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Atomic imaging of surfaces in plan view

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We report experimental results imaging the surface diffraction spots in the plan view geometry from the Si(111) surface and the Ir(001) surface. High quality images have been obtained using conventional large aperture high resolution electron microscopy (HREM), a smaller aperture to exclude the bulk diffraction spots and with highly tilted crystals. The experimental data indicates that there should be no major problems in obtaining atomic scale surface information in plan view.

1. Introduction

Atomic level information about surfaces offers the possibility of critical information that is not available from other techniques. Historically, both electron microscopy in the profile imaging mode [1–3] and scanning tunneling microscopy (STM) [4] appeared at about the same time providing essentially the same type of atomic level information. Profile imaging has seen some utilization, e.g. refs. [5–7], but has certainly not been the widespread success that STM and its children such as atomic force microscopy (AFM) have been.

Given that one can now routinely and relatively cheaply image the outermost surface with either STM or AFM, one might ask whether there is any point in using electron microscopy, a far more expensive technique, to look at surfaces. There is, since electron microscopy is highly sensitive to the bulk structure as well as the surface, while the material below the outermost surface layer is essentially invisible to STM or AFM. Given that the surface conditions can be controlled to a high level of cleanliness, something that has only recently become possible (e.g. refs. [8–10]), how should one approach imaging a surface with an electron microscope? One obvious possibility is the profile technique, but there are major questions about how representative such data is. Profile imaging requires a very thin edge (of the order of 10–20 nm at most in thickness) and there is every reason to suspect that long range elastic/electronic energy terms will be truncated by the limited thickness of the sample. A far more attractive alternative is to look at a surface with the beam normal to the surface, i.e., the plan view imaging mode, e.g. refs. [11–15]. Although there have been hints [16,17] that this can be done at the atomic level, this is not as yet a mature technique or even one whose limitations are known.

The intention of this Letter is to present initial experimental high resolution electron microscopy (HREM) results in directly imaging the atomic structures for the Si(111) and Ir(001) surfaces. Three different approaches, namely conventional HREM, aperture restricted HREM and off-zone HREM were utilized. We demonstrate atomic level information comparable to STM or AFM with simultaneous bulk information. (For reference, the Si(111) results were our initial data and were obtained at somewhat lower resolutions than the Ir(001) results.)

2. Experimental method

The silicon (111) sample was prepared by a combination of conventional ion-beam thinning

outside the microscope, and then sputter cleaning/annealing within a UHV Hitachi H-9000 microscope, as described previously [18]. Due to erosion of the aperture in the ion gun in the microscope, the area exposed to the ion beam included part of the sample holder which led to approximately a monolayer of Cu/Au (as determined later by energy-dispersive X-ray analysis) on the surface. This led to a (5×5) reconstruction with the underlying silicon flat with a (1×1) surface.

The iridium (001) sample was prepared by the same approach as above. Due to CO and CO_2 contamination during the electron beam anneals (stimulated desorption from a tungsten filament near the sample by secondary electrons from the

