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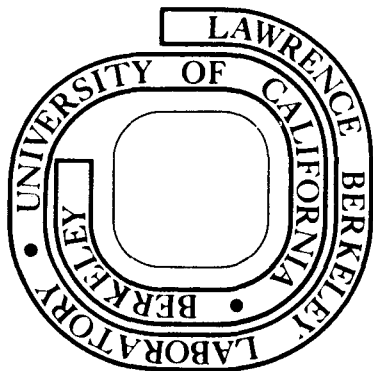
Richard Dalven

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HIGH RESOLUTION TRANSMISSION ELECTRON MICROSCOPY

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Over the past 15 years transmission electron microscopy (TEM) has played a particularly important role in the growth of materials science. It provides almost all the data needed to characterize the microstructure of materials: morphological from high magnification micrographs, crystallographic from selected area diffraction patterns and chemical from electron microprobe or velocity analysis. From correlation of the microstructure with properties the faults of materials have been isolated and the benefits of particular structures recognized. Consequently, it has become possible to design materials for a given application from first principles.

The quest by the microscopist for higher resolution to solve microstructural problems has resulted in the present generation of transmission electron microscopes. Lattice planes as closely spaced as 0.1 nm can now be resolved and a number of techniques have been developed which take advantage of this improved performance. The purpose of this article is to illustrate some of the recent advances in high resolution TEM with special reference to their relevance in materials science. (See references 1-4 for detailed information concerning electron microscopy and its applications).

CONVENTIONAL ELECTRON MICROSCOPY

The most common mode of operation of the microscope is the formation of an image utilizing only the transmitted electron beam (bright field image) or one diffracted beam (dark field image). Selection of the desired beam is achieved with a small aperture situated in the back focal plane of the objective lense.

Magnifications up to 600 000 times at the viewing screen can be achieved by modern microscopes, although it is rarely necessary or in fact desirable to operate under these extreme conditions. Smaller real areas of the specimen are imaged, intensity is weak resulting in longer exposure times and the requirement of ideal focusing and astigmatism correction becomes more stringent. Further magnification can be achieved by photographic methods and it is generally believed that a point separation of 0.05-0.1 mm on the photographic plate is sufficient for most resolution purposes. Thus for a resolution of 0.5 nm at the specimen, a magnification by the microscope 100,000-200,000 times is often adequate.

The considerable progress in materials science has come from understanding the imperfections in a material are related to its properties. Since much of the important structural detail is finer than 10 high resolution electron microscopy is invaluable. Contrast of imperfections in the microscope arises from either local variations of electron scattering power in the crystal (e.g. composition differences between precipitate and matrix) or local variations of lattice spacing and orientation which affect the diffraction conditions (e.g. near the core of a dislocation). Detailed series of the bright and dark field images under controlled specimen orientations are often necessary to characterize the defects, and

a systematic set of micrographs from the same area is normally more valuable than random high magnification pictures of several areas.

Examples of the role that TEM has played are indeed numerous, and some of the more important developments are described in references 1-4. The present experimental investigation at Berkeley into the complex nature of structural steels clearly exemplifies the application of conventional TEM. These studies have led to several suggestions for the design of martensitic and bainitic steels which are at once strong, tough and economical (5). For example at the same strength level twinned martensite has generally been found to possess inferior fracture toughness to dislocated martensite. Hence the constituents of a strong, tough martensitic steel should be those in which twinning can be avoided (i.e. carbon contents less than 0.4% minimal manganese content) and an iron-4% chromium-0.35% carbon steel has been developed to meet this requirement.

THE WEAK BEAM TECHNIQUE

One of the many problems in imaging small objects is no longer that of instrumental resolution but of obtaining sufficient contrast from the object to render it visible. The "weak beam technique" not only enhances contrast but also produces a sharper image (7).

Normally during the characterization of defects by TEM the specimen is carefully oriented so that only one beam is strongly diffracted (i.e. close to the Bragg reflection position). The electron beam in this condition is very sensitive to small strain fields and the image width is often much broader than the disturbance center (e.g. the image of a

dislocation is ~ 10 nm whereas the core may only be 0.3-0.5 nm wide). In the weak beam technique, the specimen is imaged in dark field using a reflection which is far from the Bragg position for regions of perfect crystal. The image is very dim but where it is locally under large strains the lattice planes may be distorted sufficiently to be near the Bragg position for the chosen reflection, producing a bright image. Contrast is high and the corresponding image width narrow (e.g. typical dislocation images are ~ 1.5 nm by this technique).

This method is a particularly important development for the study of dislocations and their interaction with defects (e.g. pp 1-22 of reference 4). Dislocations as closely spaced as 3.0 nm can be readily resolved, and the radiation damage produced in ion-implanted silicon is now being successfully studied at Berkeley (8).

LATTICE IMAGING

At present, one of the more controversial areas of TEM concerns the interpretation of "lattice images". These are produced when the transmitted beam and one diffracted beam (or sometimes more than one) are allowed through the aperture to form the image. The resultant image is composed of a set of fringes both parallel to the lattice planes in the crystal which give rise to the chosen reflection and of spacing equal to the separation of these lattice planes. By analogy with the Abbé principle in optical diffraction theory, it may be shown that the fringes represent the projection of the lattice planes. Thus, any distortions revealed by the fringe pattern should correspond to

real distortions in the crystal (3). However this simple treatment of lattice images has been criticized on the grounds that the more complex dynamical theory of electron diffraction must be invoked, yielding results which in general tend to invalidate the above argument (9). Whilst it was shown that interpretation of lattice images for faults inclined to the imaged planes must be treated with caution, it is still not certain whether a one to one correspondence between planes and fringes is invalid for faults parallel to the planes.

