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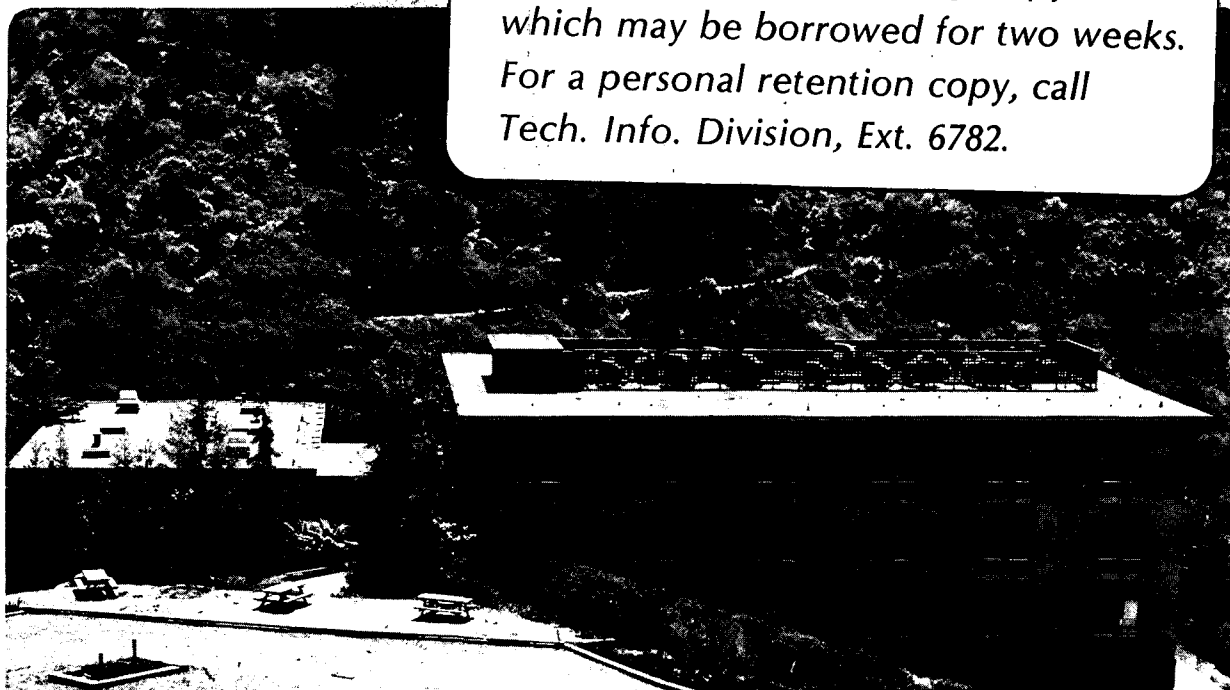
THE ARM 1000kV MICROSCOPE AT NCEM, BERKELEY
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R. Gronsky and G. Thomas

April 1984

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The ARM 1000kV Microscope at NCEM, Berkeley and Some Applications

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INTRODUCTION

The JEOL 1000kV atomic resolution microscope (ARM) is a unique instrument designed to a demanding set of performance specifications first issued in 1979. Our objective was to obtain an electron microscope capable of interpretable point resolution better than 0.18nm in order to carry out atomic resolution imaging of technologically important metals, ceramics and electronic materials. This instrument is now located at the National Center for Electron Microscopy in the Lawrence Berkeley Laboratory. The Center, with funding from the U. S. Department of Energy, was established to fill the need for a time-sharing facility where high cost instruments, support staff and facilities are all available to the scientific community. It is the first of its kind in the U. S. A. (Fig. 1), and was recently dedicated in the Fall of 1983.

Background

Although there have been no limitations on resolution imposed by instrumentation for quite some time in amplitude-contrast imaging, only the most modern microscopes have been successfully used for phase-contrast imaging of fine scale structure [1-4]. Fortunately, the sources of experimental difficulties which continue to prevent atomic resolution by this technique are also quite well known.

One set of problems has to do with beam-specimen interactions. In the phase-contrast imaging method, the specimen is frequently considered an integral part of the imaging optics since its only effect on the incident beam should be an alteration in phase [5]. This mandates that the object be very "thin", with the most severe constraints placed on preparing specimens of high atomic number. Other limitations include specimen orientation, irradiation damage, both ionization and knock-on, and contamination under the illuminating beam from both the microscope vacuum environment and adsorbed by the specimen.

Another set of problems stems from electron optics. Much like the specimen, the objective lens affects the electron beam through an overall phase distortion. This phase change (χ) in the diffraction spectrum is a function of scattering angle or position (u) in reciprocal space and can be written [6]

$$\chi(u) = \frac{2\pi}{\lambda} \left[C_s \frac{\lambda^4 u^4}{4} + \Delta z \frac{\lambda^2 u^2}{2} \right]$$

where λ is the electron wavelength, C_s the spherical aberration coefficient of the objective lens and Δz the defocus of the objective lens. Improving the contrast transfer characteristics of the objective lens, therefore, basically amounts to reducing λ and C_s while controlling Δz to match phases for the largest number of "beams" in the scattering distribution [7]. Attention must also be given to source coherence [8] and chromatic aberration [9] since these, too, serve to truncate the number of beams which can be used effectively during phase-contrast imaging.

Achieving Atomic Resolution in Close Packed Crystals

These combined effects impose limitations on the interpretation of phase-contrast images which are presently more severe than those imposed on their formation. Specifically, the point-to-point resolution of a microscope for directly

interpretable images is established by the Scherzer defocus condition [10] whereby the phases of the electron waves scattered by the specimen are uniformly controlled over the entire range of scattering vectors from $|\mathbf{u}| = 0$ to

$$|\mathbf{u}| = [0.66 C_s^{1/4} \lambda^{3/4}]^{-1} .$$

Another resolution limit is set, however, by the ultimate cut-off of spectra resulting from instrumental instabilities. This "information retrieval limit" [12] is characterized by fine detail in the image beyond the Scherzer limit where the phases of the electron waves scattered by the specimen are not uniformly controlled. For this reason the image is no longer "directly" interpretable; it must be viewed with accurate knowledge of the complete contrast transfer characteristics of the objective lens [12] and specimen conditions (thickness, orientation, etc.). One way of overcoming this limitation is to improve existing instrumentation. This was the course adopted by the Atomic Resolution Microscope (ARM) project [13,14].

INSTRUMENTATION

The ARM is a high voltage electron microscope which was built by JEOL [15] to achieve a point-to-point resolution consistently at or better than 0.18nm over its entire 400kV and 1000kV accelerating potential range. Consequently, the microscope can be tuned to a voltage which is below the threshold for knock-on damage in most specimens of interest and used to directly image its contiguous atom structure. Voltage is generated by a Cockroft-Walton system using pressurized Freon insulation, with AC and DC columns concentrically disposed about the evacuated accelerating tube, all housed within a single tank. Both ripple and stability are maintained within 1×10^{-6} per minute using highly sensitive

feedback compensation circuitry. The instrument also has a unique vacuum system to control contamination, and employs two sputter ion pumps of capacity 1000 l/sec each, backed by titanium sublimation pumps and turbomolecular pumps to maintain a pressure in the low 10^{-8} torr range throughout the electron-optical column and the accelerator tube.