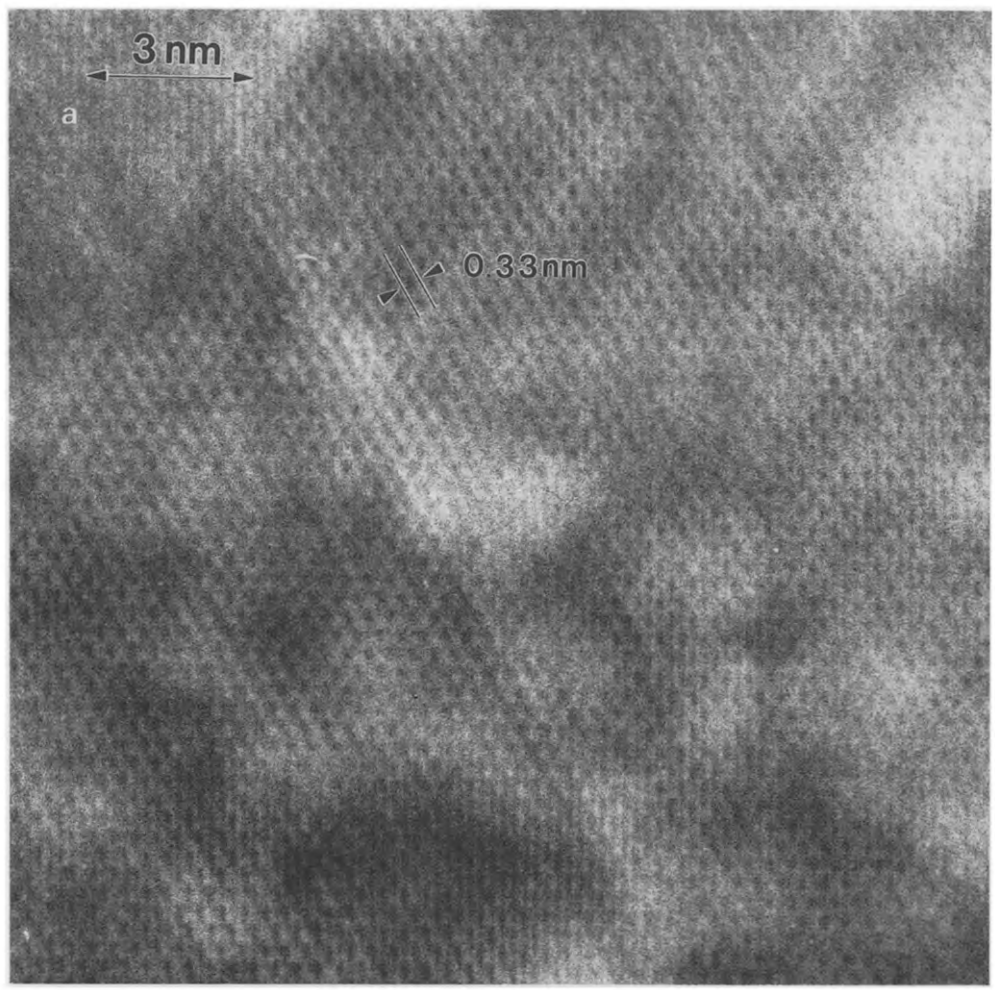

Fig. 1. (a) High resolution image of Cu/Au on silicon (111) showing the strong (1×1) fringes (0.33 nm) taken with an objective aperture large enough to include the bulk {220}. The contrast of the bulk {220} fringes was minimized at this defocus.

sample) initially only the (1×1) surface was formed. In later experiments when the filament was degassed the (5×1) reconstruction [19] was observed.

3. Results

Experimental images of the silicon (111)-(1×1) surface acquired under the three conditions are shown in figs. 1a–1c. The positions of the surface and bulk diffraction spots and the objective aperture used for fig. 1b are illustrated in fig. 2. Fig. 1a was taken with the crystal on the zone axis using a large objective aperture, i.e., conventional HREM. The image contained both (1×1) and bulk {220} fringes. Fig. 1b was with an objective aperture which included the (1×1) diffraction spots but excluded the bulk {220}. Surface imaging was also attempted by tilting the sample far