Although the meaning of the fringes is far from clear the first results of this method are extremely encouraging for its application to the study of crystal defects. The disturbance of the lattice image near a low angle grain boundary has been compared with the excellent fit of the fringes at coherent twin boundaries (3); the distortions at G.P. zones in aluminum copper alloys have been shown to be consistent with a zone model deduced from x-ray diffraction results (10); and more recently the present author has demonstrated that the out-of-phase nature of antiphase boundaries in ordered copper-gold alloys is revealed by this technique. It appears that this may become a viable and important mode of TEM in the study of imperfections in materials.

SELECTED AREA DIFFRACTION

As an extension of its role in deriving crystallographic information, the selected area diffraction technique also has considerable importance in providing information about very small heterogeneities. The shape of diffraction spots is related to the shape of the object producing them and examination of the diffraction pattern may yield information not evident in the high resolution electron images. For example streaked diffraction spots arise from thin disc shaped precipitates. Often in strong alloys the density of distribution of the strengthening particles is too high for individual precipitates to be observed (e.g. in copper-beryllium alloys) and the supplementary information in the diffraction pattern proves invaluable.

As noted previously the contrast of a small object may be insufficient to allow it to be seen in the image but carefully taken diffraction patterns may indicate the presence of an inhomogeneity. Recent work on short range order in a variety of nickel and gold based alloys is an excellent example of the use of diffraction patterns in this way (11). By thoroughly overfocusing the illumination of the specimen and, to compensate for the correspondingly weak electron signal, photographing the patterns with a long exposure time the authors found weak superlattice spots hitherto undetected. Consequently they were able to deduce the presence of small microdomains (approximately 1.5 nm) of order in the disordered state of the alloys.

CHEMICAL ANALYSIS AT HIGH RESOLUTION

The composition of small regions may be obtained from suitably modified microscopes, thus allowing direct correlation of composition variations with microstructural features in the image. To date the highest resolution attained by such techniques is ~ 10 nm and useful data is now being collected concerning for example segregation of constituents near precipitates or other defects in materials. This is proving to be particularly important to the study of grain boundaries, which are often the cause of premature failure due to brittleness.

In the energy analyzing electron microscope the spectrum of the energies of electrons leaving the specimen is determined. Energy loss peaks, which are related to the local composition, occur due to interactions between the high energy electrons traveling through the specimen and the electrons in the material. Whilst the method is confined only to the lighter elements, a variety of aluminum alloys are being studied as examples of particular metallurgical microstructures (e.g. pp 188-221 of ref. 4).

The Electron Microscope with Micro-Analysis (now available commercially through AEI as EMMA-4) has been developed from the larger scale electron microprobe, utilizing the characteristic x-rays generated by the electron beam striking the specimen. The beam can be focused to about 100 nm and the x-rays emitted from this small area analyzed by crystal spectrometry. Although the spatial resolution of this instrument is not as good as for the energy analyzing microscope, its range of application is much wider. Preliminary results (pp. 222-235 of ref. 4) have already aided in elucidation of microstructural changes occurring during superplastic

deformation of zinc-aluminum alloys and of the effect of grain boundary segregation on stress corrosion of aluminum-zinc-magnesium alloys.

HIGH VOLTAGE ELECTRON MICROSCOPY

The application of TEM to the study of materials has greatly expanded since the recent development and availability of high voltage electron microscopes. As non-metallic materials are extremely difficult to prepare as thin foils, the increased penetration with increasing energy particularly allows study of such thicker samples. Metallography is thus progressing into a wide range of materials at voltages in the range of 1 million volts compared with the normal operating voltage of 100,000 volts.

Recent work at Berkeley into the vapor deposition of epitaxial layers of gallium arsenide doped with tellurium onto gallium arsenide illustrates the advantage of the penetration (12). Combinations of such layers are used in making pulse generating devices but electrical breakdown is known to occur at the epitaxial interface. High voltage TEM showed the cause of breakdown to be defects at the interface from which dislocations and dislocation tangles emanate. The defects were subsequently identified by microprobe analysis to be silicon and thus by ensuring the absence of silicon during the evaporation failure of the devices may be averted.

HIGH RESOLUTION HIGH VOLTAGE ELECTRON MICROSCOPY

At high voltages it is not normally possible to operate under the two beam condition which is commonly used at 100 kV. Several reflections are simultaneously closer to their exact Bragg position. Certain advantages can be gained from this, especially if the specimen is oriented so that a low index systematic row of reflections is excited (i.e. all the reflections lie in a row in reciprocal space and thus have indices in the same ratio: (hkl) , $(2h2k2l)$ etc.)

