom positions: $r=r_j$, and at positions corresponding to the "twin image" $r=-r_j$. Since the precise form of I_0 is unknown experimentally, we have chosen to directly transform the measured intensity distribution using the Helmholtz-Kirchhoff formula:[10]

$$A(\mathbf{r})=\int I(\hat{\mathbf{k}})e^{-ik\hat{\mathbf{k}}\cdot\mathbf{r}}d\hat{\mathbf{k}}. \qquad (1)$$

This straightforward Fourier integral results in a three-dimensional intensity distribution in real-space coordinates, $|A|^2$, constituting the reconstructed image. Including the reference wave only contributes additional intensity near the origin of the reconstruction.[6] In order to display the information in this image, we plot two-dimensional intensity distributions (sections) which correspond to the expected positions of atomic planes. Two such sections are displayed in Figs. 3 and 4.

We define our origin as being at the position of the source atom (point O in Fig. 1), and our coordinate axis directions as in Fig. 1. Figure 3 shows the intensity distribution in the (001) plane containing atoms labeled A, B, C, and D. The crosses are at the expected positions of the atoms in that plane. We see excellent agreement between the intensity maxima and the atomic positions.

Figure 4 shows the intensity distribution in the (110) plane passing through atoms C, D, E, and F. Again, the crosses indicate the expected positions of the atoms, and the intensity maxima in Fig. 4 show excellent agreement with these positions. The combination of the planar sections of two different crystal planes (Figs. 3 and 4) aptly illustrates the three-dimensional nature of the holographic reconstruction.

The Kikuchi hologram consists of superimposed interference fringes due to all reference-object wave combinations. However, not all atoms which are crystallographically equivalent nearest neighbors to the source atom are reconstructed with equivalent intensity in the real-space map, due to the effects of forward scattering. The strongest interference fringes in the hologram will be produced by atoms which lie near a forward-scattering path between the source atom and the detector. In Fig. 1, these atoms are labeled A through D, and produce the reconstruction shown in Fig. 3.

In contrast, we do not find significant intensity in the reconstruction for atoms in the same plane as the source atom. Scattering from the emitter through an in-plane atom and finally to the detector requires a large angle

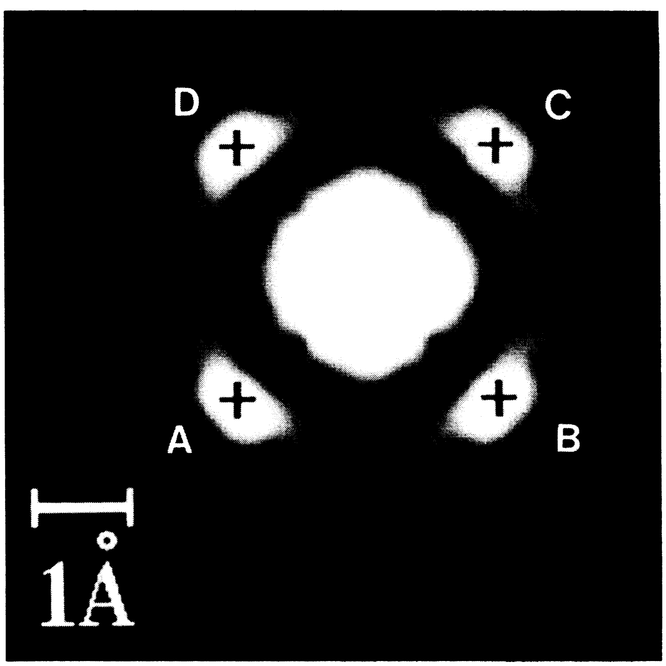

FIG. 3. Holographic reconstruction of the Kikuchi pattern in the previous figure, corresponding to a plane of atoms lying above the source atom (i.e., between the source and detector). The crosses mark the positions of atoms in the ideal crystal.

