Microdiffraction from Stacking Faults and Twin Boundaries in F.c.c. Crystals

BY JING ZHU* AND J. M. COWLEY

Department of Physics, Arizona State University, Tempe, Arizona 85287, USA

(Received 12 July 1982; accepted 4 October 1982)

Abstract

Electron microdiffraction patterns have been obtained from regions of diameter about 15 Å in thin crystals of stainless steel containing twin boundaries and stacking faults. The diffraction spots show splittings which are characteristic of the type of defect present as a result of interference effects in the coherent convergent electron beam. The observations of spot splitting are in good agreement with calculations based on simple theoretical models. In conjunction with previous work on antiphase domain boundaries [Zhu & Cowley (1982). Acta Cryst. A38, 718–724] these results suggest that the observation of spot splitting is of general relevance for the study of all planar faults in thin crystals.

1. Introduction

By use of a scanning transmission electron microscopy (STEM) instrument equipped with a field emission gun it is routinely possible to get microdiffraction patterns from specimen regions 15 Å or less in diameter. Because the incident electron beam comes from a very small source it may be assumed to be completely coherent. When there is appreciable overlap in the diffraction patterns produced by different beam directions within the incident beam cone, interference effects dependent on the relative phases of the diffracted waves will be produced. For ideally perfect thin crystals no such interference effects occur unless the Bragg reflection diffraction spots are so large that they overlap (Spence & Cowley, 1978). When crystal defects are present, giving rise to continuous distributions of diffuse scattering in the parallel-beam diffraction patterns, interference effects are observable and are characteristic of the defect.

It was shown by Cowley & Spence (1981) that the discontinuities at edges of crystals give rise to a splitting of all diffraction spots. Usually there are intensity maxima at the opposite edges of a diffraction spot disc, separated along a line perpendicular to the crystal boundary. Some planar defects in crystals produce discontinuities with respect to some reflections but not others. For example, antiphase boundaries in ordered alloys involve no discontinuities for the sublattice reflections but produce discontinuities in the superlattice so that some superlattice reflections may show splitting. It was shown by Zhu & Cowley (1982) that by observation of spot splittings from thin films of partially ordered Cu₃Au it is possible to deduce the nature of the antiphase boundary illuminated by an electron beam.

In the present work we extend the observations and analyses to the case of stacking faults and twin boundaries in face-centered cubic metals. The structures of these faults are well known and their diffraction effects have been extensively studied (e.g. Hirsch, Howie, Nicholson, Pashley & Whelan, 1977). The results of the analysis on spot splittings are sufficiently different from those for antiphase boundaries, however, to justify this separate treatment and discussion.

2. Theoretical treatment

Although the diffraction of electrons by thin metal crystal films is marked by strong dynamical diffraction effects, the treatment of the diffraction problem may be made almost as simple as the kinematical approximation by use of the projection approximation which, in our case, is equivalent to the column approximation used for most calculations of electron-microscope contrast.

The two-dimensional wave function $\psi(xy)$ leaving a thin crystal will have the symmetry of the projection of the potential distribution in the incident beam direction (with exceptions which are not important in the present context). For a sufficiently thin crystal in a principal orientation this wave function may be considered as made up of contributions from the individual projected atom positions, so that

$$\psi(xy) = \sum_i \psi_i(xy) \delta(x-x_i,y-y_i),$$

where the characteristic wave function contribution, $\psi_i(xy)$, replaces the projected potential peaks, $\phi_i(xy)$, of the kinematical theory.

In the column approximation the wave function at a point in the exit face of a crystal is assumed to be
influenced only by a column of crystal extending through the sample in the incident beam direction. The width of the column for crystals a few hundred Å thick for 100 keV electrons is normally 3–5 Å. Since we are concerned here with interference effects associated with phase changes over distances comparable with the incident beam diameter of about 15 Å, giving rise to intensity modulations within the extended diffraction spots, the use of the column approximation or equivalent projection approximation will not affect our results to any appreciable extent.

(1) Stacking faults in f.c.c. crystals

For an intrinsic stacking fault viewed in the [110] direction, the projection of the structure is as represented in Fig. 1(a). From the two-dimensional unit cell as indicated, the projected atom positions are designated on $O(0, 0), \alpha(1/3, 1/3)$ and $b(2/3, 2/3)$ on one side of the fault and as $c(1/3, 2/3), d(2/3, 0)$ and $e(1, 1/3)$ on the other side.