As the specimen is tilted so that higher order reflections of the systematic set are excited the image width of defects in the bright field image reduces considerably (12). However the contrast of the defect also decreases so that a compromise must be met at intermediate reflections to optimize resolution and visibility. Nevertheless the improved clarity of the image can be quite striking and is often comparable with that of the weak beam technique (13). Although originally developed under high voltage conditions a systematic row orientation can be produced at 100 kV. Thus with the ease of focusing a bright field image and the shorter exposure times involved it may become an attractive alternative to weak beam imaging.

An exciting development in high voltage electron microscopy has evolved since the discovery of the "critical voltage effect". For particular incident electron energies the intensity of the second order Bragg reflection in a systematic row is a minimum and the critical voltage (V_c) at which this occurs can be determined by suitably varying the accelerating voltage during microscopy. V_c depends on the electron scattering factors of the first and second order reflections of the set

and on the unit cell dimensions of the material, and is thus sensitive to any inhomogeneity which locally changes these parameters. It has already been used to study segregation effects in alloys (e.g. V_c for the (400) reflection of Nickel decreases from 600 kV to 200 kV on the addition of 30% gold), lattice parameter changes, degree of order of partially ordered alloys, and also to determine accurate atomic scattering factors and Debye temperatures (14 and pp 23-59 of ref 4).

Furthermore it has also been shown that dark field imaging under critical voltage conditions enhances the contrast and reduces the image width of defects which locally alter V_c . The possibility of readily deriving both chemical and microstructural information is provided by this technique and it is one which should assume greater importance over the next few years.

CONCLUSIONS

Several new techniques in transmission electron microscopy have been described, both for operation at conventional and at high voltages. These considerably diversify the study of materials at high magnification and render the correlation of microstructure and properties a more precise science. In the future TEM should continue in its importance to the materials scientist and the emphasis of microscopy should become increasingly more positive in its approach to the design of materials.

ACKNOWLEDGMENTS

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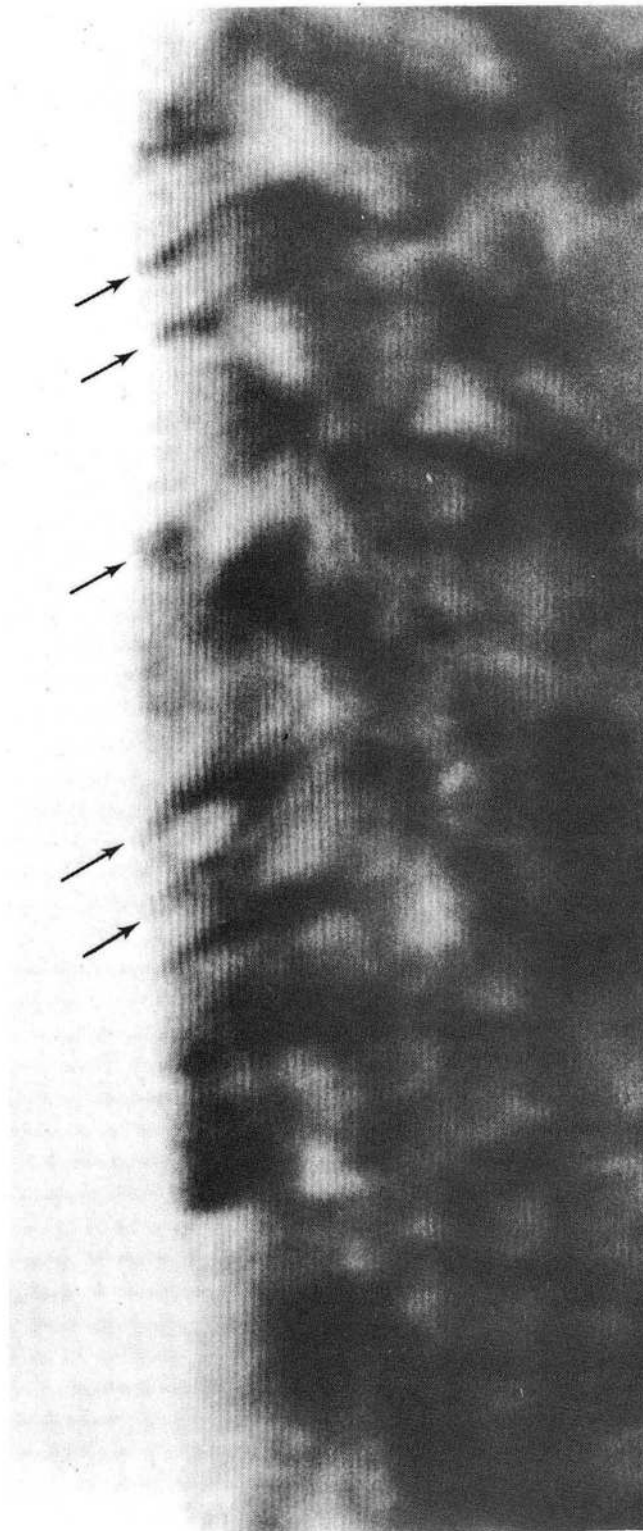
The author

Robert Sinclair is a National Science Foundation postdoctoral research fellow at the Department of Materials Science and Engineering, College of Engineering, University of California at Berkeley. He gained the degrees of B.A., M.A. and Ph.D. in Materials Science at the University of Cambridge, England and was a postdoctoral research associate at the University of Newcastle-upon Tyne, England for 18 months before taking up his present position.