To remove mechanical instabilities the microscope has been attached to a 100 ton inertial block and mounted on ten pneumatic isolators capable of dynamic leveling to within ± 0.001 inch from a preset value [Fig. 2]. The natural vibrational frequency of this system is 0.64Hz in the horizontal plane and 1.15Hz in the vertical.

Special attention has also been given to beam brightness in the ARM. At 1000kV and 25 μ A current, the LaB₆ cathodes in this microscope yield 10^8 amps/ cm^2 ---str., enabling 2 sec. photographic exposure times at $\sim 300,000\times$ magnification. This level of brightness is furthermore preserved through the viewing glass (made under special subcontract by Nikon Optics) due to its tailored transmission coefficient which is matched to the wavelength of the photons emitted from the screen phosphor.

The key to the variable-voltage, atomic resolution performance of the ARM is its top entry objective stage, which in addition to $\pm 40^\circ$ biaxial tilting, incorporates a height (z) control to alter specimen position along the optic axis over a 2mm range within the objective lens. Using the z-control to focus the specimen, the microscope can be operated at the appropriate objective lens current which maintains a constant $C_s \lambda$ product for any accelerating voltage. This principle is in fact put into operation automatically on the ARM; the instantaneous orientation of the specimen is furthermore displayed via a graphics software package on the monitor screen.

PERFORMANCE

The versatility of the high resolution, high angle goniometer of the ARM is demonstrated in Fig. 3. Moreover, the z-control stage enables the specimen to be lowered within the lens pole piece to such an extent that the pre-field of the objective can be used as a probe-former. In this way, convergent beam electron diffraction (CBED) patterns can also be recorded from the same specimen areas imaged at high resolution (Fig. 4), providing greater precision in diffraction studies through the analysis of higher order Laue zone (HOLZ) lines [16].

Another effect of the height adjustable stage is its influence upon the optical properties of the objective lens. In general, the lens spherical aberration coefficient decreases with increasing lens excitation although the exact functional variation is not a simple one [17]. At higher excitation, the lens focal length shortens, making it necessary to drop the specimen deeper into the focusing field. The most serious complication of this process is that the lens is physically constructed nearer the pole piece gap, and this in turn reduces the maximum tilting range of the specimen goniometer. In the ARM, the specimen in its lowest z position is "restricted" to an otherwise generous $\pm 25^\circ$ tilting range.

However, the primary purpose of the height control is to optimize lens performance. An example of its proper use is shown in Fig. 5, the computed phase contrast transfer function for the ARM at 1000kV operation with the specimen now located at an optimum 1.9mm above its deepest immersion position. This calculation reveals that with proper care the ARM is theoretically capable of 0.13nm point-to-point resolution.

Experimentally, the contrast transfer function of the ARM has been measured by the optical diffractogram method [18]. The 1MeV results are shown in Fig. 6, where the point-to-point resolution is demonstrated to be 0.16nm.

APPLICATIONS

The ARM accepts common 3mm diameter specimens which have been thinned to electron transparency by standard techniques and screened for suitability in another TEM. Three cartridges can be simultaneously loaded into the specimen airlock and individually withdrawn by a turret mechanism for rapid insertion into the goniometer stage. Once in position, focusing and astigmatism correction are carried out while viewing the image directly on the screen phosphor. Through-focus series are facilitated by infinite-turn, continuously variable potentiometer control of the objective lens current, with a minimum focus increment of 0.3nm. In addition, the stigmator coils are fed by six independent CPU channels, permitting different reference settings to be stored in memory for rapid recall. Operation is further simplified by digital readout of all relevant lens and deflector coil excitation values at the console. Specific data concerning film number, operator code, magnification or camera length, and a 14 character text assigned by a user terminal are also recorded in the margin of each negative, of which there are 50 per camera load.

Initial Applications of the ARM

At the time of this writing the ARM has just become available for research applications. Figure 7 shows an example of the structure and interface of precipitates in an Al-20% Ag alloy. In Fig. 7b the ledge mechanism can be seen at the single planar level. Figure 8 shows the potential of the ARM for distinguishing between different atomic species in a specimen--in this case barium titanate. Here the computed image and experimental image are in very good agreement. Figure 9 is an example from a study on precipitation in a Mo-Hf-N alloy [19]. This alloy is a dispersion of HfN on {001} planes of the Mo matrix. HfN has the NaCl crystal structure and is related to the bcc Mo matrix by the Bain correspondence which for

this particular combination of lattice parameters describes an invariant plane strain, a simple extension normal to the precipitate habit plane. Like other precipitates formed by an invariant plane strain transformation, HfN has been reported to grow by a ledge mechanism. The large volume change of about 40% that accompanies the transformation must be accommodated by lattice vacancies. If stacking violations of {001} planes are to be avoided, vacancies must condense in double layers. This will place discrete limits on the optimal precipitate thicknesses and ledge heights. An example of an image in the symmetrical $\langle 100 \rangle_{\text{Mo}}$ orientation is shown in Fig. 9. The thick plate of fcc HfN is viewed precisely edge-on and is readily distinguished from the Mo matrix. Two features are clearly evident: (1) the precipitate-matrix interface is atomically smooth and (2) the precipitate contains no faults in the stacking sequence. This evidence supports the proposed model and the resulting plate is predicated to have an 8% interstitial strain field.

SUMMARY

Electron microscopy is now over 50 years old. It is of central importance in the field of materials science and engineering, i.e., in the solving of fundamental scientific, as well as practical-technological problems. Consequently, progress in the applications of this subject to understanding the structure, morphology and composition of materials has been very rapid, so much so that it is almost impossible to keep track of all the research currently underway. This paper does not attempt to review the field but covers some typical research projects currently being investigated at Berkeley involving high resolution.

The ARM is just in its infancy but it has proved its unique capability for superior resolution. The demonstrated resolution of $1.8\text{\AA}$ at 400kV on the ARM

exactly overlaps with the $1.8\text{\AA}$ resolution measured at 400kV on the JEOL 4000EX by Hashimoto, a pioneer in HREM [20] and whose work along with others especially in Japan has been an inspiration for this project. It can be forecast that a new wave of data will be generated by such atomic resolution microscopes and the information explosion will continue.

Acknowledgements

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Figure Captions

Fig. 1. View of the NCEM showing the two high voltage microscope silos. The ARM is on the right.

Fig. 2. Showing the support system to provide anti-vibration and earthquake proofing to a shock of 7.8 on the Richter scale.

Fig. 3. Tilting experiments to show the full crystallographic range of orientations possible at all voltages in the ARM (specimen: silicon).

Fig. 4. Central disc of CBED pattern from [011] silicon specimen.

Fig. 5. Computed phase contrast transfer function for the ARM, assuming 1000kV accelerating voltage, $C_s = 2.3\text{mm}$, $C_c = 3.4\text{mm}$, defocus = 52nm (Scherzer condition), and a disc-shaped source with a divergence half angle of 0.5mrad and 2eV energy spread. The theoretical resolution at first crossover of the zero axis is $1.3\text{\AA}$. The value at 400kV is $1.7\text{\AA}$.

