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ABSTRACT

High temperature electron microscopy experiments on stacking faults in silicon performed under controlled contrast conditions show that the extinction distance for the 220 reflection increases by about 30% over a 600°C rise in temperature. This result is explained by the effect of temperature in reducing atomic scattering.

* On leave 1964/65 in the Miller Institute for Basic Research, University of California, Berkeley, California.

INTRODUCTION

Although high temperature electron microscopy experiments have been carried out to investigate dislocation climb [1-3] and precipitation-dissolution phenomena [4,5], little has been reported concerning specifically the changes in contrast that occur on heating. Howie and Valdrè [6] observed a strong decrease in electron transmission near bend contours in copper and gold foils. These workers suggested that two important effects occur on heating, viz. 1) a decrease in atomic scattering leading to a slight increase in extinction distance t_0 , 2) that due to atomic vibrations, a term $\overline{\Delta s}^2$ must be added to the usual factor s in the dynamical equations which describes the macroscopic crystal orientation with respect to the Bragg condition. $\overline{\Delta s}^2$ is the mean square deviation from the macroscopic s value due to thermal vibrations.

The object of the experiments described in this paper was to try to obtain direct experimental evidence for the change in extinction distance with increasing temperature. Silicon was chosen because of its hardness and resistance to bending and because growth stacking faults can be produced under certain conditions [7], and which are stable during annealing. The observations of fault fringes with change in temperature thus enables one to estimate the change in t_0 .

To determine the true change in extinction distance, it is better to measure thickness fringe spacings [8,9] at the edges of foils rather than stacking fault fringes [10]. However, because the edges of the foil bend appreciably (on heating) and because our hot stage is non-tiltable, it was decided to carry out the experiments on stacking faults.

EXPERIMENTAL PROCEDURE

The Siemens hot stage was used as the specimen holder. This stage was precalibrated using a constant voltage power supply, but due to beam heating of the specimen the actual temperature of the area illuminated can be estimated only to within about $\pm 10^\circ\text{C}$ [12]. The beam was switched off except when taking exposures on the photographic plate. All precautions were taken to avoid specimen contamination. We have found that the use of silicone oil in the diffusion pumps greatly reduces this problem. With the microscope operated at pressures of between 10^{-4} and 10^{-5} mm Hg, no observable contamination occurs above about 300°C . Epitaxial silicon foils in [111] were used. The preparation of these is described elsewhere [7].

CONTRAST CHANGE ON HEATING

The dynamical transmitted intensity, neglecting absorption and temperature, (which describes the bright field image) is given by (see e.g. refs. 8, 11)

$$|\psi|_T^2 = \cos^2 \frac{\pi t}{t_0} \sqrt{1 + w^2} + \frac{w^2}{(1+w^2)} \sin^2 \frac{\pi t}{t_0} \sqrt{1 + w^2} \quad \text{where } w = t_0 s \quad (1)$$

which reduces to $\cos^2 \pi t/t_0$ at the exact Bragg reflection whence $s = 0$.

With increase in temperature, the transmitted and diffracted intensities are modified by the Debye factor e^{-2M} where M is related to the atomic displacements due to thermal scattering [8]. The Debye factor essentially damps the lattice periodicity and decreases atomic scattering. Thus, on heating there will be a loss in diffracted intensity and an increase in thermal diffuse scattering, i.e. the "signal to noise" ratio

in the image decreases. There is thus an advantage to be gained in using dark field images (by gun tilting), of high order reflections in order to reduce chromatic aberration.

The decrease in diffracted intensity due to atomic thermal vibrations can be physically regarded as arising from increased out of phase-ness of successively diffracted wavelets and can be described in equation (1) by adding to s the term $(\overline{\Delta s})^2$ [6]. Also, on heating, the crystal expands so that Bragg's law is modified to $2(d + \Delta d) \sin(\theta - \Delta\theta) = \lambda$, i.e. the Bragg angle decreases. Thus, if originally a crystal is set macroscopically at $s = \text{zero}$, s will become positive on heating.* For example, for the 220 reflection in silicon the d spacing increases by about 0.3% for a 400°C rise in temperature. Consequently, the 220 Kikuchi spacing should decrease by this amount.

The diffraction pattern has been calibrated [13] such that a shift on the plate of a Kikuchi line corresponds to a known angular change in orientation. Thus, as the orientation changes on heating the appropriate correction can be made to retain constancy of orientation and diffracting conditions, by observing the diffraction pattern.