FIGURE CAPTIONS

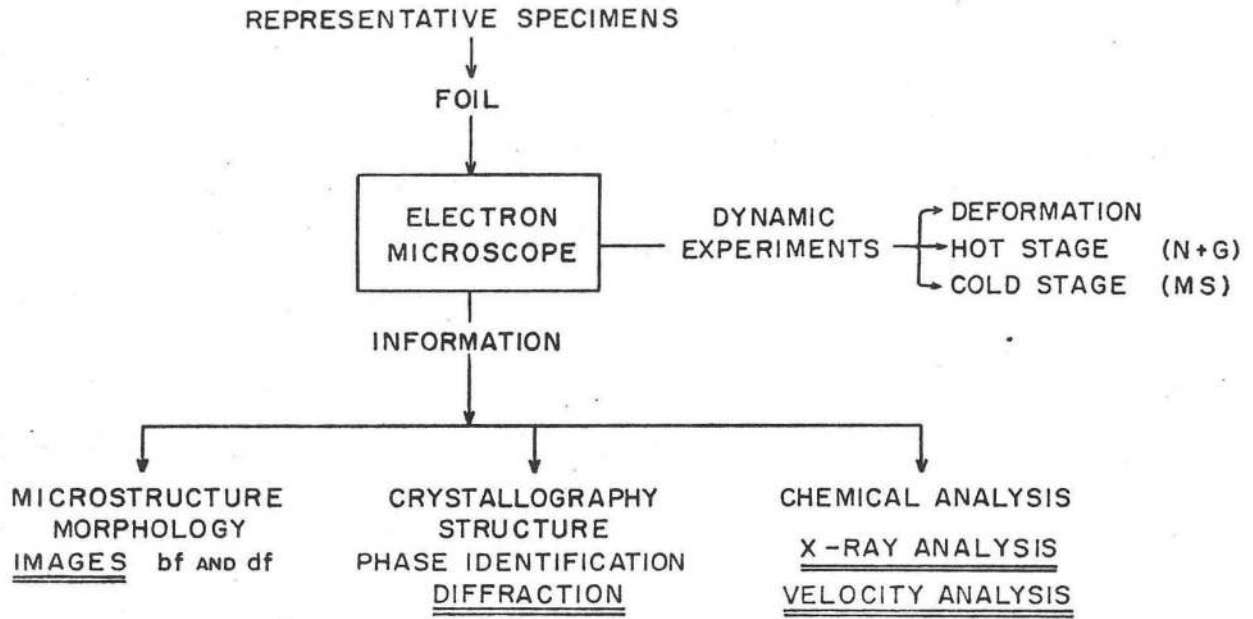
- Fig. 1. A lattice image of gold, showing resolution of the (200) planes spaced 0.205 nm apart. (Magnification
- Fig. 2. A schematic diagram illustrating the main functions of the transmission electron microscope.
- Fig. 3. Fracture toughness versus yield strength for Fe-Ni-Co-C martensitic steels. Low fracture toughness is associated with twinned martensite, high fracture toughness with the dislocated martensite as indicated by the inset micrographs. (Courtesy ASM, ref. 6).
- Fig. 4. Bright field (a) and weak beam dark field (b) images of ion-implantation damage in the same area of a silicon specimen. The improvement of the weak beam image allows the crystallographic nature of the defects to be clearly recognized. (Courtesy Radiation Effects, ref. 8).
- Fig. 5. Lattice image of periodic antiphase domain boundaries (APB's) in ordered copper gold. The superlattice (110) planes (spacing 0.276 nm) abutt at each APB illustrating their antiphase nature (Magnification
- Fig. 6. Selected area diffraction patterns of disordered Ni_3No showing the presence of weak superlattice reflections. (a) [001] pattern (b) [110] pattern. (Courtesy Acta Metallurgica, ref. 11).
- Fig. 7. High voltage bright field electron micrographs (at 650 kV) of dislocations emanating from faults, at epitaxial interfaces in GaAs electrical devices (Courtesy R. Osiecki, ref. 12)

Fig. 8. High voltage dark field image of dislocations in aluminum-7% magnesium taken at 400 kV, the critical voltage conditions for the (222) reflection (courtesy W. Bell and G. Thomas, pp. 23-59 of ref. 4).