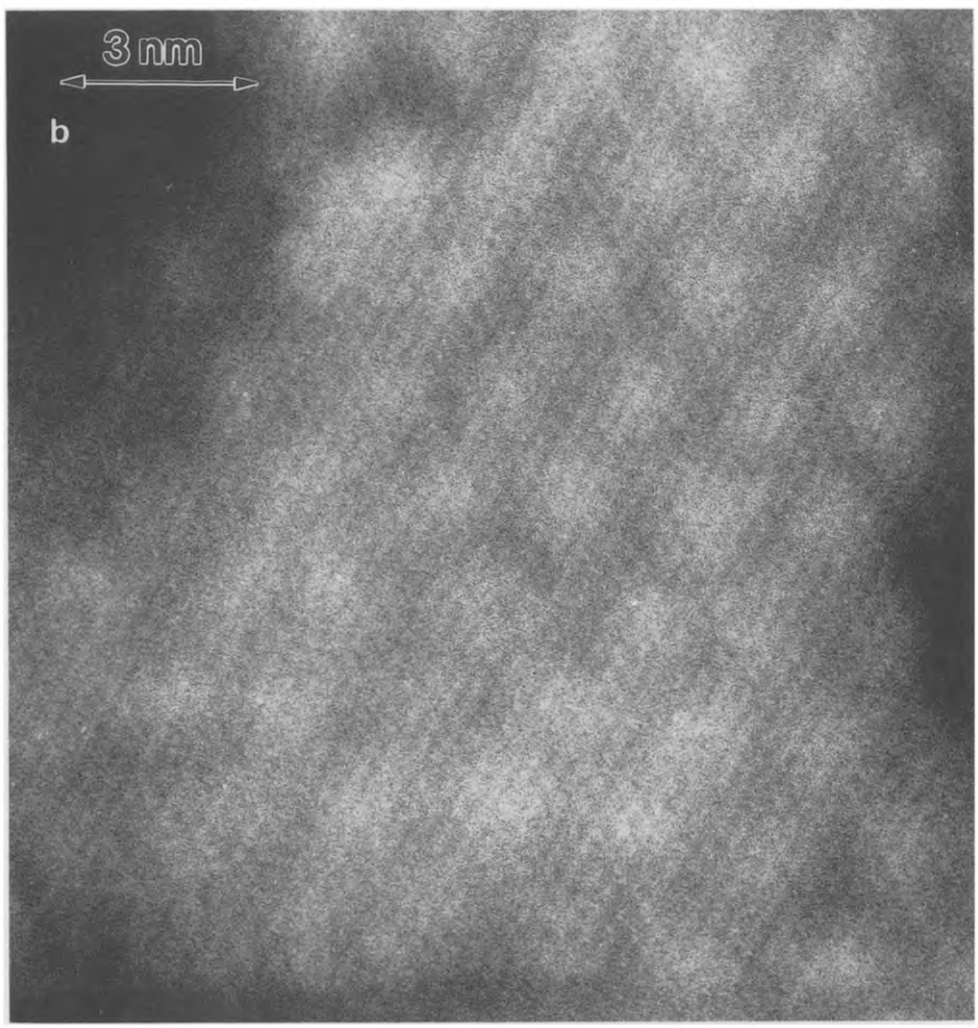

Fig. 1. (b) High resolution image of silicon (1×1) surface and moiré fringes of (5×5) reconstruction from the Cu/Au on Si taken with an objective aperture which included the (1×1) spots but excluded the bulk {220} spots.