The incident beam amplitude distribution is $b(xy)$, given by Fourier transform of the aperture function $B(u, v)$ describing the aperture and aberrations of the objective lens which forms the incident beam, where $u, v$ are reciprocal-space variables. To form a beam of 15 Å diameter with a lens giving an optimum STEM image resolution of 5 Å or better, the objective aperture size is so small that lens aberrations are negligible. Hence $B(u, v)$ is a real symmetric function due to the physical aperture only.

The discontinuity at the fault in the crystal may be represented by use of the step function

$$s(x) = \begin{cases} -1 & \text{for } x < 0 \\ 1 & \text{for } x \geq 0. \end{cases}$$

Then the wave function at the exit face of the crystal is

$$\psi(xy) = b(xy) \left\{ \psi_0(xy) \ast \sum \sum \delta(x - n, y - m) \\
\ast \frac{1}{2}[\delta(x, y) + \delta(x - 1/3, y - 1/3) + \delta(x - 2/3, y - 2/3) \\
+ \delta(x - 1/3, y - 2/3) + \delta(x - 2/3, y) + \delta(x - 1, y - 1/3)] \\
+ \psi_0(xy) \ast \left[ \sum \sum \delta(x - n, y - m) \frac{1}{2} s(x) \right] \\
\ast \left[ -\delta(xy) - \delta(x - 1/3, y - 1/3) - \delta(x - 2/3, y - 2/3) \\
+ \delta(x - 1/3, y - 2/3) + \delta(x - 2/3, y) \\
+ \delta(x - 1, y - 1/3) \right] \right\}. \quad (2)$$

The diffraction amplitude given by Fourier transform of (2) is then

$$\Psi(u, v) = \frac{1}{2} \sum \sum \Psi_0(h, k)P(h, k)\delta(u - h, v - k) \ast B(u, v)$$

$$+ \frac{1}{2} \sum \sum \Psi_0(h, k)Q(h, k)\delta(u - h, v - k) \ast B'(u, v). \quad (3)$$

The Fourier transform of $s(x)$ is $S(u) = (\pi u)^{-1}$. As in our previous paper (Zhu & Cowley, 1982), we have made the approximation that convolution by $u^{-1}$ has
the effect of differentiation so that
\[ [S'(u)]^o(w)B(u, v) \approx icB'(u, v), \]
where \( B'(u, v) = \partial B/\partial u \) and \( c \) is a constant. The influence of \( \Psi_o(w) \) is neglected since this function varies more slowly with \( u \) than does \( S(u) \). In (3),
\[
P(h, k) = 1 + 2 \cos(2\pi/3)(h+k) + \exp\{(2\pi i/3)(h-k)\} \\
+ \exp\{-2\pi i h/3\} + \exp\{2\pi i k/3\}
\]
\[
Q(h, k) = -[1 + 2 \cos(2\pi/3)(h+k)] + \exp\{(2\pi i/3)(h-k)\} \\
+ \exp\{-2\pi i h/3\} + \exp\{2\pi i k/3\}.
\]
The intensity is zero for \( h + k \neq 3n \). For reflections with \( h=3n, k=3m \), \( P=6 \) and \( Q=0 \) and the intensity distribution of the spots is given by \( B^2(u, v) \), i.e. each spot is an image of the objective aperture and is not split.

For \( h \neq 3n, k \neq 3m \) but \( h + k = 3n \), \( P=1/2 - \sqrt{3i}/2 \), \( Q = -(7/2) - \sqrt{3i}/2 \).

The addition of some portion of the symmetrical function \( B(u, v) \) to the antisymmetrical function \( B'(u, v) \) will produce an asymmetric function, which, when squared, will give split spots with an asymmetric intensity distribution, as suggested in Fig. 2. Asymmetry in the intensities of the split spots can also be introduced if the fault is not in the center of the incident beam. This asymmetry may reinforce or cancel that derived above. However, the presence of the \( B(u, v) \) component will ensure that the splitting will usually be superimposed on a constant background and so will show less contrast than in the case of the antiphase boundaries previously considered.

(2) Twin boundary

The [110] projection of a f.c.c. crystal with a twin boundary is represented in Fig. 3(a). With the two-dimensional cell indicated the atom positions are \( O(0, 0), a(1/3, 1/3) \) and \( b(2/3, 2/3) \) on one side of the boundary and \( O(0, 0), b(1/3, 2/3) \) and \( d(2/3, 1/3) \) on the other side. The expression for the diffracted amplitude is then identical with (3) except that
\[
P(h, k) = 1 + \cos \frac{2\pi}{3}(h+k) + \cos \frac{2\pi}{3}(h-k)
\]
\[
Q(h, k) = \cos \frac{2\pi}{3}(h-k) - \cos \frac{2\pi}{3}(h+k)
\]

Fig. 2. An origin of the appearance of asymmetric intensities in the microdiffraction spots.