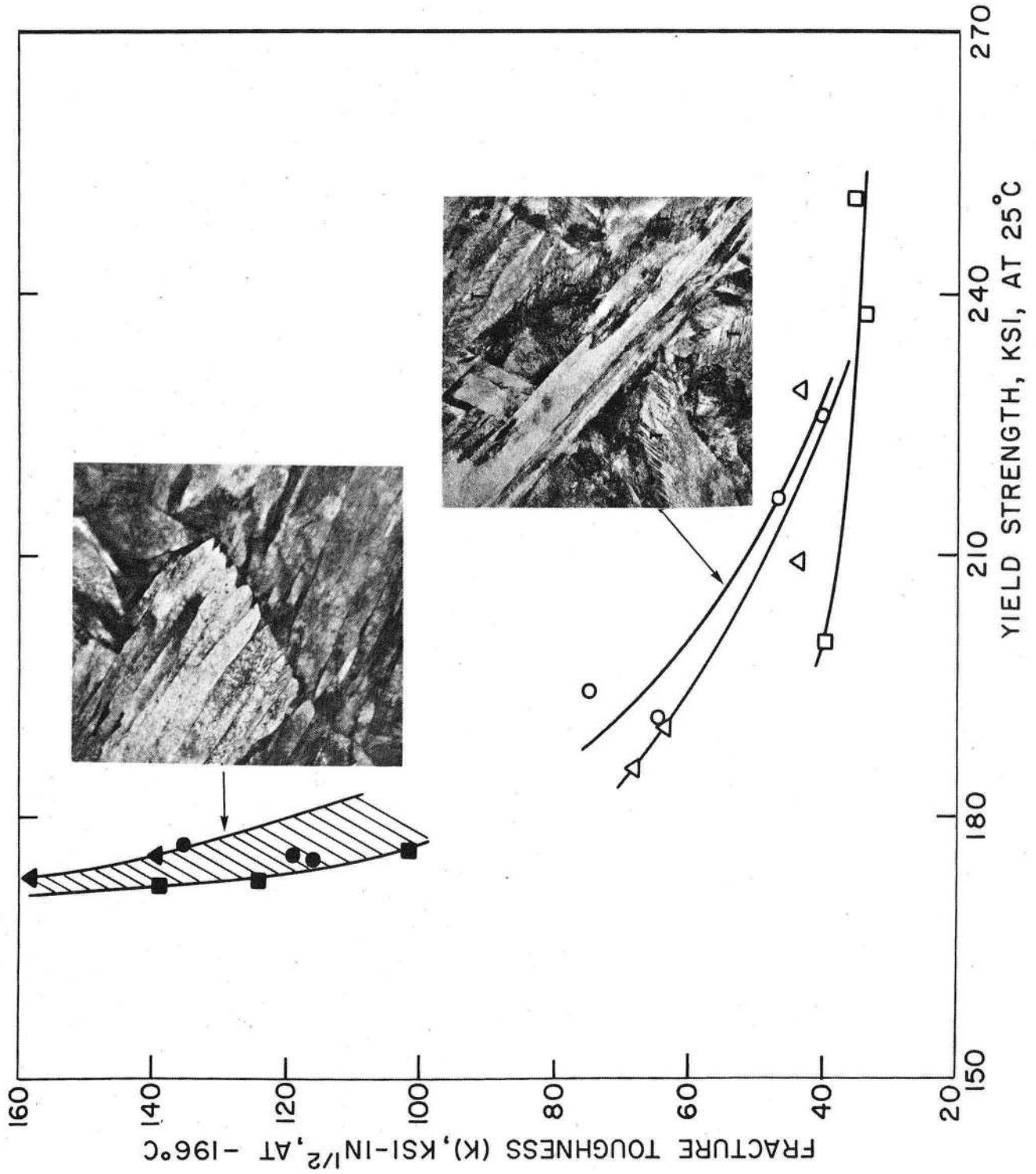
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Fig. 1



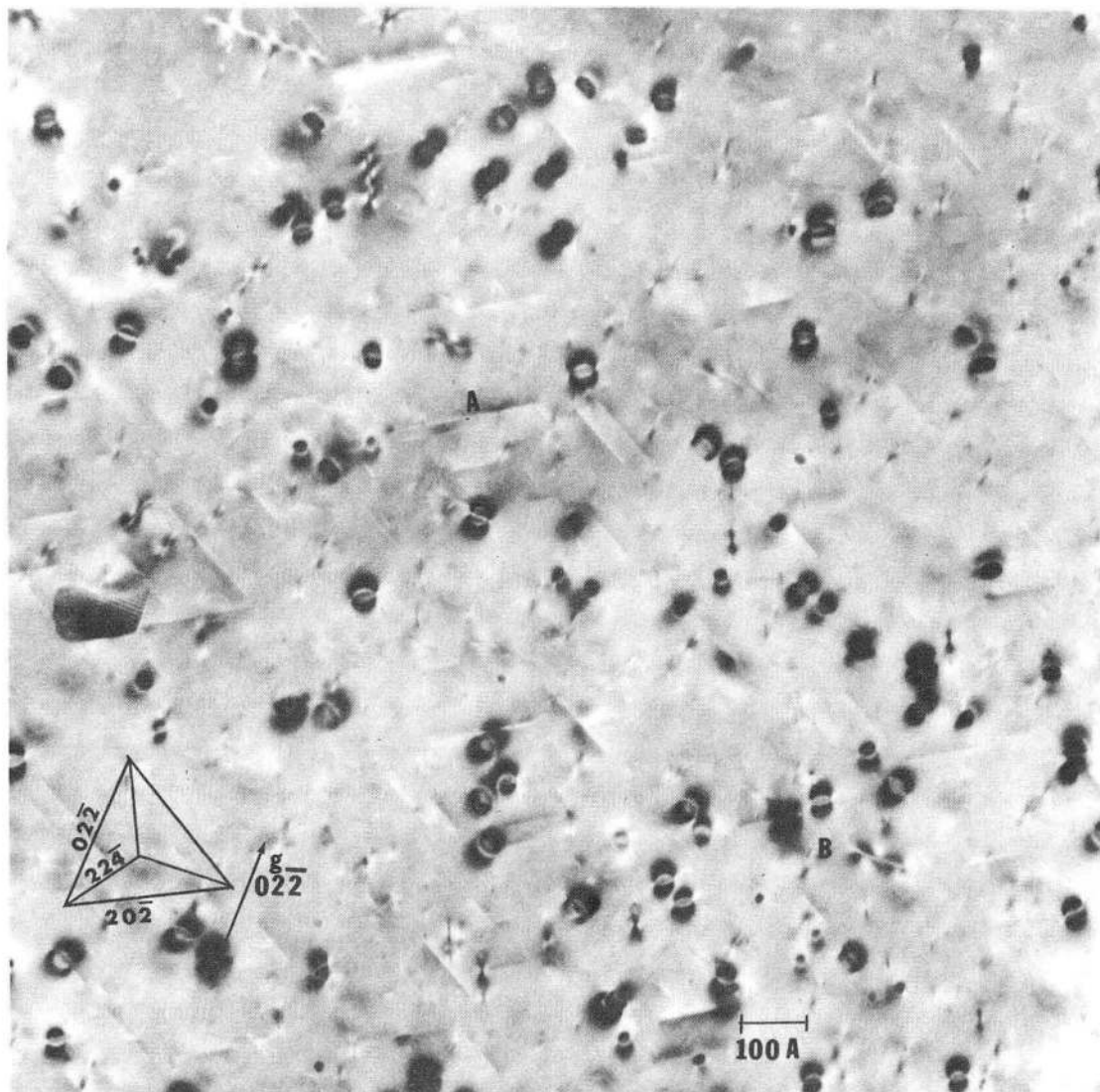
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Fig. 2