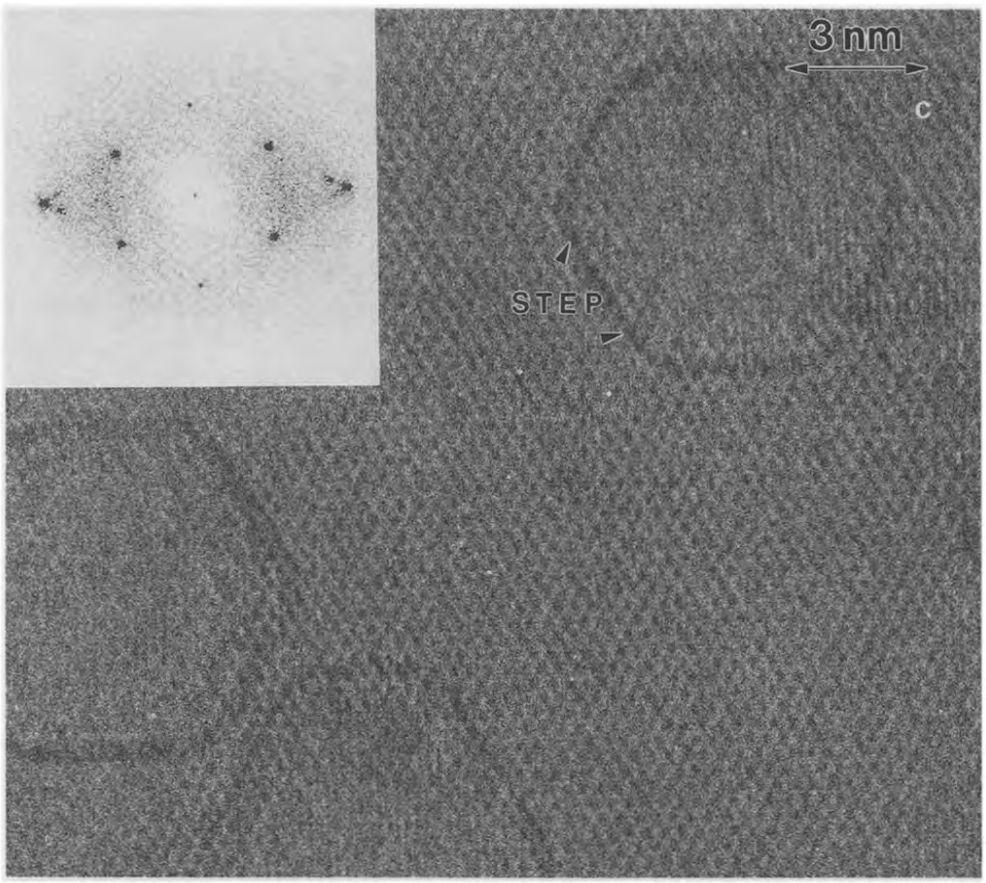

Fig. 1. (c) Off-zone high resolution image showing the silicon (1 × 1) lattice and surface steps, an optical diffraction pattern is shown in the inset.

off the zone where no bulk spots were strongly excited. The principle behind this technique is the long extension in reciprocal space of diffraction spots associated with the surface as illustrated in fig. 3; the bulk diffraction spots attenuate far faster as the crystal is tilted off the zone axis. Fig. 1c was obtained at such a condition with no objective aperture. The most apparent spacings in the image in fig. 1c are from the (1 × 1) surface spots. However, an optical diffraction pattern (inset) shows that one set of bulk (220) spacings and one of the fringes from the Cu/Au layer are present in the image as well. As mentioned earlier, the resolution in these images were not so good (due to vibrations from a diffusion pump); if it was better the Cu/Au layer would have been visible in the images similar to the Ir data shown below.

All three imaging techniques are viable in terms of imaging the surface, and each has its advantages and disadvantages. The large aperture images on the zone axis are quite focus dependent as would be expected. Therefore, the surface signal can be buried in the "noise" of the bulk fringe contrast unless care is taken. However, surface information can be obtained with the bulk at the same time for appropriate defoci, and it is therefore powerful for deriving the relative registry information.

The contrast for the case with an aperture

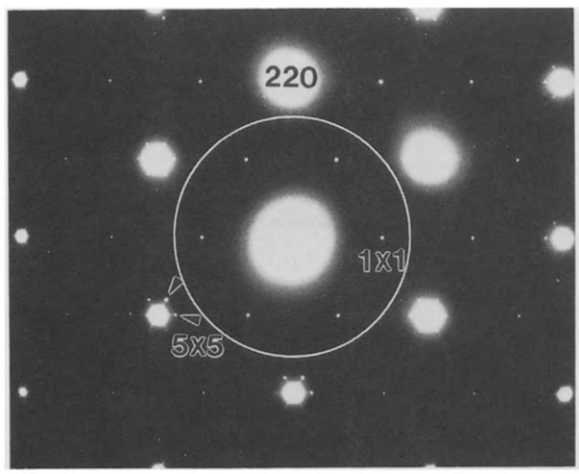

Fig. 2. Diffraction pattern of Cu/Au on Si(111) showing the strong bulk {200}, relatively weak first order Laue zone spots of {111} or so called (1×1) surface spots. Spots due to the Cu/Au (5×5) reconstruction are indicated by arrows. The big circle represents the approximate objective aperture size used for fig. 1b.

