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SOME PRELIMINARY RESULTS ON DEFECTS IN IRRADIATED UO_2 SINGLE CRYSTALS AS REVEALED BY TRANSMISSION ELECTRON MICROSCOPY

by

S. AMELINCKX (C.E.N.)
H. BLANK (EURATOM)

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Work performed under the Euratom/C.E.N. contract No. 063-61-10 RDB
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Some Preliminary Results on Defects in Irradiated UO_2 Single Crystals as Revealed by Transmission Electron Microscopy

H. BLANK* AND S. AMELINCKX
Solid State Physics Department, S.C.K.–C.E.N., Mol, Belgium
(Received 29 January 1963)

Vapor-grown single crystals of nearly stoichiometric UO_2 have been thinned for transmission electron microscopy. The as-grown crystals show a dense pattern of precipitates and may contain loops due to quenched-in vacancies but contain hardly any dislocations. Two models for the production of loops are proposed. Dislocation networks (small angle boundaries) are produced on annealing the crystals. Irregular patterns of dislocations arise when sufficient thick parts of the crystal foils are treated with heat shocks in the microscope suddenly increasing the beam current. After exposure to 10^{15} *nvt* the crystal foils show fission tracks, short segments of straight dislocation lines and black dots, which are partly distributed at random and partly are aligned on fission tracks. Interaction between fission tracks and dislocations has been observed. The conditions for contrast production on fission tracks have been investigated. The experimental findings can be explained by a model of a displacement field around the fission tracks.

I. INTRODUCTION

FOR many years there has been a growing interest in the transmission electron microscopy of UO_2. The problem of getting suitable thin foils transparent for electrons is rather delicate in this material because of poor cleavage properties and because of the brittle nature of UO_2. Therefore, the first investigations were

* Permanent address: Euratom, Brussels, Belgium.

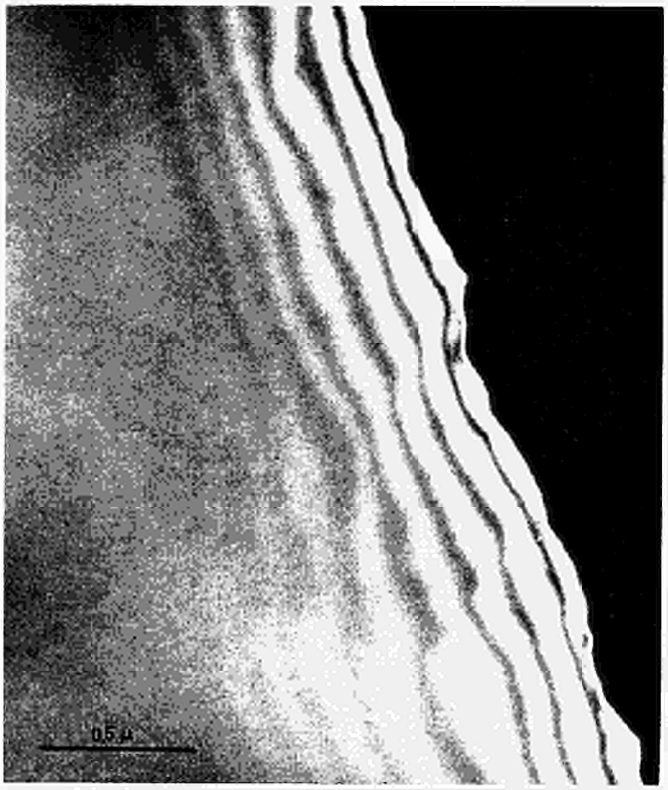


FIG. 1. Dark field picture of a crystal wedge. The diffraction vector operating is $\bar{g}=[220]$, $t_{220}=330$ Å. One can easily count seven extinction contours which correspond to more than 2000 Å thickness at the thick part of the wedge.

carried out on evaporated thin films.[1] A more recent study very elegantly makes use of the fact that thin uranium foils can be transformed into UO_2 by heating in the presence of some oxygen. This transformation can be achieved directly in the microscope with the uranium foil on the supporting grid of the specimen holder.[2]

To study defects introduced by various means into bulk single crystals of known stoichiometry, however, it seems worth while to develop a method for thinning large crystals to dimensions which allow transmission of electrons. The purpose of this paper is to give some first results obtained by such a procedure.

II. EXPERIMENTAL TECHNIQUE

Single crystals of composition $UO_{2.006}$, with uranium of natural isotope content, were grown by condensation from the vapor phase.[3] They were cut and ground into disks of about 3-mm diameter so that they fit into the cap of a slightly transformed specimen holder of an Elmiskop I. The thickness of the disks may vary between 0.1 and 0.2 mm. The final thinning is done chemically with hot orthophosphoric acid[4] in a small container heated from the outside by an electrical furnace. The shape of the container is such as to produce in the acid a convection current[5] which is directed against a sheet of platinum bearing a small hole. This hole is covered by the single-crystal disk which is illuminated from underneath and observed from above with a low-magnification light microscope. As soon as the UO_2 becomes optically transparent it shows a ruby-red transmission color that turns slowly into yellow as the disk becomes thinner. Regions near holes appearing pale yellow are more or less transparent for 100-kV electrons. To get good specimens the surface should be attacked evenly without production of etch pits and with no preferential etching of certain lattice planes. The optimum conditions seem to depend on temperature, impurities in the acid, and on the orientation of the plane to be attacked.

Apart from annealing, the bulk crystals used have not been given any special treatment before the final thinning. However, deformation experiments are in preparation. On the other hand, the as-prepared foils can be treated in the microscope by manipulating the electron beam. Other specimens have been irradiated for short periods in the BR-1 reactor at 20°C. Observations were made in an Elmiskop I at 100 kV.