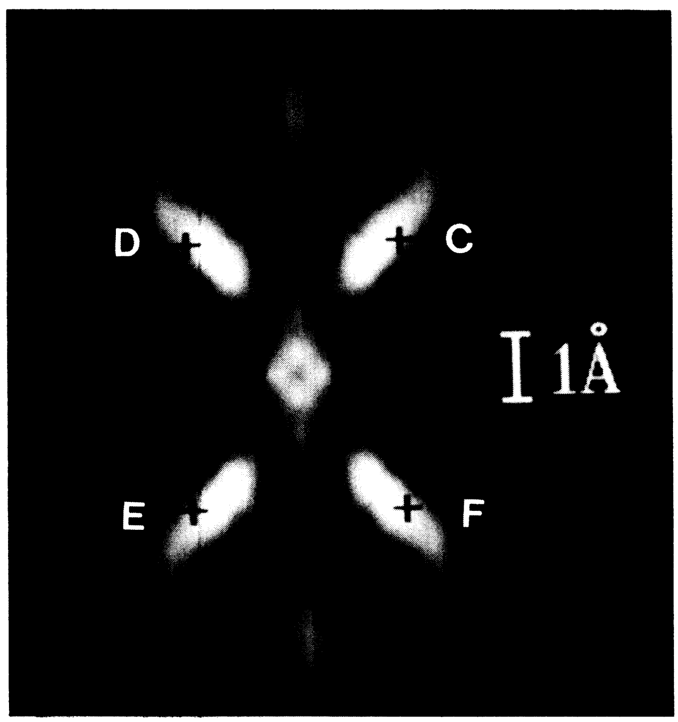

FIG. 4. A view in the plane perpendicular to the surface of the holographic reconstruction of the data from Fig. 1. The crosses mark positions of atoms in the ideal lattice. The elongated shape of the atom image is due to a reduction in resolution parallel to the electron emission direction.

($\sim 90°$) of scattering, for which the atomic scattering factor is relatively small. We emphasize this important feature of holograms which show large forward scattering: The holographic interference pattern for a particular string of atoms will be strongest in the portion of the hologram surrounding the forward-scattering peak for that particular atom string. In order to reconstruct atoms in the plane of the source, the hologram must extend to near-grazing collection angles.

The features at F and E in Fig. 4 are primarily due to the holographic twin images of atoms A and B, produced by inversion around the source atom. The twin images overlap the true holographic images of these atoms, since the crystal itself possesses inversion symmetry. The true image of atoms below the source is expected to be weak, because of the small contribution of backscattered waves to the overall diffraction pattern.

The intensity at the center of Fig. 3 does not correspond to an atom position. Although it represents a local maximum in the displayed plane of the reconstruction, the absolute maximum of this feature occurs at the origin, and corresponds to the source atom. Since we do not remove the reference wave from the Kikuchi hologram, the direct wave from the source produces an intense feature in the reconstruction.

Until now, it has not been appreciated that the forward-focusing pattern of a single-crystal sample contains this kind of holographic information. Here, we have demonstrated that forward-focusing patterns of quasielastic reflected electrons may be used to reconstruct a three-dimensional image of a single-crystal sample. We have also found similar results from the reconstructions of photoemission and Auger diffraction patterns, and these results will be discussed in a paper that is to follow. The advantage of photoemission and Auger reconstructions is that they give the local crystal environment of a particular chemical species. This will be an important asset in structural measurement for a wide range of multicomponent systems, ranging from chemisorption to interfaces to epitaxial films.

These images convincingly prove the predictions of previous multiple-scattering simulations[5,6] that electron holography with local atomic sources is indeed feasible, and that this may be used to generate striking three-dimensional images of the atoms in the regions immediately surrounding the electron sources. We have extended the generality of the holographic interpretation of electron diffraction to include geometries in which forward scattering is dominant. In the previously considered backscattering geometries, higher electron energies are preferred in order to achieve atomic resolution, but at the cost of severely reduced interference fringe intensity due to the low backscattering probability of high-energy electrons. In contrast, the forward-scattering geometry for holography produces strong holographic fringes, yet is compatible with the use of high-energy electrons to achieve atomic resolution.