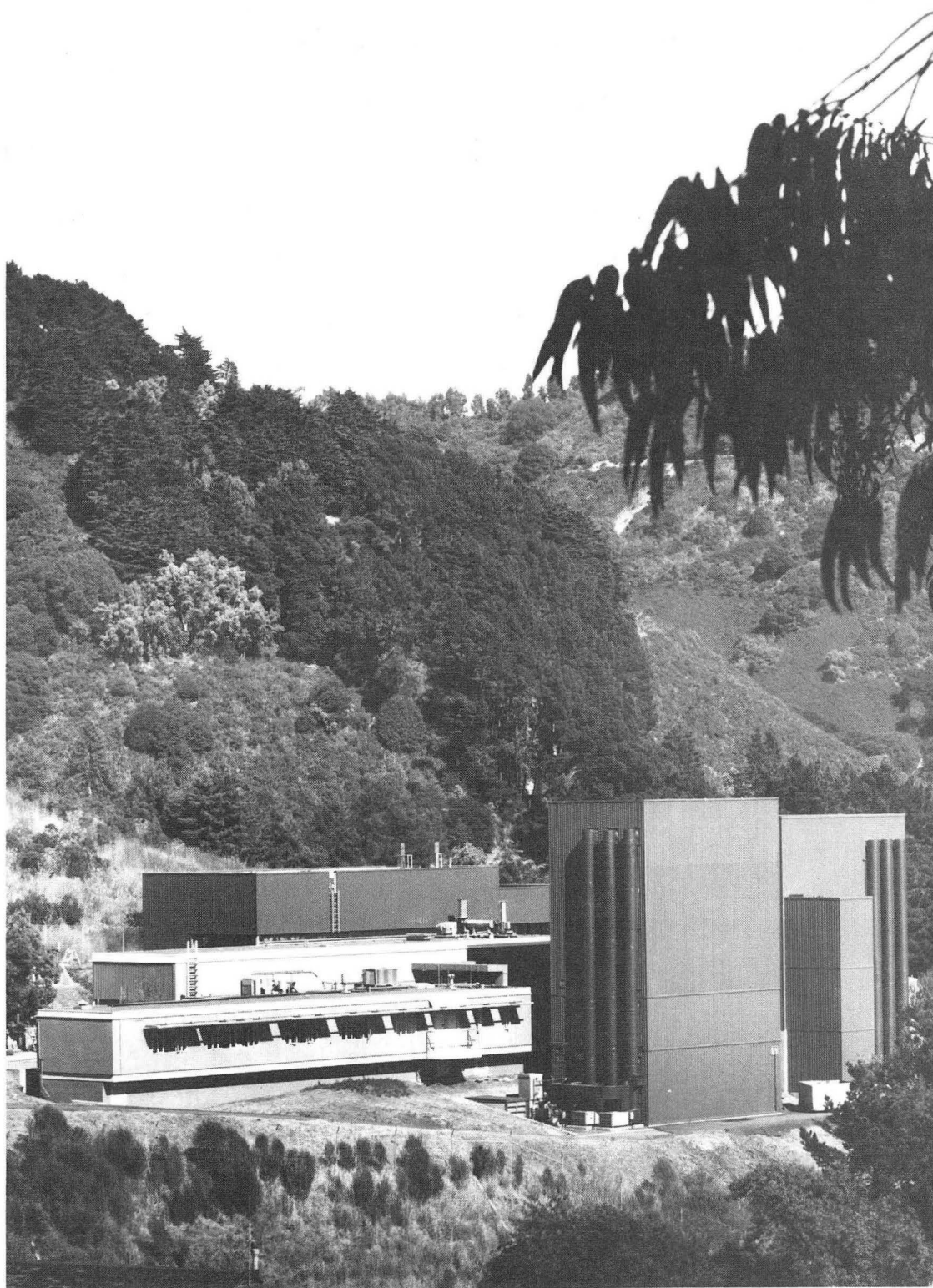
Fig. 6. Optical diffractograms from high resolution images of (a) gold particles showing discrete 0.23, 0.20 and 0.14nm reflections (left) and (b) amorphous Si (right) at Scherzer defocus, 1000kV. The diffractogram halo in (b) is continuous out to a resolution limit of 0.16nm.

Fig. 7. (a) Atomic resolution image of a four atom layer γ' platelet in an Al-Ag alloy. The long particle dimension is 15nm. (b) Atomic resolution image of

an impingement event between γ' platelet and large ordered γ plate in an Al-Ag alloy. The impingement region again shows four atom layer stacking (arrow), demonstrating the role of Shockley partial dislocations in the growth process. Here the ledge mechanism is observed to be operable at the single atomic plane level.

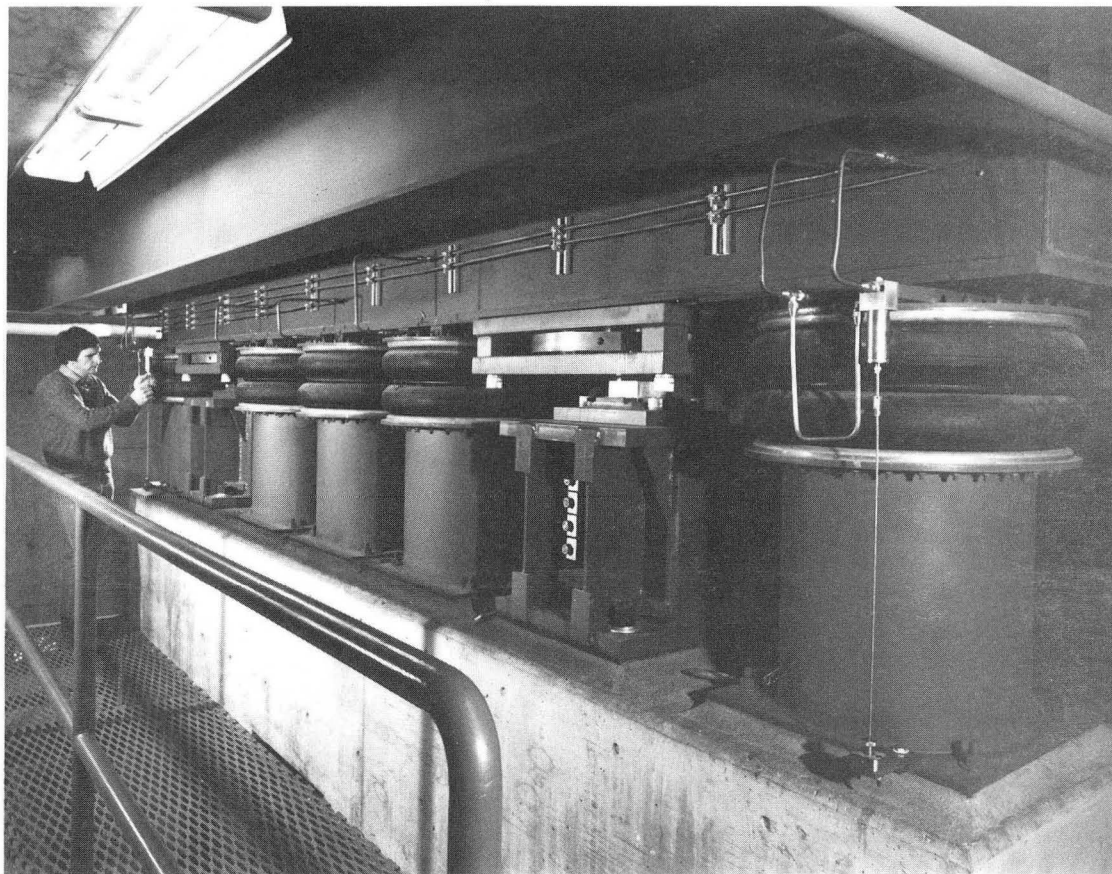
Fig. 8. (a) Ionic positions, (b) diffraction pattern and (c) atomic resolution image of barium titanate in [001] orientation. Powdered crystallites were suspended on a holey carbon film; goniometer was used to tilt one crystallite which extended beyond film edge for this image. (d) Simulated image ($160\text{\AA}$ thick crystal) at energy of 1MeV, objective lens defocus and C_s of -90nm and 2.6mm. (Courtesy of Dr. M. A. O'Keefe).