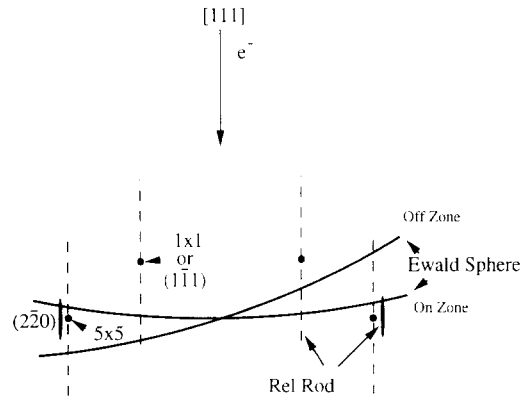

Fig. 3. Schematic illustrating the position of the Ewald sphere with respect to the reciprocal lattice spikes (or rel rods) of the bulk and surface spots when the sample is on zone and off zone.

excluding the bulk diffraction spots is also good, and the images are less defocus dependent. However, bulk diffraction contrast, which is not so

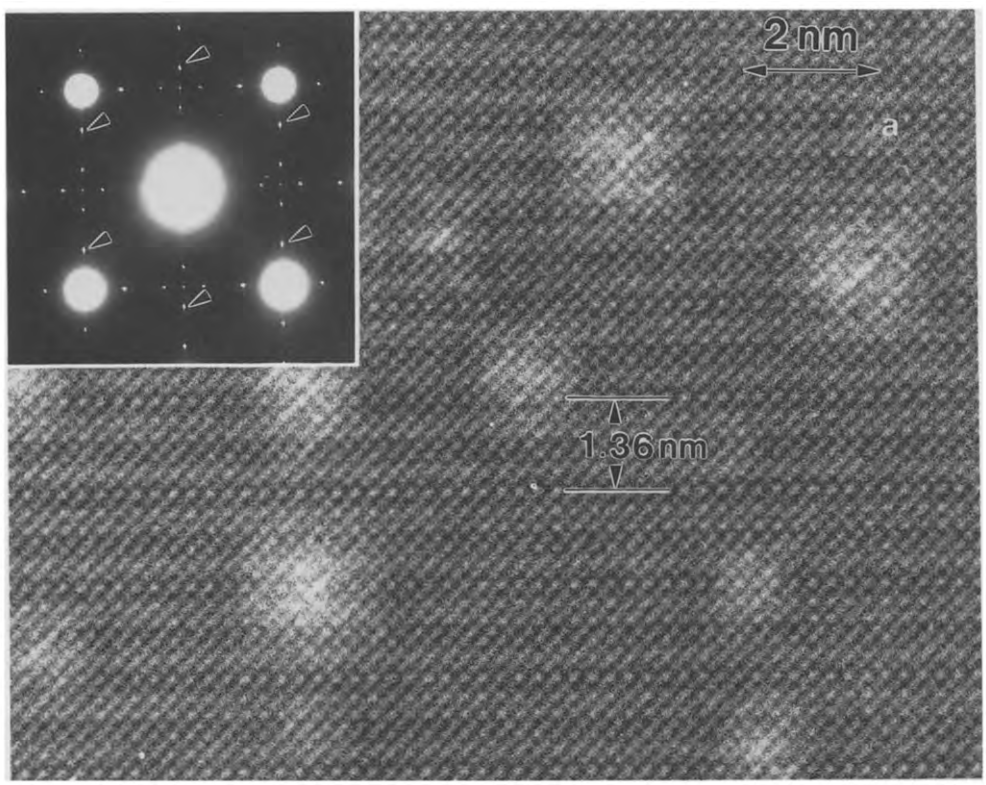

Fig. 4. (a) On-zone high resolution image of iridium (001)-(5 × 1) reconstruction showing the {200} bulk fringes and the moiré fringes of 1.36 nm along the "5" dimension of the reconstruction unit cell. Diffraction pattern inset shows two orthogonal domains of the (5 × 1) reconstruction, main spots forming a hexagon due to one of the domains is arrowed.

