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50 Years of Transmission Electron Microscopy of Dislocations: Past, Present, and Future[1]

A Paper by Laureate of the Lomonosov Grand Gold Medal Professor P. B. Hirsch

INTRODUCTION

The first observations of dislocations by transmission electron microscopy (TEM) were published in 1956. Since then the technique has been developed into an indispensable tool for the materials scientist, not only for the characterization of the extended defect structure of materials, but also for the elucidation of the mechanisms controlling their properties. This talk will discuss briefly the historical context of these observations, trace the major developments in the technique since 1956, present a snapshot of the technique as it is today, and suggest some challenges for the future. The lecture will be illustrated by various applications of the technique in materials science. The written background material published here is taken from the preface to the special issue of the *Philosophical Magazine* that marks the 50th anniversary of the first observations of dislocations by TEM, which is due to be published later this year.

HISTORICAL CONTEXT

In the 1940s and 1950s, major advances were made by solid-state physicists in the development of the theory of dislocations in crystals, and its application to crystal growth phenomena and mechanical properties (Fig. 1). Two important books were published, one in 1953 by A.H. Cottrell entitled *Dislocations and Plastic Flow in Crystals* and the other in 1954 by W.T. Read called *Dislocations in Crystals*.

The theoretical developments stimulated the search for techniques to observe dislocations directly under the microscope. Growth spirals were observed on the surface of crystals, confirming F.C. Frank's predictions of growth of crystals containing screw dislocations under low supersaturation conditions. Etch pit techniques were developed that revealed the presence of dislocations intersecting the surface, and these were used to check Frank's formula for the dependence of misorientations at a low-angle boundary on dislocation spacing. Another technique was preferential precipitation at dislocations, as used, for example, by J.M. Hedges and J.W. Mitchell to reveal dislocation networks inside AgBr. However, these techniques were rather limited in scope and not universally applicable, and for metals and alloys, dislocations could only be revealed at the surface. In fact, dislocation theory had far outstripped in complexity what could be observed experimentally; for example, dissociated dislocations had already been suggested by R.D. Heidenreich and W. Shockley in 1948, and reactions resulting in sessile dislocations, e.g., the Lomer–Cottrell dislocations, had been proposed.

The lack of an experimental technique to check proposed theoretical models led to a plethora of dislocation models of mechanical properties. To quote Read, "It became the fashion to invent a dislocation theory of almost every experimental result in plastic deformation. Finally, it became apparent that dislocations could explain not only any actual result but virtually any conceivable result, usually in several different ways." It was not surprising that some metallurgists were rather skeptical of the relevance of dislocations in metallurgy.

While the motivation for development of a universally applicable technique for the observation of dislocations was clear, it was the arrival of a new generation of electron microscopes that made the observation of dislocations by TEM possible. J.W. Menter, working at the Tube Investments Research Laboratories near Cambridge, realized that the resolution of the new Siemens Elmiskop I electron microscope was sufficient to resolve the crystal lattice in crystals with large unit cells and, therefore, potentially the distortion of the lattice planes close to dislocation cores. This led him to produce in December 1955 and to publish in 1956 his beautiful end-on images of edge dislocations in platinum phthalocyanine (Fig. 2) by direct resolution of the lattice planes spaced 1.2 nm apart.

The approach taken by Hirsch and M.J. Whelan was quite different. They also used a Siemens Elmiskop I microscope in Cosslett's group in the Cavendish Laboratory in Cambridge, but revealed the dislocations by "diffraction contrast" (Fig. 3). Whelan had access to the microscope (which was operated in collaboration with