Fig. 9. High resolution image of HfN precipitate plate in a molybdenum matrix. The precipitate is seen to be three unit cells thick (see outlines), free of stacking faults, and bounded by an atomically smooth interface.



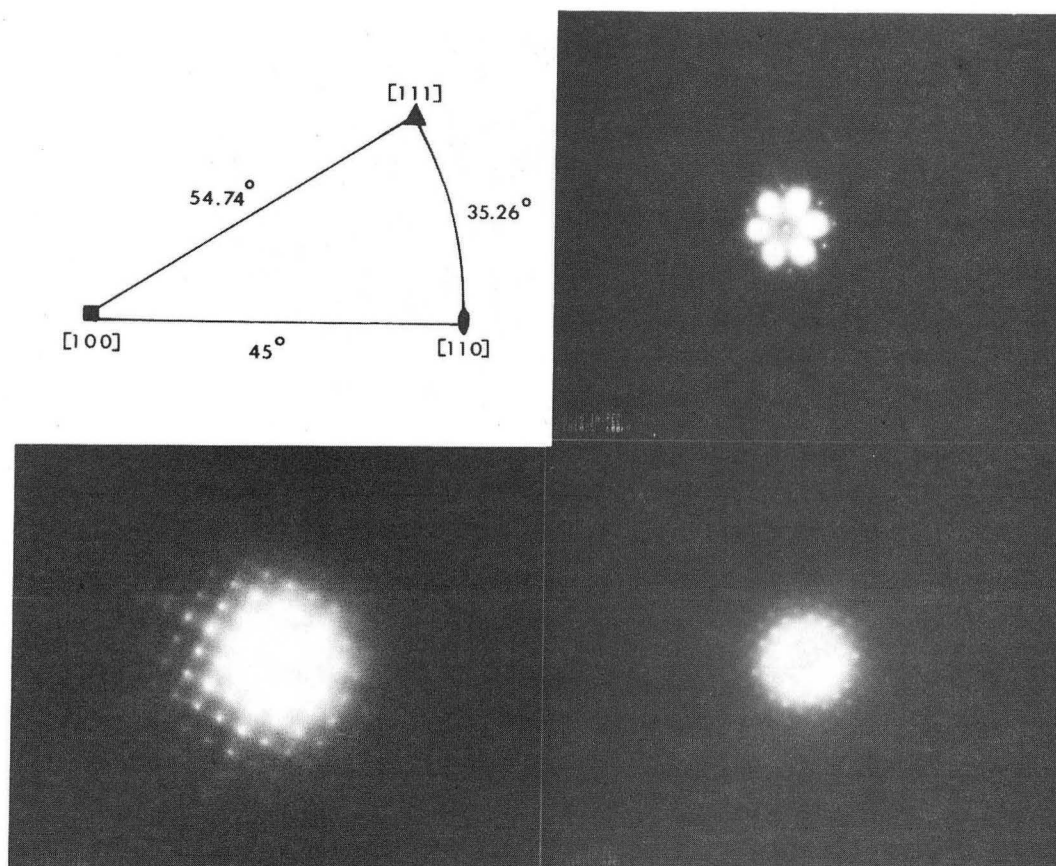
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Fig. 1



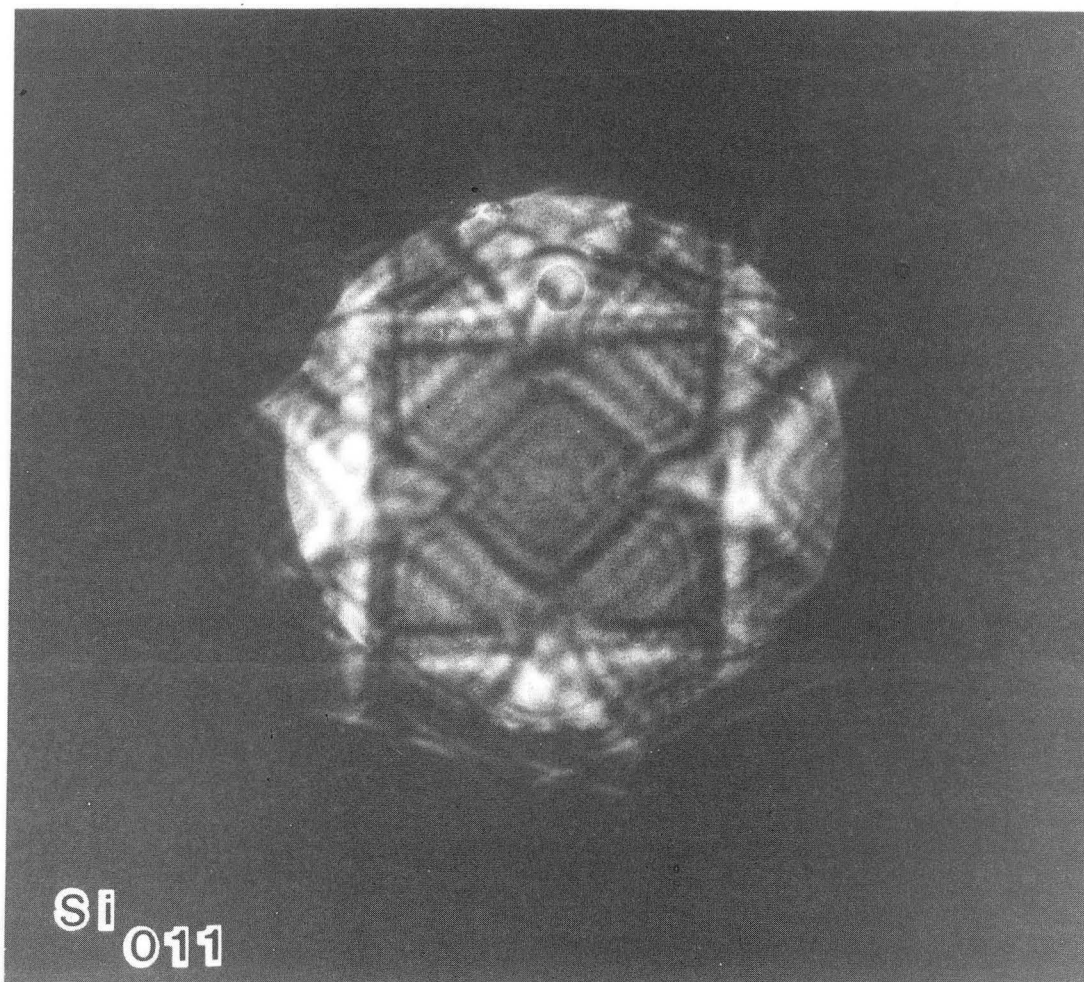
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Fig. 2