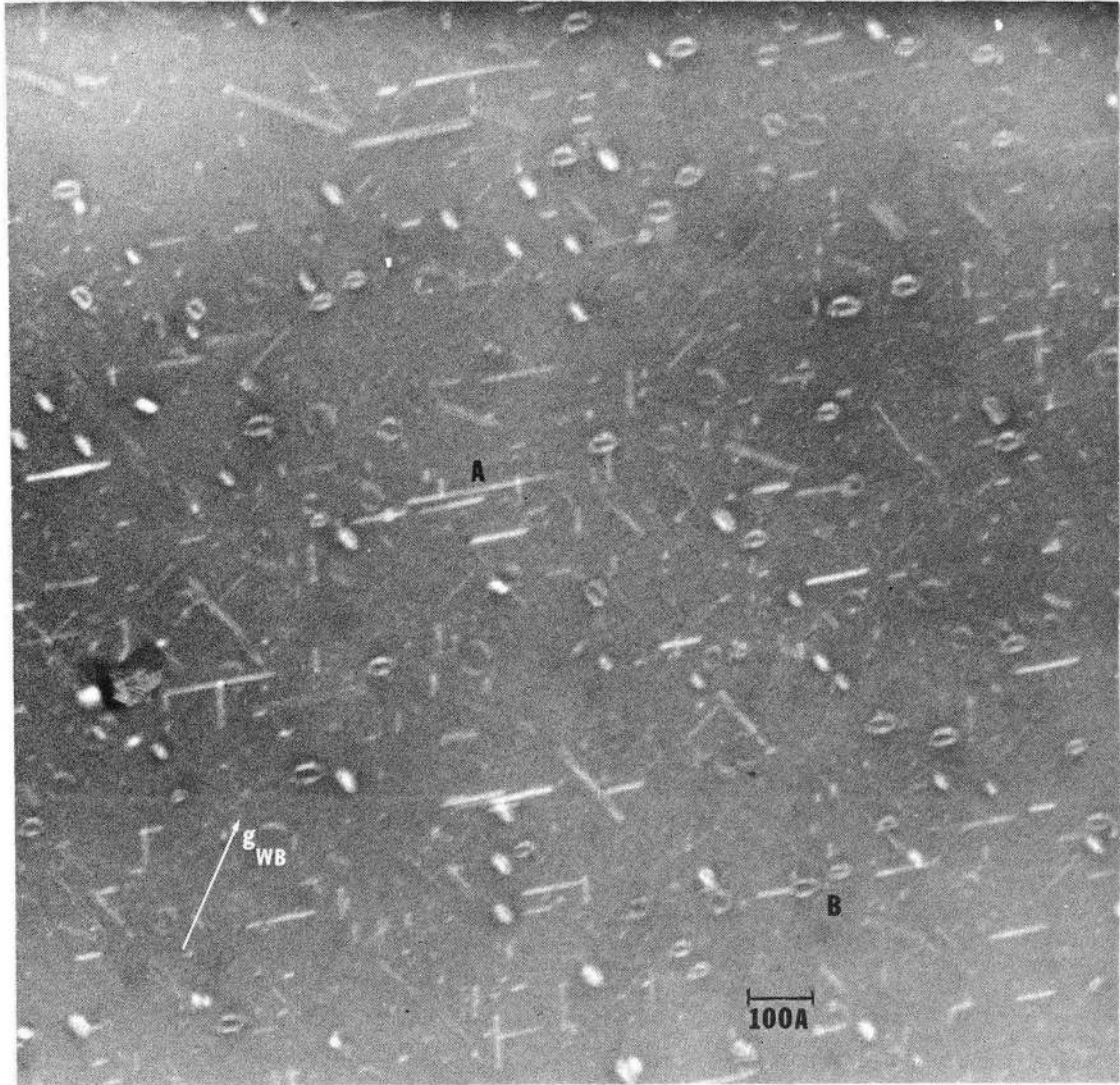
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Fig. 3