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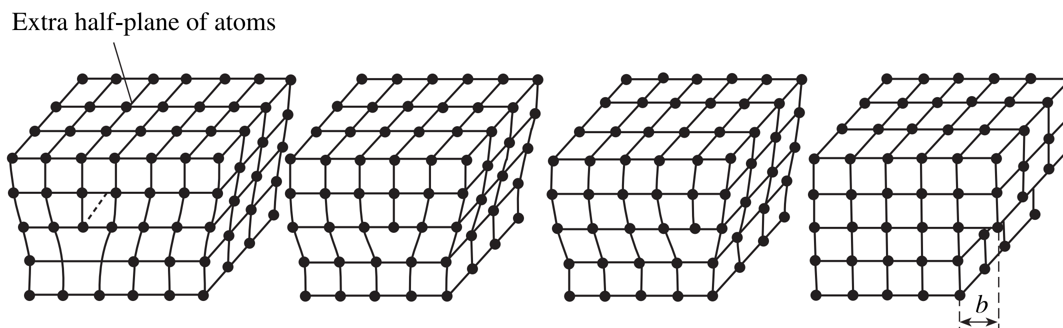


Fig. 1. Slip caused by the movement of an edge dislocation.
The dislocation moves due to crystalline planes slipping from one side of the crystal to the other to form a step (*b*).

R.W. Horne) from about October 1955 and during the next few months studied beaten and subsequently annealed Al foil, and observed, inter alia, regular dots along subgrain boundaries. There was uncertainty as to whether these were dislocations or moiré patterns from overlapping misoriented crystals. The problem was resolved during a session on May 3, 1956, when the specimen was observed with double condenser illumination at 40 000 times magnification (Fig. 4).

On removing the aperture, the dots and lines moved across the specimen leaving pairs of parallel lines behind, which gradually faded. There was no doubt now that the static features previously observed were dislocations, which now moved under the influence of local stress and left slip traces parallel to the projection of (111) planes. The work was published by Hirsch, Horne, and Whelan in the July (1956) issue of *Philosophical Magazine*.

At about the same time, W. Bollmann at the Battelle Memorial Institute in Geneva examined thin foils of stainless steel, prepared by electropolishing, in a Philips EM100 microscope, in the hope of seeing dislocations directly by TEM. Bollmann saw rows of short lines on his images which he interpreted as dislocations. However, no movements of the dislocations were observed; his microscope lacked the advantage of the double condenser illumination system of the Siemens Elmiskop I.

This is the history of events that led to the original publications in the mid-1950s.