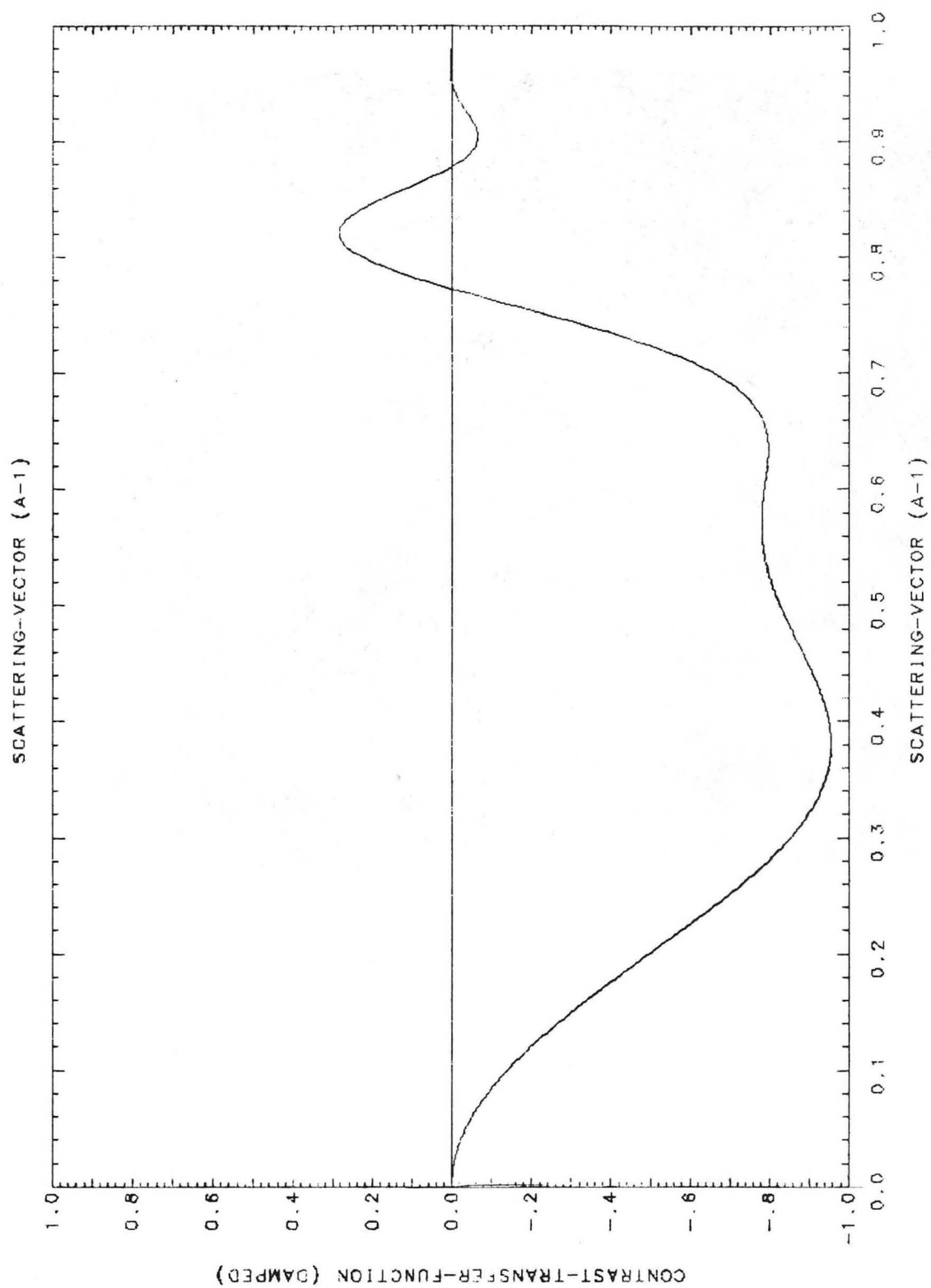
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Fig. 3



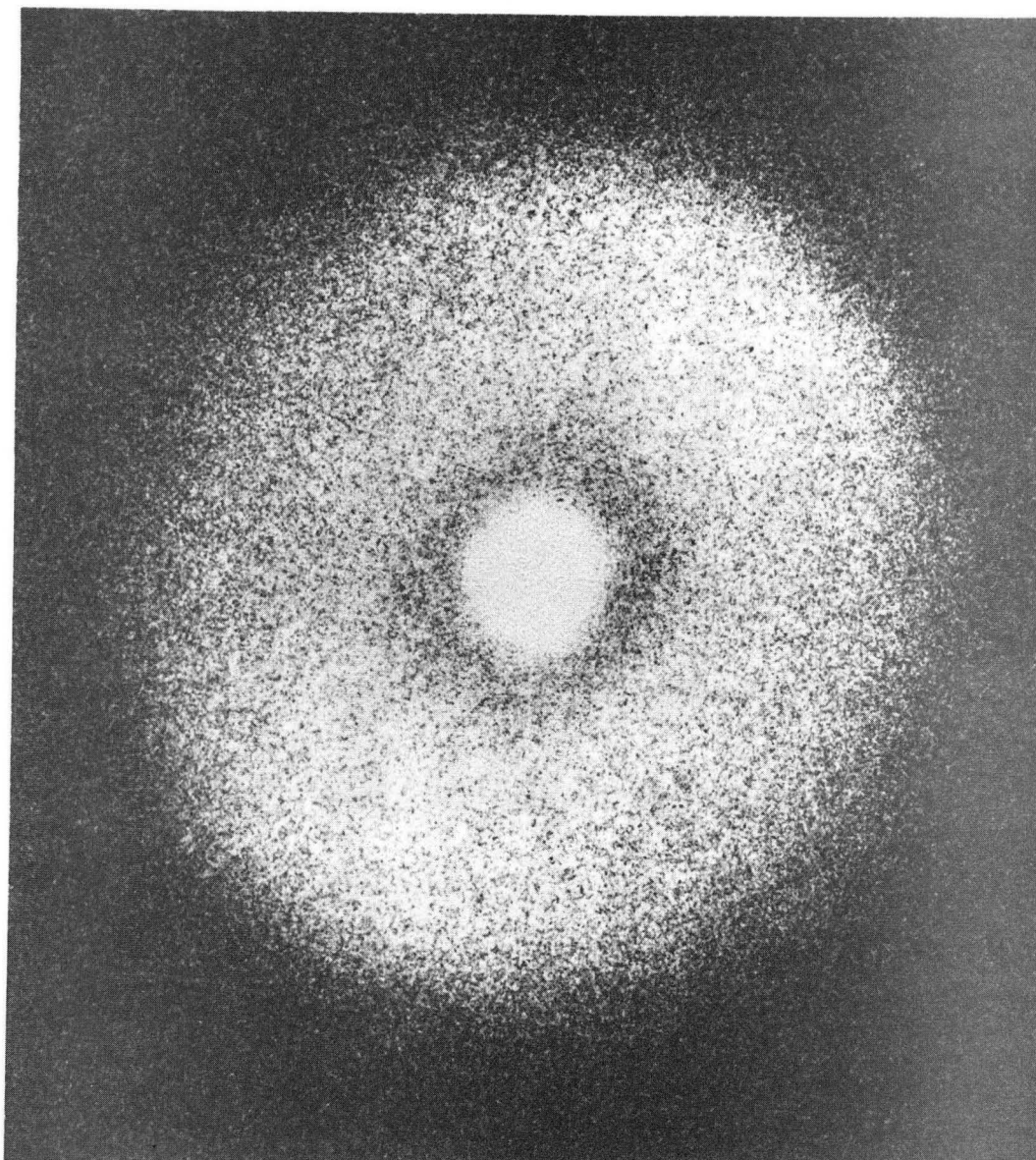
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Fig. 4