Figure 1 shows a series of sketches illustrating how the contrast changes on heating and how this change can be corrected. In Fig. 1a, the two beam $s = 0$ case is shown. Upon heating (Fig. 1b) the reciprocal lattice point P^* moves inwards along $O^* P^*$ to a new position P_1^* . Thus, s becomes positive and the Kikuchi spacing decreases by Δg . If the foil also bends P_1^* will rotate about the arc $P_1^* R$ as indicated in Fig. 1b,

* The usual conventions for s are adopted [11].

causing further changes in s (back to zero, negative or more positive depending on the bend). If the hot stage is tiltable, then the foil can be tilted as shown in Fig. 1c to restore the orientation back to $s = \text{zero}$. If the hot stage is non-tiltable, the gun can be tilted to correct the change in orientation. This is shown in Fig. 1d. Tilting the gun effectively translates the reciprocal lattice and the spots will be observed to move across the Kikuchi pattern. The gun tilt method is very convenient because the contrast is maintained without tilting the foil, hence the area of interest in the foil is unchanged. However, some foil tilting is inevitable due to heating, but this can be accounted for from the diffraction pattern.

If one works near a bend contour, the profile of the foil will change on heating. If this change is such as to make s negative a serious loss of intensity and contrast [11] will result. For this reason, it is very important to avoid buckling the specimens during preparation and mounting in the hot stage and to avoid working near sharp bend contours.

The dark field by gun tilt is very useful for contrast experiments because then one is always in a two beam situation. However, for the present purposes, bright field images were used because under these conditions fringes from single stacking faults are symmetrical [10]. Two beam cases with $s = 0$ were employed throughout. Since the Siemens hot stage cannot be tilted, the constancy of orientation was maintained by gun tilting. The change in orientation during heating was small ($< 1^\circ$) so that the resultant spherical aberration caused by moving into off-optic axis orientations (the distance B in Fig. 1d) was negligible and did not adversely affect the resolution (see Figs. 4,5).

THE EFFECT OF SMALL CHANGES IN DIFFRACTING CONDITIONS ON THE FRINGE PATTERN

Prior to the high temperature experiments, other factors which might affect the number of fringes in a stacking fault were investigated using a double tilting stage. This was necessary in order to ensure that any small changes in s at constant thickness which might occur during heating, do not produce a change in the number of fringes. According to the dynamical theory of stacking fault contrast including absorption [9,10], at the reflecting position subsidiary maxima or minima tend to disappear except near the center of the fault. The spacing between the first two or last two fringes near the foil surfaces corresponds to the extinction distance. Due to absorption the visibility of the fringes is decreased near the center of the fault.

Figure 2a shows typical single stacking faults in a [111] Si foil. For the right hand fault s is slightly positive. As predicted by the dynamical theory with absorption [10] the central fringes are less visible than the outer fringes. Slight doubling of the central fringes is also observed. This has no effect, however, on the total number of dark fringes present, which is 8. Figure 2b was taken with $s = 0$. The fringes in the center are slightly more damped out. For s negative, Fig. 2c, the resolution of individual fringes has diminished radically and the fault appears asymmetrical in bright field. This apparent asymmetry often occurs during heating when s becomes negative. The missing outer bright fringe is probably not resolved because its intensity is close to background intensity, and thus, the asymmetry is only apparent. Similar contrast effects in bright field with change in s have been observed by Booker [14]. They appear to be characteristic

of s negative and strong absorption.

Figure 2d shows the effect on the fringe pattern of changing the sign of $\bar{g}$ keeping $s = 0$. As expected [15], fringe reversal has occurred and there are now 7 dark fringes and 8 light fringes. The number of dark fringes has decreased by 1, but the number of light fringes has increased by 1, i.e. the total number of fringes is constant (compare to Fig. 2b).

Thus, care must be taken to ensure that the observed fringes at the high temperature are symmetrical and that if a fringe change occurs it is not simply due to fringe reversal. If these precautions are observed, small, local variations in tilting conditions should not affect the number of fringes. As predicted [10] the fringes are only clearly resolved when the orientation is very close to $s = 0$, or s slightly positive. A good approximation to $s = 0$ at the fault is approached when all the fringes are resolvable. In a non-tiltable hot stage, it is only necessary to search the foil for a fault in which the fringes are completely resolved. A selected area diffraction pattern from this fault invariably shows s to be close to zero.

If the fault is set for $s = 0$ at room temperature, any rotation out of this condition at the higher temperature would result in a smaller extinction distance (and thus more fringes) since the apparent fringe spacing is related to s through w in equation (1). Experiments have shown that for this to occur a considerable deviation from s must take place ($> 0.2^\circ$).

Another parameter investigated was the effect of three beam orientations. Figure 3a shows symmetrical faults in a two beam $s = 0$ case.