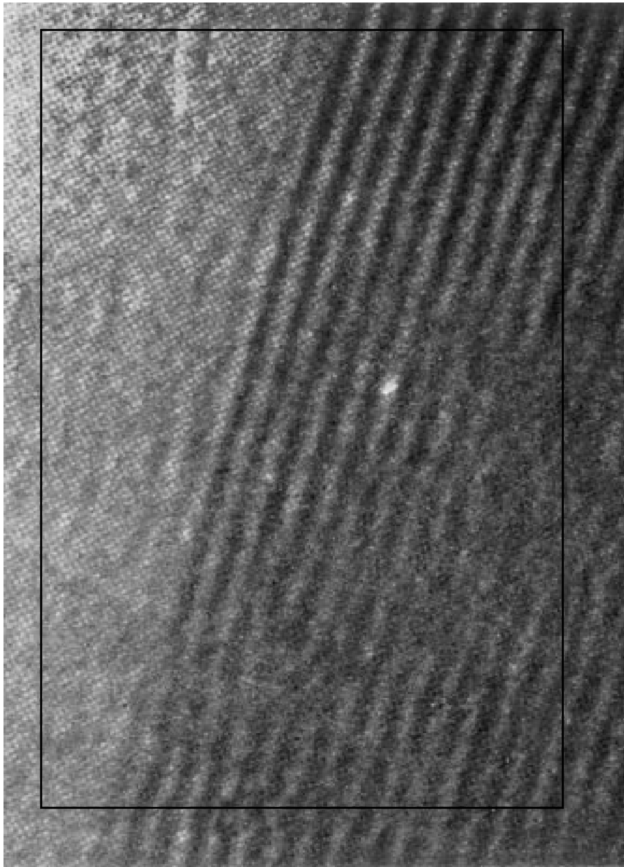
MAJOR DEVELOPMENTS IN TECHNIQUE SINCE 1956

The development of the diffraction contrast technique was much more rapid than that of the high-resolution imaging of the dislocation cores. The reason was that, whereas the application of the latter technique to materials of interest to materials scientists depended on the development of microscopes with much higher resolution, the resolution of the state-of-the-art microscopes of the 1950s and 1960s was sufficient to test the theories of diffraction contrast image and to apply this technique to observe and characterize the nature of the dislocations, their Burgers vectors, distributions, and details of their interactions in many materials. The main requirements were the development of goniometer stages and specimen thinning techniques. Many of the basic dislocation mechanisms, which form part of the theory of dislocations in crystals, were confirmed, and this did much to remove the skepticism of some metallurgists. Furthermore, whereas initially existing theoretical models provided guidance for the experimenters, new defect structures, not previously predicted, were observed. Many of these emphasized the three-dimensional nature of dislocation structures and also the importance of elastic anisotropy.

Generally, the scale of many of the phenomena observed by this technique (~10 nm) was such that they could be interpreted using elasticity theory. The development of the weak-beam technique in 1969 by D.J.H. Cockayne, I.L.F. Ray, and Whelan constituted a step-function improvement in resolution of the technique. The dislocation images could now be reduced to ~1.5 nm in width on a routine basis, and this enabled the dislocation geometry to be determined in unprecedented detail. Important applications include, for example, the measurement of stacking fault energies (Fig. 5). This technique is still today the standard method for the study of dislocations, other than their core structures.

The year 1969 also saw the development by A.K. Head of methods for simulating images of dislocations inclined in the foil, based on the Howie–Whelan equations. These methods have proved to be a powerful tool for distinguishing between alternative models of the defect being imaged. Another technique developed in the 1970s was that using forbidden reflections to image individual atomic height surface steps on single crystal foils, which can be thought of as an extension of the weak-beam technique.

Other major advances included the development of many heating, cooling, straining, and environmental cell stages, stimulated by the arrival of high-voltage



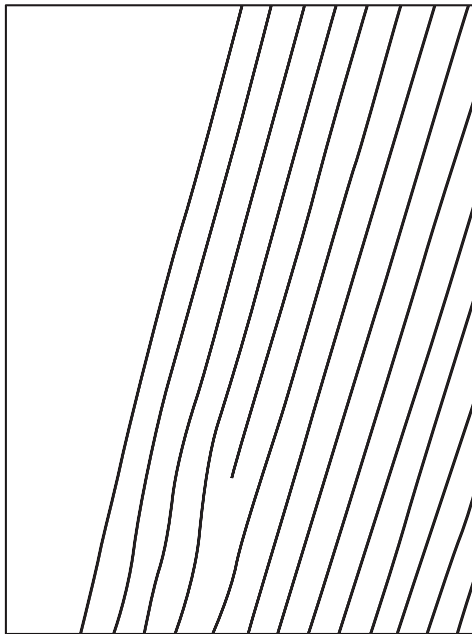


Fig. 2. A photograph and a diagram of an edge dislocation in platinum phthalocyanine.
A 1500000× magnification.

microscopes in the 1970s, which were developed to improve resolution and penetration, and which facilitated specimen manipulation. In situ observations of materials under controlled conditions provided a powerful means of studying dislocation mechanisms; for example, observations of the motion of screws in body-centered cubic (b.c.c.) metals at low temperatures showed clearly that lattice friction (Peierls force) was the velocity controlling mechanism. Various very useful techniques for determining the Burgers vectors of dislocations were developed, based on the effect of dislocations on thickness contours, bend contours, and large angle convergent beam patterns.

But essentially there have been no large changes in the resolution of the diffraction contrast technique for 25 years or more. Of course, there have been improvements in the way the technique has been applied, for example, the more extensive use of image simulation, and microscopes with higher resolution and better vacuum and stability have enabled sharper images to be obtained by using larger deviation parameters providing some increase in resolution. Most recently, the introduction of Omega imaging filters has allowed weak-beam images to be obtained. These images show the greatly increased contrast, which results when most inelastic scattering is removed, allowing thicker samples to be studied. Specimen preparation techniques have also seen considerable improvements, e.g., the focused ion beam (FIB) method.