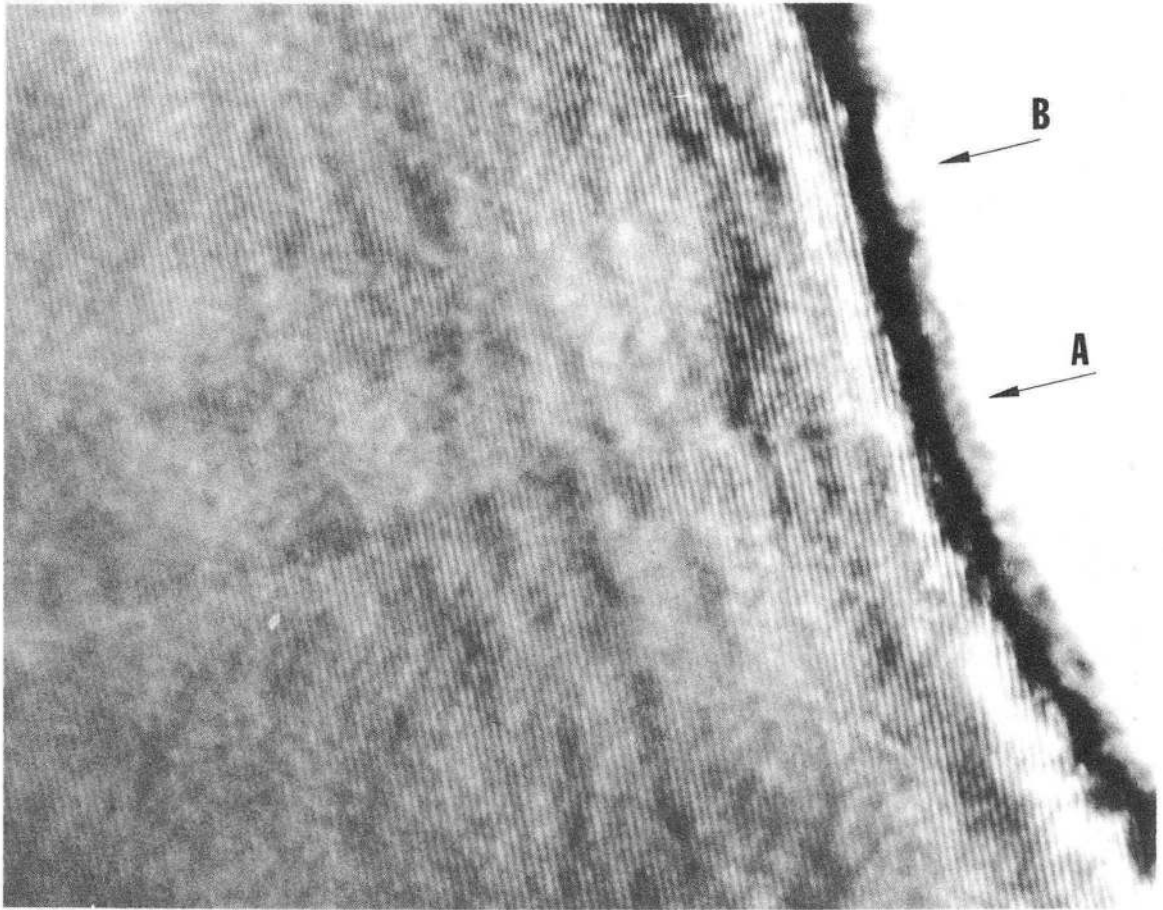
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Fig. 4a



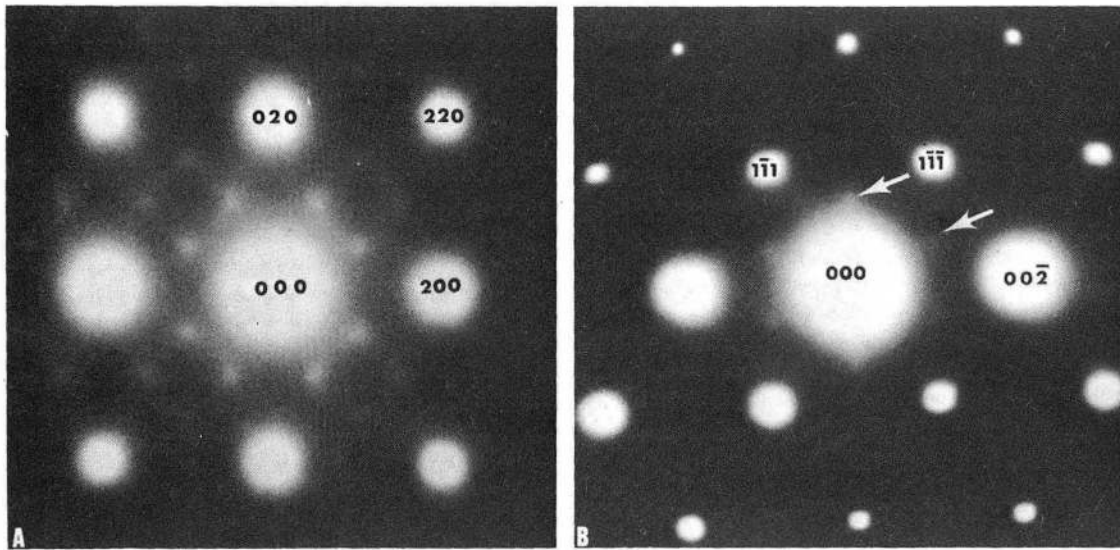
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Fig. 4b