The doubling and diminished visibility of the central fringes is evident. Figure 3b is the same fault in a three beam case with $s = 0$. The number of fringes has increased from 8 to 13, a ratio of 1.6. Heidenreich [8] has estimated that a change from a two beam to a three beam case for the same $\{hkl\}$ reflections should change the extinction distance by a factor of 1.4. Also of interest is the resolution of fringes in the three beam case (Fig. 3b). No doubled fringes are visible and there appears to be less damping in the foil center. The latter result possibly indicates that there is a decrease in anomalous absorption in multi-beam cases.

If a crystal is set for $s = 0$ and a two beam case at room temperature, a slight variation of s , still staying in the dynamical region or a change to a three beam case will cause an increase in the number of fringes by virtue of a decrease in extinction distance. Increasing temperature is expected to lead only to an increase in extinction distance and thus less fringes should be observed.

Some consideration should be given to the result of thermal expansion which would cause an increase in thickness of the foil and thus the projected width. This increase for Si at 400°C would only amount to 20-30Å which would not lead to any additional fringes as this change is small in comparison to the calculated $\{220\}$ extinction distance for Si (900Å). The effect would, in any case, again cause an increase in the number of fringes at the high temperature. Also any small rotation of the plane of the foil with heating would not cause any change in the number of fringes as the latter depends only on the magnitude of t_0 compared to the thickness.

The discussion of the above possible errors in observing the number of fringes has shown that any extraneous parameters introduced due to the difficulty of maintaining constant diffracting conditions would always lead to an increase in the number of fringes at high temperatures. In practice, two beam cases with $s = 0$ could be obtained at all temperatures after several foils had been examined.

OBSERVED CHANGES IN EXTINCTION DISTANCE WITH TEMPERATURE

Figures 4,5 are examples of the results of the high temperature experiments. All areas selected were close to a strong diffracting position yet away from sharp bend contours. Figure 4a is an area at room temperature. Figure 4b is the same area after attaining equilibrium at about 630°C . As shown by the Kikuchi centers of the diffraction patterns the macroscopic orientation was kept within one degree of each other by gun tilting, maintaining a constant $\bar{g}$ and s close to zero. The effect, if any, of the additional beams observed in Fig. 4b would be one of decreasing the fringe spacing and thus increasing the number of fringes (as in Fig. 3b). However, comparing Fig. 4a to 4b one observes a decrease in the number of fringes at the higher temperature. A comparison of the outer fringe extinction distances in Figs. 4a and 4b showed about a 30% increase in extinction distance from room temperature to 600°C . Figure 5 similarly shows a decrease in the number of fringes at about 430°C indicating an increase in extinction distance, of about 25%.

Measurement of Kikuchi pair spacings as a function of temperature was done on the photographic plate with the aid of a microdensitometer which was used to ascertain the point of maximum or minimum in intensity

within a line. For a 220 reflection at $s = 0$ both lines appear dark (on a plate), but are very diffuse and thus it is difficult to obtain the exact center with any certainty. Also, interference from the transmitted and diffracted beams with the Kikuchi line precludes any precise determination of change in spacing with temperature. By using higher order beams such as the 224 Kikuchi pair a sharper peak is obtained, and from Fig. 6 the d spacing was observed to increase by approximately 1% for about a 400°C rise in temperature. The predicted change based on the linear coefficient of expansion of Silicon is .33%. Possible explanations of this discrepancy are a strong tendency for anisotropy of the given coefficient in thin films and/or possible additional expansion due to elastic stresses.

DISCUSSION

The 220 calculated extinction distance neglecting the temperature factor and using the relativistic corrections is 900Å. The experimental extinction distance calculated from the spacing of the outermost bright field fringes of the stacking faults shown is 640Å at room temperature. Using dark field with s exactly zero a value of 660Å was obtained for the room temperature extinction distance. The discrepancy between the calculated and the observed values at room temperature is being further investigated. The high temperature experiments indicate that the extinction distance increases to about 800Å at 400°C and 850Å at 600°C. As shown earlier the possible orientation variables such as small changes in s or the operation of more than one diffracted beam all cause an increase in the number of fringes and a decrease in extinction distance. Thus, the observed increase of extinction distances is a result of a change in the physical properties of Si with temperature.