Quite frequently, the weak-beam technique gives information on atomic scale phenomena, without having atomic resolution. B.C. Carter's observations of climb of jogs in Cu alloys are an example of this, the jogs being inferred by the observable constrictions on the dislocations. Recently, Arakawa (private communication) made direct in situ observations of the rate of growth of interstitial Frank platelets in Cu in a high-voltage electron microscope under conditions of interstitial supersaturation, by following the motion of a bright dot along the edges of the platelet, this bright dot corresponding to the jog in the Frank partial dislocation.

By contrast, the development of high-resolution electron microscopy (HREM) as a universally applicable tool has been rather slower, being critically dependent on instrument development and the accurate measurement of instrument parameters. The generation of instruments in the 1950s had a point-to-point resolution of ~2 nm; by about 1970, this had been reduced to ~0.3 nm. The development of 300–400 kV microscopes helped to improve the resolving power further; by the 1980s, some instruments had resolving powers of ≤0.2 nm, and by the 1990s, ~0.15 nm or better. These advances were made by reducing spherical aberration and improving the mechanical and electronic stabilities. This meant that

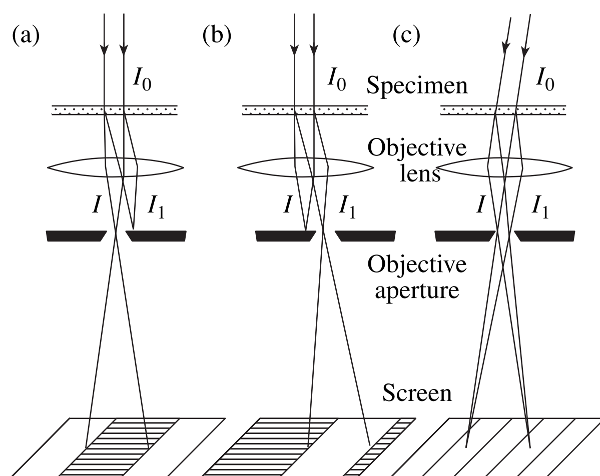


Fig. 3. Diagram of atomic faults in a crystal observed by the diffraction contrast method.

(a) Bright-field image, (b) dark-field image, (c) image produced by interference, (I_0) initial beam, (I) straight beam, and (I_1) reflected beam.

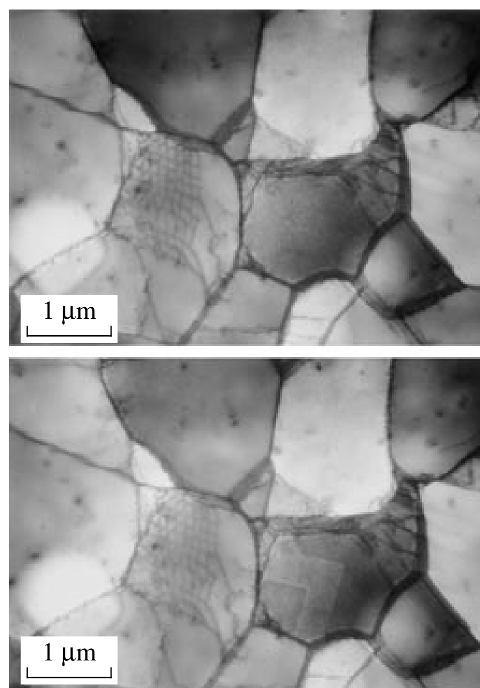


Fig. 4. Dislocations and their motions in the subgrains of aluminum foil.

Imaged by the diffraction contrast method using Bragg diffraction.

defects in an increasing range of materials, including metals, semiconductors, and ceramics, could be investigated at atomic resolution.