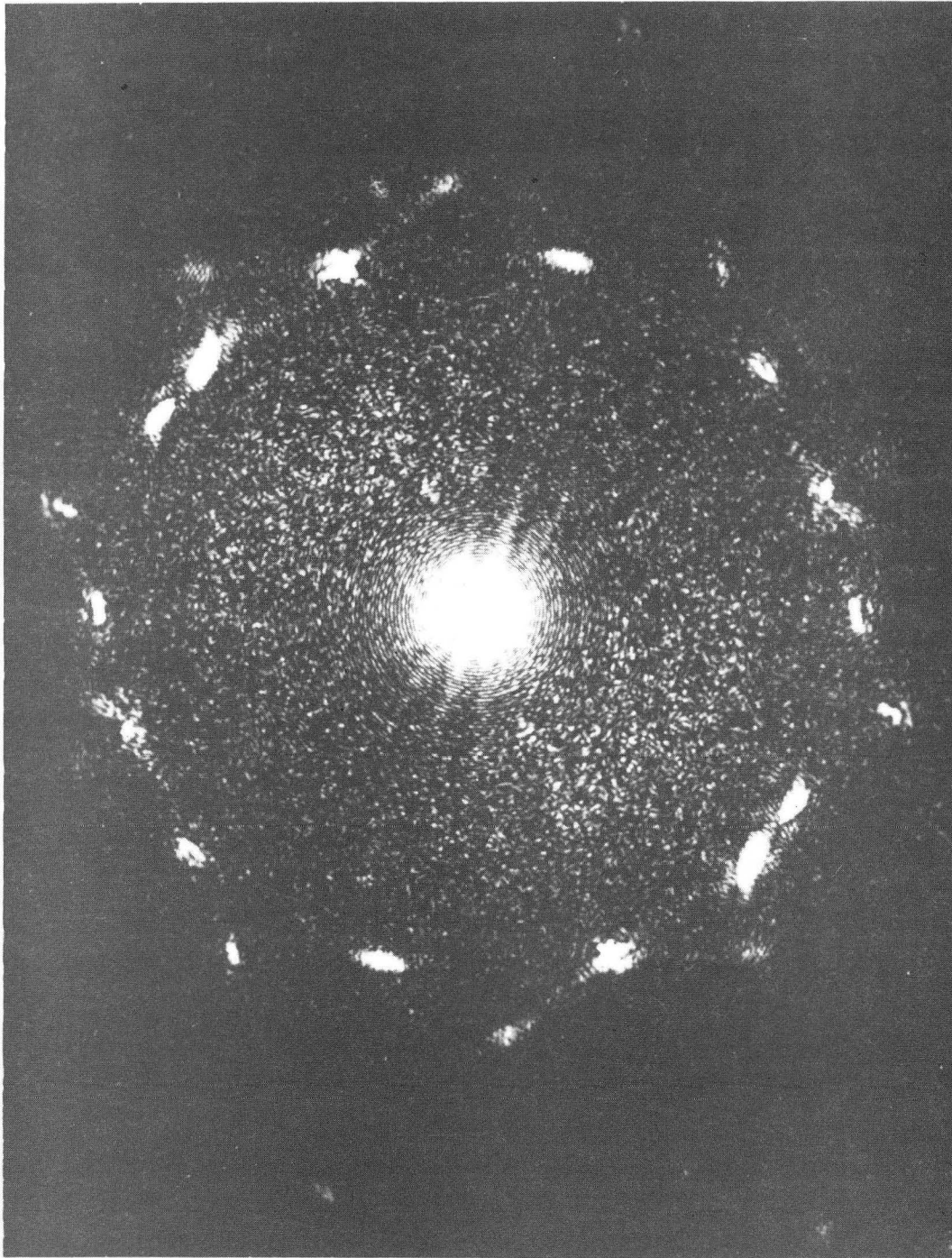
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Fig. 5



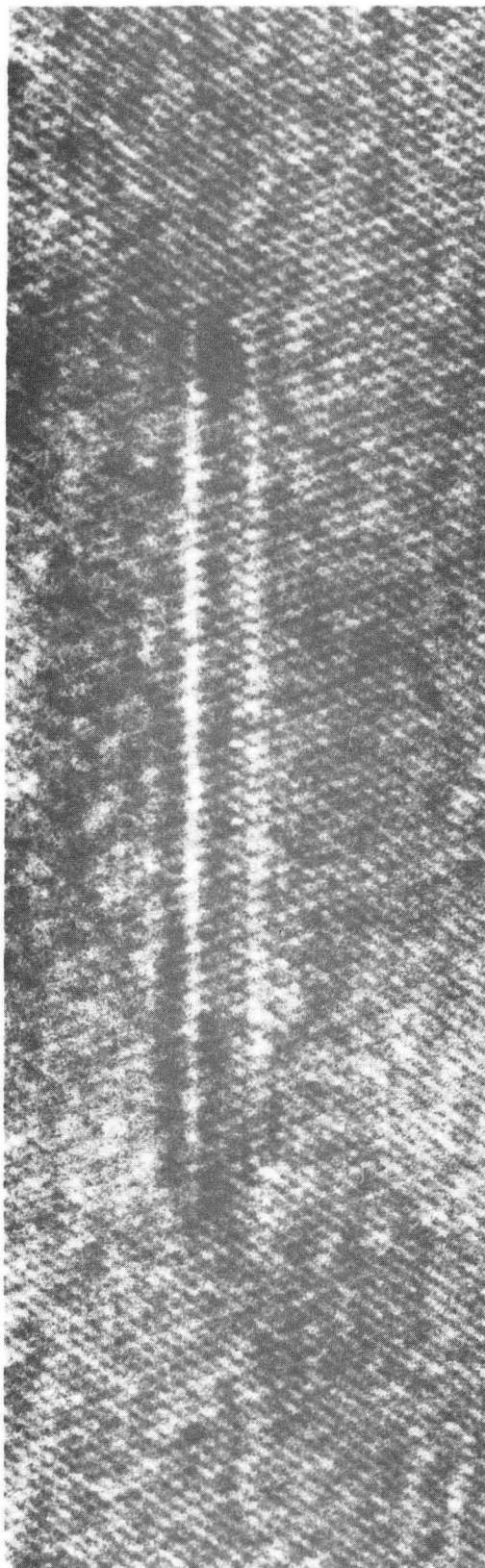
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Fig. 6a