The extinction distance for a perfect crystal at the Bragg angle is given by $t_o = \frac{\lambda E}{V_g}$ where V_g is the Fourier coefficient of the operating reflection $\bar{g}$. A change in temperature would have no effect on the numerator, but will affect V_g . The value of V_g is given by [8]

$$V_{hkl} = \frac{4\pi}{\Omega} \sum f_j(s) e^{-2\pi i (\bar{g} \cdot \bar{r}_j)}$$

where Ω indicates integration over the volume of the unit cell and $f_j(s)$ is the electron atomic scattering factor for the j th atom at position r_j in the unit cell. Since the same macroscopic orientations were maintained in all cases, the observed increase in the extinction distance indicates that the effect of temperature is to reduce the Fourier coefficient by a reduction in the atomic scattering factor and, consequently, increase the value of the extinction distance. This effect is stated quantitatively by Heidenreich [8], $f'(s) = f(s) e^{-M}$, where $f'(s)$ is the scattering factor for a vibrating lattice and M is proportional to $\bar{\mu}^2$, the mean square amplitude of vibration of an atom. An increase in M with increasing temperature results in a decrease in the atomic scattering factor and in the Fourier potential of the lattice. The new extinction distance may be expressed by $t'_o = \frac{\lambda E}{V_g} e^M$.

The observed increase in extinction distance of 30% over 600°C is very large. Dynamical theory [8] predicts that near a Brillouin zone boundary perturbation splits a given energy level into two new levels. The energy gap is given by $\Delta E_g = 2|V_g|$ at the exact Bragg condition. Therefore, the extinction distance may be expressed as $t_o = \frac{2\lambda E}{\Delta E_g}$ where ΔE_g is the energy gap at the Brillouin zone boundary corresponding to the operating reflection $\bar{g}$. The effect of temperature is to cause a

change in the magnitude of the energy gap which modifies the dispersion surfaces.

It is interesting to note that for silicon, due to thermal expansion, a decrease in the energy gap between the valence and conduction bands of 4.5×10^{-4} e.V./°C has been observed [16]. This is also of the same order as the observed decrease in ΔE_g (220) of Si from room temperature to 600°C found in our work.

Thus, the primary result of increasing temperature is to cause the Fourier coefficients to diminish leaving the diffuse component (total inelastic scattering) enhanced [8]. This may be expressed as a change in the dispersion surfaces (ΔE_g decreasing) or as a decrease in the atomic scattering factor with temperature.

Finally, it may be remarked that from a practical viewpoint provided that the orientation (i.e. the diffracting conditions), is maintained constant, no great loss in image quality with increasing temperature is observed.

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Figure Captions

Figure 1 Scheme showing changes in contrast on heating and how to correct them.

- (a) Initial orientation, two beam case, $s = 0$.
- (b) After heating showing decrease in $|\bar{g}|$ and θ . The foil may also tilt due to buckling whence P_1^* , will also move along $P_1^* R$.

(c) Correction of the change in (b) by foil tilting.

(d) Correction of the change in (b) by gun tilting.

Notice that a small amount of spherical aberration $\propto B$ now occurs.

Figure 2 Effect of changes in diffracting conditions on the fringe pattern (selected area centered on right fault).

- (a) $g = 2\bar{2}0$, s slightly positive as shown by the diffraction pattern.
- (b) $g = 2\bar{2}0$, $s = 0$.
- (c) $g = 2\bar{2}0$, s negative, notice apparent asymmetry in (c) and the constancy of the number of fringes in (a) and (b).
- (d) $g = \bar{2}20$, $s = 0$, fringe reversal, but total number of fringes is the same as in (a), (b).

Figure 3 Effect of multiple beams on fringe spacing

- (a) $s = 0$, 2 beam case, $g_1 = 0\bar{2}2$.
- (b) $s = 0$, 3 beam case, $g_1 = 0\bar{2}2$, $g_2 = \bar{2}02$.

Notice the decrease in extinction distance resulting in a change from 8 to 13 dark fringes from (a) to (b).

Figure 4 Change in extinction distance with increase in temperature, selected area centered on right fault.

(a) Room temperature.

(b) After 15 mins. at about 630°C.

Notice the decreased number of fringes at the higher temperature.

Figure 5 Change in number of fringes on heating to about 430°C.

(a) Room temperature, selected area centered on right fault.

(b) After 15 mins. at about 430°C.

Figure 6 Showing change in Kikuchi spacing on heating.

(a) Room temperature.

(b) After 15 mins. at about 430°C. The 224 Kikuchi spacing has decreased by about 1% from (a) to (b).

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Fig. 1

(a)

(b)

ZN-4898

Fig. 2 a, b

(c)

(d)

ZN-4899

Fig. 2 c, d

(a)

(b)

ZN-4900

Fig. 3

(a) 0.25μ

(b)

ZN-4901

Fig. 4

(a)

(b)

ZN-4902

Fig. 5

(a)

(b)

ZN-4903

Fig. 6

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