The theory of interpretation of HREM images had already been developed extensively before the advent of microscopes with near-atomic resolution. But over the last 10 years or so, there have been very important advances in image interpretation, in particular, the reconstruction of the electron exit wave function, which now enables directly interpretable images to be obtained, whose resolution is governed by the information limit, a considerable improvement on the usual Scherzer criterion. This has provided subangstrom resolution (0.08 nm) from the aberration-corrected Berkeley High Resolution Microscope. The development of spherical aberration correctors has led to similar important improvements.

In the 1970s, microscopes became available in which electrons could be focused into small probes on the specimens and which combined imaging, diffraction, X-ray analysis by energy dispersive spectrometry (EDS), and later electron energy-loss spectrometry (EELS) using energy-dispersive spectrometers. Dedicated scanning transmission electron microscopes (STEM) were developed, and over the last 10–20 years, there have been great improvements in these instruments. Probes 0.1 nm in diameter have recently been produced by advances in lens design and aberration correctors, and the field emission guns, first developed by A.V. Crewe (which are also used in high-resolution TEMs), have provided increased brightness. By collecting the signal in a high-angle annular aperture, it has become possible to image crystal structures and the cores of dislocations by the Z (atomic number) contrast

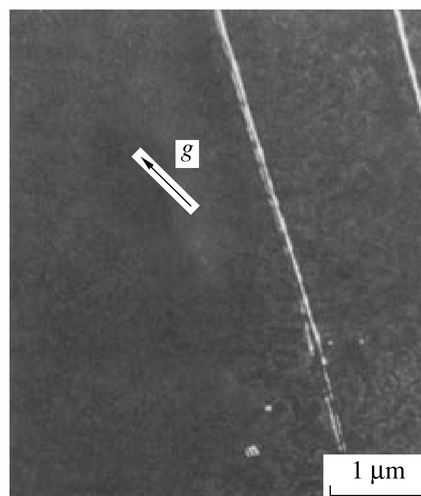


Fig. 5. A dark-field image of a 30° dislocation (bright line) in silicon, obtained by the weak-beam method.

Separating dislocations are visible, and a stacking fault appears between the separated dislocations. The maximal distance between the separated dislocations is 4.8 nm.

technique, which often enables more straightforward interpretation of the images (Fig. 6). This has become established as a powerful tool for the study of dislocation cores, particularly when used in conjunction with EELS at atomic resolution.

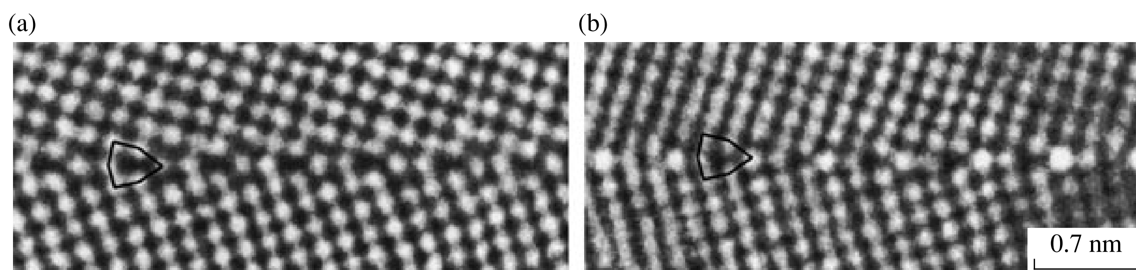


Fig. 6. Z-contrast images of the grain boundary region in pure Cu (a) and Bi-doped Cu (b). Bi settles on the grain boundary, making Cu more brittle.

Another important advance in instrumentation in the last few years is the development of energy filters, which, as mentioned above, eliminate some of the background due to inelastically scattered electrons and enable images to be obtained that have suffered a particular energy loss. Monochromators have also been developed to minimize the energy spread in the incident electron beam, and these can give EELS spectra with an energy resolution of 0.15 eV. In short, the last 10–20 years have seen quite spectacular advances in instrumentation, techniques, and interpretation for structural imaging and the study of dislocation cores with atomic resolution, the structure of which can then be compared with models computed using, e.g., density functional theory.