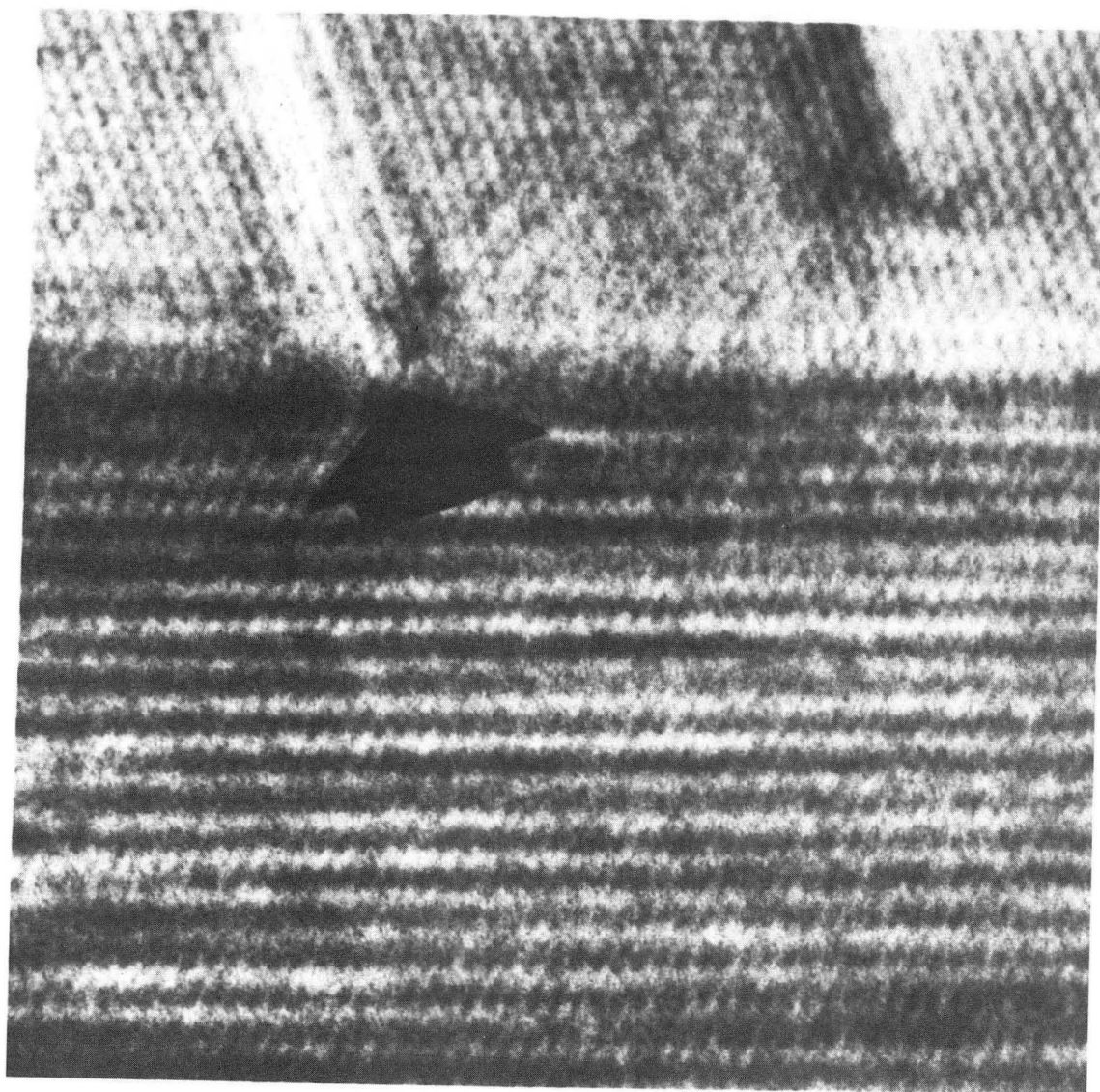
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Fig. 6b



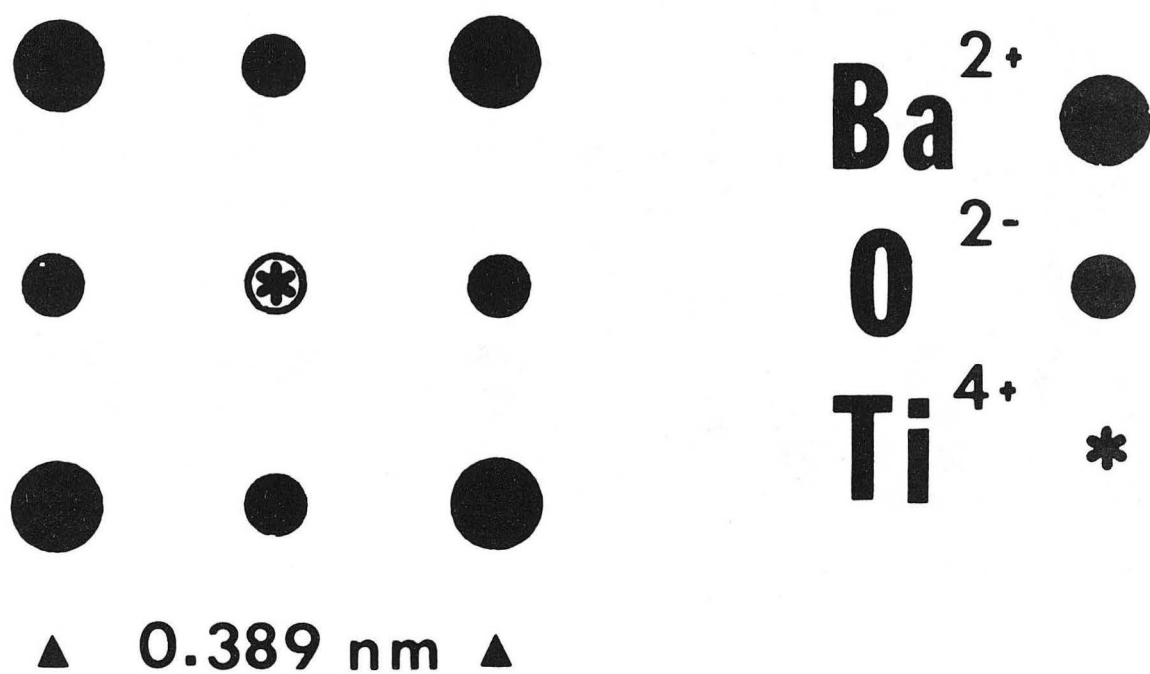
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Fig. 7a