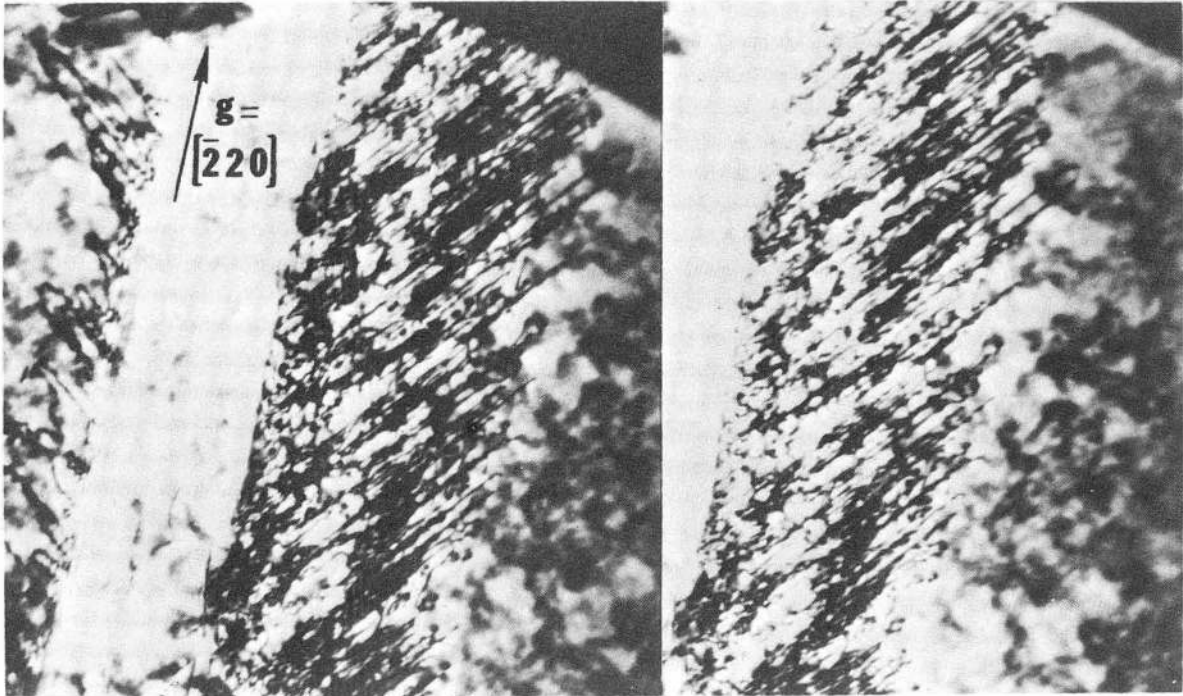
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Fig. 5



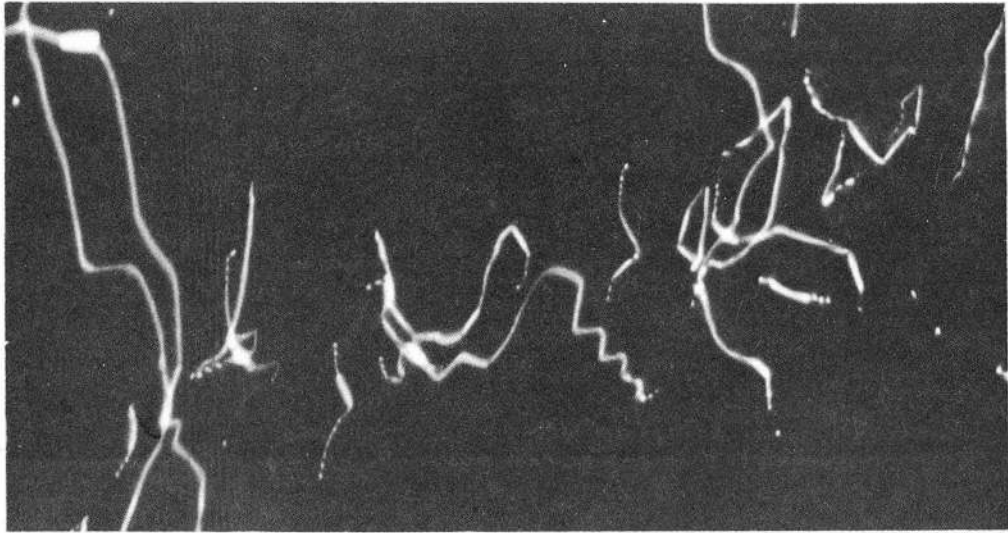
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Fig. 6



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Fig. 7



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Fig. 8

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