CHALLENGES FOR THE FUTURE

The challenges and opportunities for the future have already been addressed by a number of the contributors to the *Philosophical Magazine's* special issue, so this section will be limited to a few additional issues and speculations.

The current HREM and Z-contrast techniques are very powerful for resolving the arrangement of columns of atoms in end-on images parallel to the beam. The techniques are therefore well suited to study the cores of edge dislocations or the edge dislocation components of mixed dislocations, because the images can also distinguish between different atomic species. Since the techniques effectively give projections of the structure, it is more difficult to check for changes in atomic composition along the length of the columns, but this should be amenable to comparison between observed and simulated images. EELS has been shown to be a powerful means of giving additional chemical information on an atomic scale, for example, on the presence of impurities. As pointed out by M.F. Chisholm and S.J. Pennycook, it may be possible to reduce the depth of focus sufficiently in the Z-contrast technique, by increasing the convergence angle, to give nanometer resolution along the column, and providing perhaps the possibility of 3D reconstruction of images of dislocation cores.

However, so far it has proved very difficult to determine the core structure of screw dislocations because end-on images are relatively insensitive to displacements along the columns. At present there are two types of structure of interest. First, in some cores, e.g., the 30° partials in the diamond cubic structure, reconstruction is expected to take place along the column, and end-on images are insensitive to such displacements. They do of course affect the higher order Laue zone (HOLZ) reflections, but so does the elastic strain field, which would swamp any core effect. The predicted reconstruction results in a doubling of the lattice period along the (110) core direction, and it has been suggested by J.C.H. Spence et al. that the diffraction pattern corresponding to this core structure when viewed end-on could be detected as a ring where the disc in reciprocal space normal to the dislocation intersects the Ewald sphere. So far this has not proved successful. Alternatively, it might perhaps be possible to detect period doubling by the diffraction effect from dislocations parallel to the foil, for example, from arrays of 60° misfit dislocations (consisting of 30° and 90° partials) at heteroepitaxial interfaces, by the streak normal to the dislocation direction, or even in dark-field images using an aperture that accepts the streak.

In other cases, the core structures may involve displacements normal to the screw, as well as spreading of the shear displacements parallel to the screw along particular planes, as in current models of the screw dislocations in b.c.c. metals. Two HREM studies have been performed so far on end-on screw dislocations in Mo by W. Sigle and B.G. Mendis et al. aimed at revealing the edge-type displacements. The former obtained some evidence for the proposed edge displacements, but the latter did not.

The largest displacements observed are due to the surface relaxations (the Eshelby twist). Mendis et al. have devised an elegant technique for separating out the incompatible deformation by the use of the Nye tensor, which constitutes an important advance in interpretation. There remains some doubt as to whether the inter-

action between the surface relaxation stresses and the edge fractional dislocations is sufficient to change the nature and distribution of the latter. Ideally, one needs a technique which looks at the structure only in the middle of the foil, away from the surfaces. Perhaps the Z-contrast technique with depth resolution might provide the answer. Such a study, if it becomes possible, might also limit the interference from kinks. However, end-on images of screws are unlikely to give any information on the distribution of the shear displacements in the core.

Surface relaxation plays an important role in the image contrast, particularly from end on screws, and sometimes also by affecting their configurations at the surface. This must be taken into account. The Eshelby twist is of course well known to make it possible to image end-on screws by diffraction contrast. In the HREM image simulation of screw dislocations in Mo, displacements corresponding to the Eshelby twist are observed and are found to be those expected at the exit surface. The information on the opposite twist at the entrance surface appears to be lost by the multiple scattering. The surface twist will result in the Bloch waves initially excited undergoing interband and intraband scattering as they reach the perfect region in the middle of the crystal. Those that have undergone intraband scattering will tend to follow the directions of the channels and at the exit surface will be sensitive to, and give information on, the twist at the exit surface. The effect on the final image of the interband scattering, which is expected to be quite strong and lead to redistribution of the Bloch waves, is not clear.