XBB 836-5041

Fig. 7b



XBL 835-9630

Fig. 8a

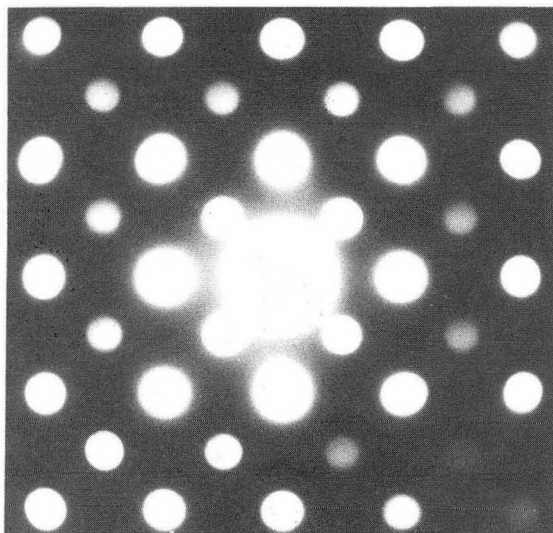
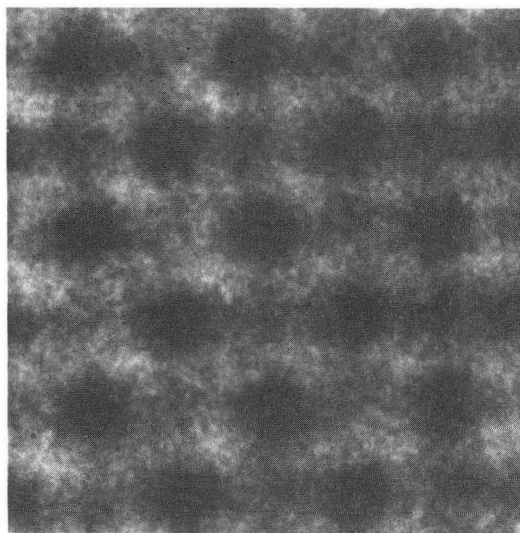
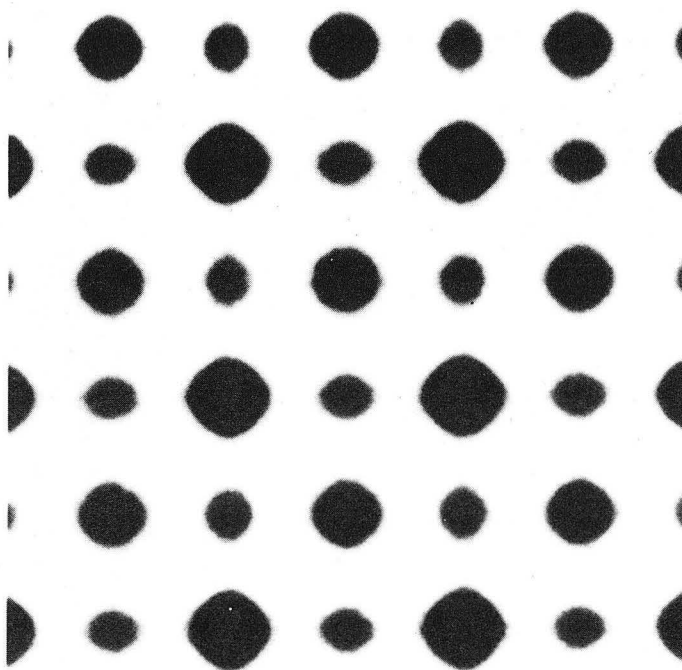


Fig. 8b



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Fig. 8c



XBB 843-2552A

Fig. 8d

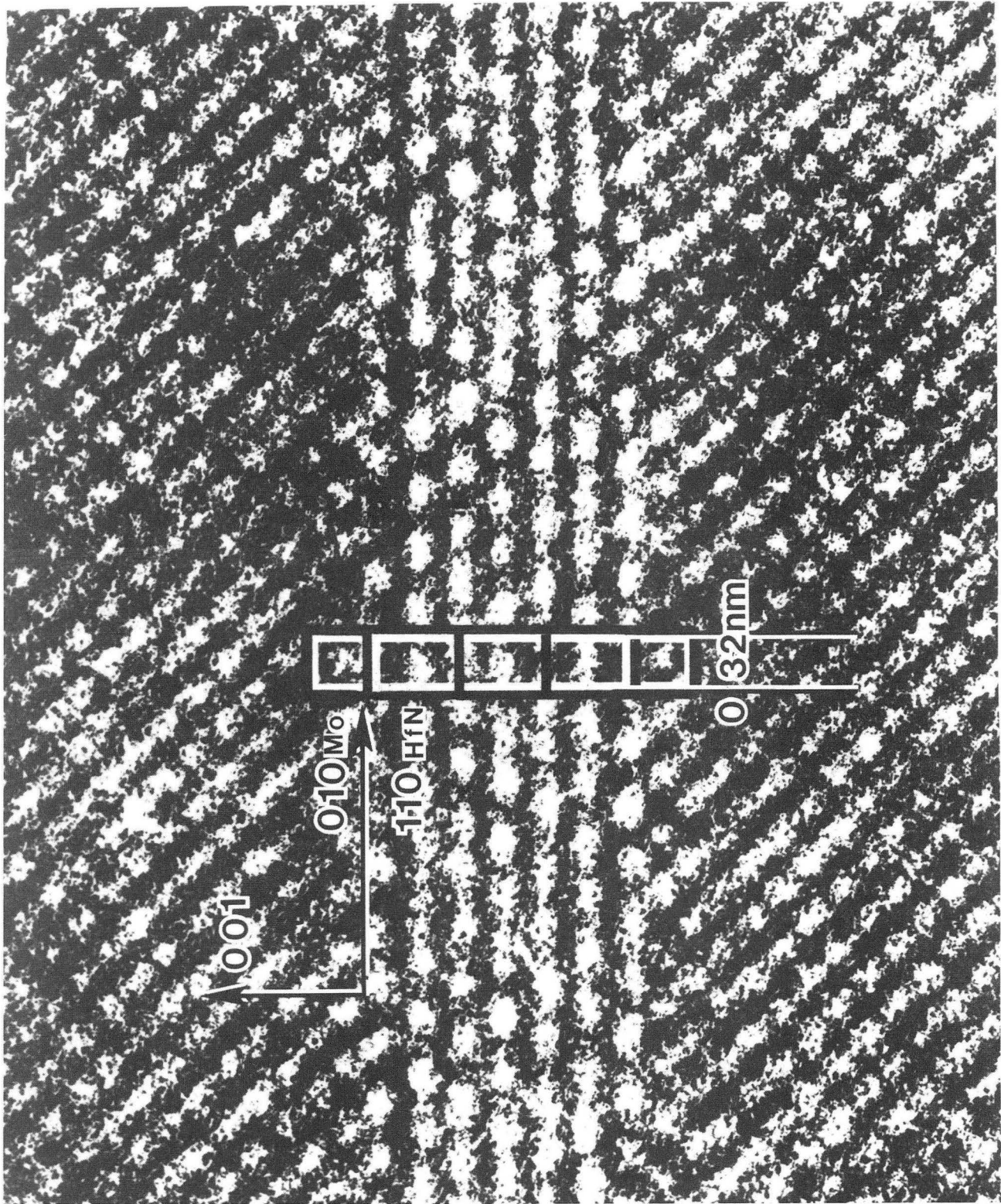


Fig. 9

XBB 839-7867

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