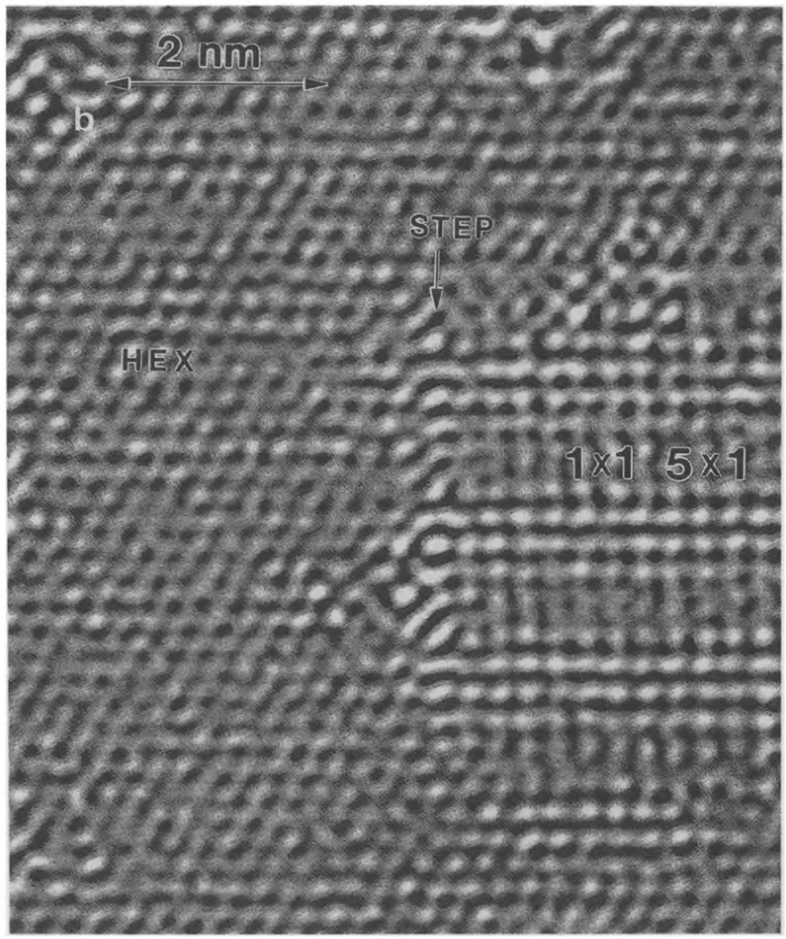

Fig. 4. (b) Off-zone high resolution image of the Ir(001) (5 × 1) reconstruction. Hexagonal surface structure (left) and the (1 × 1) structure (right) are visible in different areas separated by surface steps. Gentle Fourier filtering was employed to remove noise from the image.

significant when all the beams are included, complicates the images.

Finally, the image quality off the zone axis is also high. Due to the much higher transmitted beam intensity, the image contrast is lower than the on-zone cases. With appropriate printing or image processing, images show more surface sensitivity, and steps are much more visible.

The second example, with higher resolution, is shown in fig. 4 for the Ir(001)-(5 × 1) surface [20]. The on-zone image in fig. 4a is complicated and shows the bulk {200} fringes and the moiré fringes of the "5" dimension of the reconstruction, but no surface information is directly available in this image. The off-zone image in fig. 4b has the bulk contrast damped and shows the hexagonal surface layer structure and the (1 × 1) fringes much more clearly. The off-zone image is less sensitive to the surface registry which, in principle, is obtainable from the on-zone image and work is in

progress to obtain this information. We were not able to obtain useful images with a small objective aperture for this surface.

4. Discussion

The experimental results shown here in imaging silicon (111) and iridium (001) surfaces are quite encouraging. It does appear that conventional on-zone HREM or off-zone HREM are readily useable to obtain atomic scale images (the restricted aperture HREM is not a good technique). The conventional HREM is useful in that the images show simultaneously the bulk and surface information, so this is the optimal technique for obtaining, for instance, registry information. For completeness, we should mention that such information cannot be immediately obtained in most cases by simply looking at the images. However, electron diffraction and imaging theory is very mature, so the registry data can in principle be backed out from the images with some additional effort using well-established simulation techniques. Unfortunately on the zone axis some surface information such as surface steps is buried in the images and does not appear readily to the eye. In contrast, since there is little to no bulk contrast in the off-zone images, steps are quite clear, so are the surface structures. There is ample surface information available from an electron microscope, and coupling the absolute surface diffraction intensity information and HREM images for the phase information should provide a powerful approach to solve surface structures.

Acknowledgements

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