In Z-contrast imaging of end-on screws, the interband scattering due to the Eshelby twist would be expected to give rise to contrast from dechanneling of the Bloch waves. Strain contrast is observed for end-on screw and mixed dislocations. It is clear that the mechanisms leading to strain contrast in Z-contrast imaging are not completely understood and need detailed investigation.

Surface relaxation is also relevant to the elegant investigations by M.J. Hytch and his colleagues, who are interested in the deviations in displacements around dislocations from those expected from anisotropic elasticity. Again, it is the screw component of any dislocation that is likely to have the larger effect. At the present time, the displacements used to account for the surface relaxation of end-on dislocations are those due to J.D. Eshelby and A.N. Stroh, for isotropic elasticity. It is now necessary to solve the problem for anisotropic elasticity.

As pointed out by Spence et al., there is still little information from TEM on dislocation kinks and their structure. What is needed is a technique to image dislocations normal to the beam. Spence et al. demonstrate for the diamond cubic structure the use of imaging in forbidden reflections. Consideration should be given to using the effect of defects on the Z-contrast image obtained with good lateral and depth resolution, to yield information on the nature of the defects.

The fact that with atomic size probes it is possible to resolve individual atomic columns suggests that this might also be possible at the surface of bulk specimens in an SEM with similar size probes, by detecting the back-scattered electrons. The information on the atomic columns is carried in both cases effectively by the electrons which have undergone Rutherford scattering. In the SEM case, the problem is the large background due to multiply inelastically scattered electrons, but in principle, at least this could be alleviated by using suitable filters. It should be noted that dislocations have been imaged using an SEM using the channeling contrast mechanism even without filtering.

So far only structural imaging has been discussed. With regard to the weak-beam technique, in principle it is a powerful method for detecting particular components of the displacements near the core by using larger deviation parameters. For example, for screw dislocations in the b.c.c. lattice, appropriate weak-beam images will include information on the distribution of shear displacements in the core. The problem is the small signal-to-noise ratio due to inelastic scattering. Some of this can be removed by filters, but inelastic scattering with small energy loss and thermal diffuse scattering remains. Experiments at low temperatures should provide some improvement. The increased contrast obtained with Omega imaging filters, referred to above, opens the way toward much more quantitative analysis of the images.

A. Howie has suggested that reconstruction from diffuse scattering around weak beams in the sideband hologram would produce "purely elastic" weak-beam images, greatly improving contrast and quantification, but this idea has yet to be tested. In conventional weak-beam imaging, care must be taken to ensure that the resolution of the image is not diffraction limited by the objective aperture used.

Again, in principle, the weak-beam technique should give information for screw dislocations on displacements normal to $\underline{b}$, by imaging with $g.\underline{b} = 0$. This is relevant, inter alia, to screw dislocations in b.c.c. metals. Depending on its strength, perhaps it may be possible to detect the signal using filtering and low temperatures to reduce the thermal diffuse background. Information on the presence of kinks should also be present in the signal. Comparison with simulated images would be essential. Calculations with the Howie–Basinski equations might well prove useful.

There are some special cases where it might be possible to obtain sharper images by observing the dislocations in more unconventional orientations. It is well

known that the images of edge dislocations lying in slip planes normal to the beam are wider than those of screw dislocations. However, for edge dislocations lying in slip planes parallel to the beam, and which are not normal to the beam, the images are expected to be narrower than those of screws, as has been observed in experiments. This suggests that weak-beam images should be sharper, and this might increase the sensitivity for detecting and measuring the separation between edge dislocations that have undergone climb dissociation, e.g., in intermetallics or some ceramics, perpendicular to the slip plane. But the images are likely to be weaker. No weak-beam experiments have been carried out to date to check this suggestion.

CONCLUSIONS

As the contributions to the special issue show, tremendous advances have been made in the study of dislocations by electron microscopy during the last 50 years, and any prospects envisaged by the early workers have surely been exceeded beyond all expectations.

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