Fig. 3. (a) The projection of the structure for a twin boundary viewed in the [110] direction. (b) The calculated microdiffraction pattern obtained with the incident beam including a twin boundary. (c) The microdiffraction pattern obtained with the incident beam including the twin boundary of (d). (d) The STEM bright-field image of a twin boundary.
Then $P = 0, Q = 0$ for $h = 3n, k \neq 3m$ or $h \neq 3n, k = 3m$

$P = 3, Q = 0$ for $h = 3n, k = 3m$

$P = 3/2, Q = \pm 3/2$ for $h \neq 3n, k \neq 3m$.

The intensities of the fundamental reflections thus depend on $B^2(u, v)$ only and show no splitting. The intensity distributions of other spots depend on combinations of $B(u, v)$ and $B'(u, v)$ and so show, in general, asymmetric splitting. In Fig. 3(c) the two sets of spots corresponding to the two twinned orientations are indexed in terms of the f.c.c lattice. For the two sets of spots, the sign of $Q$ differs so that they show asymmetry of the spots in opposite directions. Since in this case the values of $P$ and $Q$ are more nearly equal than for the stacking fault, the asymmetry may be greater and the contrast for the splitting less.

3. Experimental procedures and results

The sample, cut from a ClCr18Ni9Ti stainless steel sheet 100 μm thick, was heat treated at 1403–1433 K, water quenched and annealed at 1073 K for 10 h. It was thinned using jet thinning equipment with a solution of 42% H₃PO₄, 34% H₂SO₄ and 24% water at room temperature with a current of 80–100 mA at 5–8 V.

Microdiffraction patterns and images were obtained using the HB5 scanning transmission electron microscope with a phosphor screen followed by an image intensifier and low-light-level TV camera to record the diffraction patterns (Cowley, 1980). With a 10 μm objective aperture the beam diameter at the specimen level was about 15 Å. Fig. 1(d) shows the bright-field image of a stacking fault. The sample was tilted away from this orientation until the incident beam was in the [110] orientation. The (111) planes were then parallel to the beam and the stacking-fault contrast was very weak. Fig. 1(c) shows the microdiffraction pattern obtained with the incident beam including the stacking fault. The spot splitting is seen to be essentially the same as suggested by Fig. 1(b).

Fig. 3(d) shows the bright-field STEM image of a twin boundary. The microdiffraction pattern obtained near the [110] orientation, Fig. 3(c), is seen to show spot splittings as suggested in Fig. 3(b).

Conclusions

The characteristic spot splittings predicted by our simple theoretical treatment for twin and fault planes are clearly observable in microdiffraction patterns obtained from regions of about 15 Å diameter provided that the crystal films are sufficiently thin and there is no great perturbation from other effects. As suggested by the theory the splitting is in the direction perpendicular to the boundary and is usually asymmetric. The contrast of the splitting effect is usually less clear than for the case of the antiphase boundaries previously considered because of the presence of the symmetrical component in the diffraction amplitude expression.

For thicker crystals a number of complications can be introduced. If there are appreciable changes of orientation or thickness of the film associated with the fault, the amplitude or phase of the fundamental reflections may be affected. Then these reflections also show some splitting. Additional phase changes of all spots may be introduced by local variations of thickness, orientation or contamination of the sample or by modification of the incident beam phase distribution due to lens aberrations, defocus or distortion caused by dirty apertures. An extreme case is illustrated by Fig. 4(a) which is a microdiffraction pattern given by a twin boundary in stainless steel. The asymmetry of the spots is in opposite directions for the two sides of the boundary and these directions are almost parallel to the boundary, giving the appearance of grossly distorted rows of spots.

For thick crystals and extended faults the more conventional methods of bright-field and dark-field TEM or STEM are usually adequate for the identification of faults. The use of splitting effects in microdiffraction patterns may be an important supplementary technique for use with very thin crystals and very localized faulted regions. Our results reported here serve to confirm our previous conclusion (Zhu & Cowley, 1982) that the splitting effect has general application as a means for identifying planar defects and is particularly effective in cases which are inaccessible to the more conventional methods.

The authors thank Mr H. Q. Ye for his helpful suggestions and discussions. This work was supported by National Science Foundation Grant DMR 7926460 and made use of the resources of the Facility for High Resolution Electron Microscopy supported.
by the NSF Regional Instrumentation Facilities Program, Grant CHE 7916098.

References