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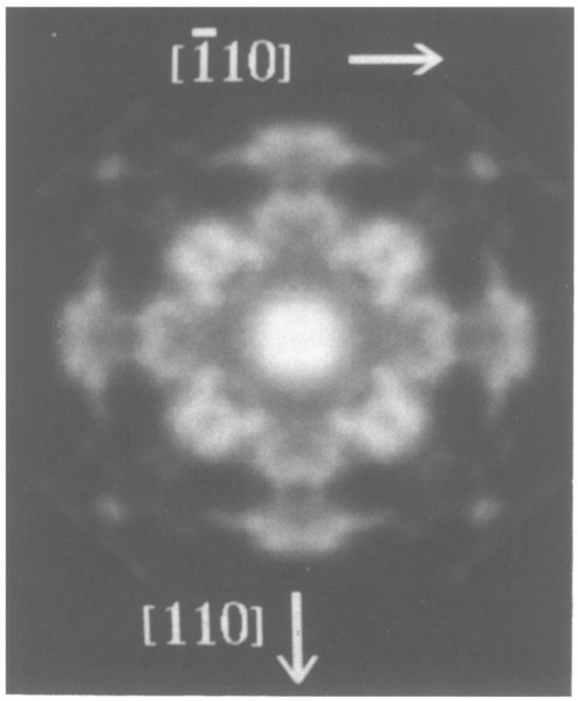

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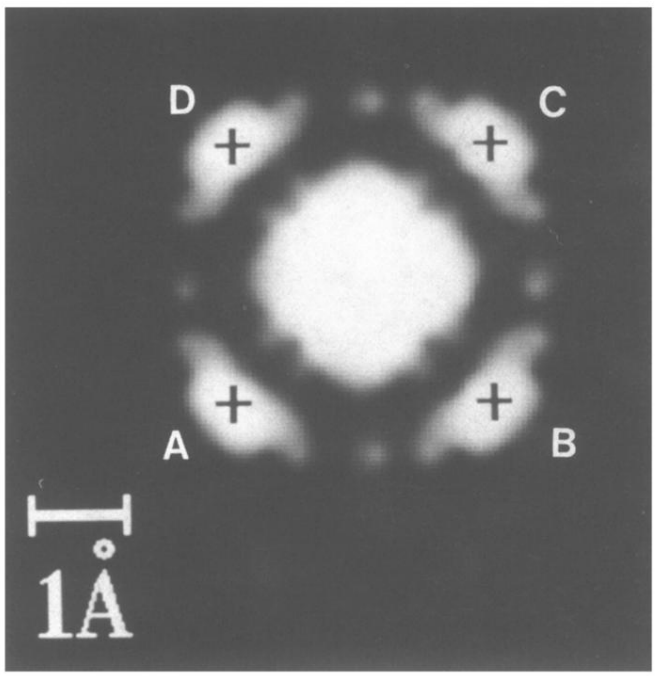

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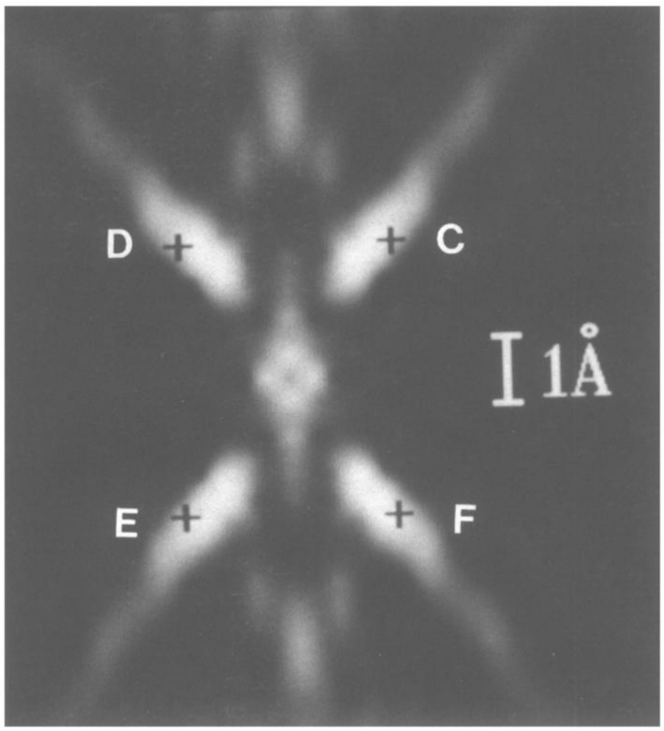

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