III. RESULTS AND DISCUSSION

A. General Remarks

With the thinning technique used one sometimes produces crystal wedges which can give some indication on the transparency of UO_2 for electrons.[5] Figure 1 shows the dark field image of such a wedge with the diffraction vector [220] operating in approximately a two-beam case. The calculated extinction depth for [220] is about 330 Å and one can count at least seven successive extinction contours, which means that the crystal thickness was more than 2.000 Å. However, it is more convenient to use only parts of the specimens with a thickness of up to something more than 1.000 Å. For foils having thicknesses in the vicinity of 1.000 Å and more, it is essential to make use of anomalous transmission and, hence, to bring a suitable diffraction vector into operation by proper tilting. The lateral extension of the suitable areas seems to depend sensitively on the orientation of the foil plane during the thinning operation.

B. Precipitates and Loops

The as-grown crystals contain precipitates of two distinct sizes and possibly small dislocation loops. It has not been possible yet, to discriminate between small loops (diameters 120 to 350 Å) and precipitate particles

[1] T. K. Bierlein and B. Mastel, J. Appl. Phys. 31, 2314 (1960).
[2] A. D. Whapham and M. J. Makin, Atomic Energy Research Establishment Report, AERE-R 4066 (1962).
[3] W. Van Lierde, R. Strumane, E. Smets, and S. Amelinckx, J. Nucl. Mater. 5, 250 (1962).
[4] A. Briggs, Atomic Energy Research Establishment Report, AERE-M 859.
[5] H. B. Kirkpatrick and S. Amelinckx, Rev. Sci. Instr. 33, 488 (1962).

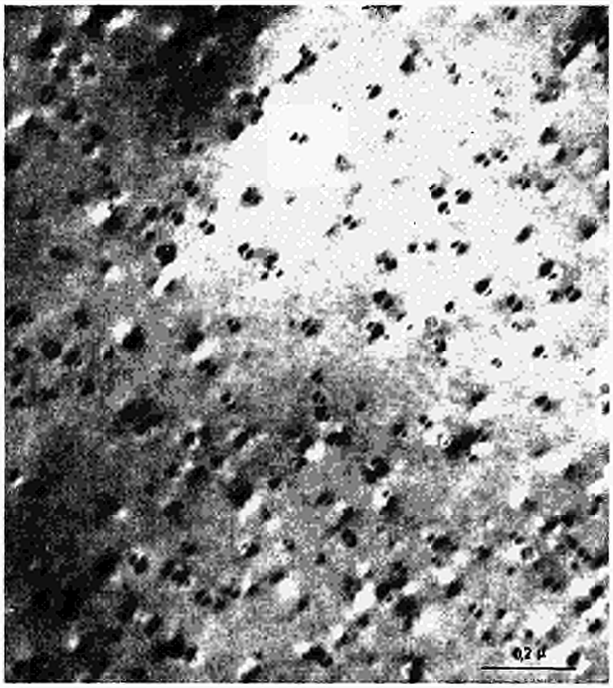


FIG. 2. Pattern of precipitates in a nonannealed crystal.

of about the same size. Big precipitates with extensions of the order of 2 to 5000 Å have the shape of crosses of more or less regular geometry their arms extending in ⟨100⟩ directions. The small type of particles, if it has a size of at least 500 Å, can be recognized as small squares (Fig. 2) with sides somewhat curved towards the interior of the square and their diagonals again extending in ⟨100⟩ directions. Up till now no platelets as viewed from the side have been found so these particles may consist of small cubes. It is highly probable that both big and small precipitates are of the same kind as bigger precipitates always show a zone around them denuded from smaller particles, see Fig. 3.

Annealing the crystals for 1 h at 1500°C under hydrogen removes all precipitates and loops, so we conclude that the precipitates consist of a higher oxyde of uranium. A more detailed account on these precipitates in UO_2 crystals, will be published elsewhere.

After sublimation the bulk crystals are cooled fairly rapidly from about 2300° to 1000°C, as a consequence the occurrence of quenched-in vacancies coalesced into loops seems possible.

For reasons of lattice geometry, the following models for loops may be proposed. The most favorable planes for the precipitation of vacancies in a compound crystal are those where the ions are present in the correct stoichiometric ratio, in our case O:U = 2:1. Part of such a plane can be removed without disturbing the stoichiometry. The simplest family of planes that satisfies this condition is {110}.

Removing part of a (110) plane and letting the resulting penny-shaped cavity collapse would give rise to a loop with a Burgers vector $(a/4)[110]$. This loop would contain a severe fault, the fault energy being mainly due to the fact that like ions are in direct contact [Fig. 4(b)]. This fault can be removed by a shear motion in the (110) plane. Considering the detailed geometry the shortest vector that would bring the ions again in almost correct environment is $(a/4)[1\bar{1}0]$ or $(a/4)[\bar{1}10]$. The resulting loop would then have a Burgers vector $(a/2)[100]$ or $(a/2)[010]$ as a consequence of the reactions:

$$\frac{a}{4}[110]+\frac{a}{4}[1\bar{1}0]\rightarrow\frac{a}{2}[100]$$

or

$$\frac{a}{4}[110]+\frac{a}{4}[110]\rightarrow\frac{a}{2}[010].$$

Furthermore, the loop would still contain a stacking fault as can be seen in a sideview given in Fig. 4(e).

A perfect loop can be generated by a shear motion in the (110) plane with a Burgers vector $(a/4)[1\bar{1}2]$ or $(a/4)[\bar{1}12]$ according to the reaction

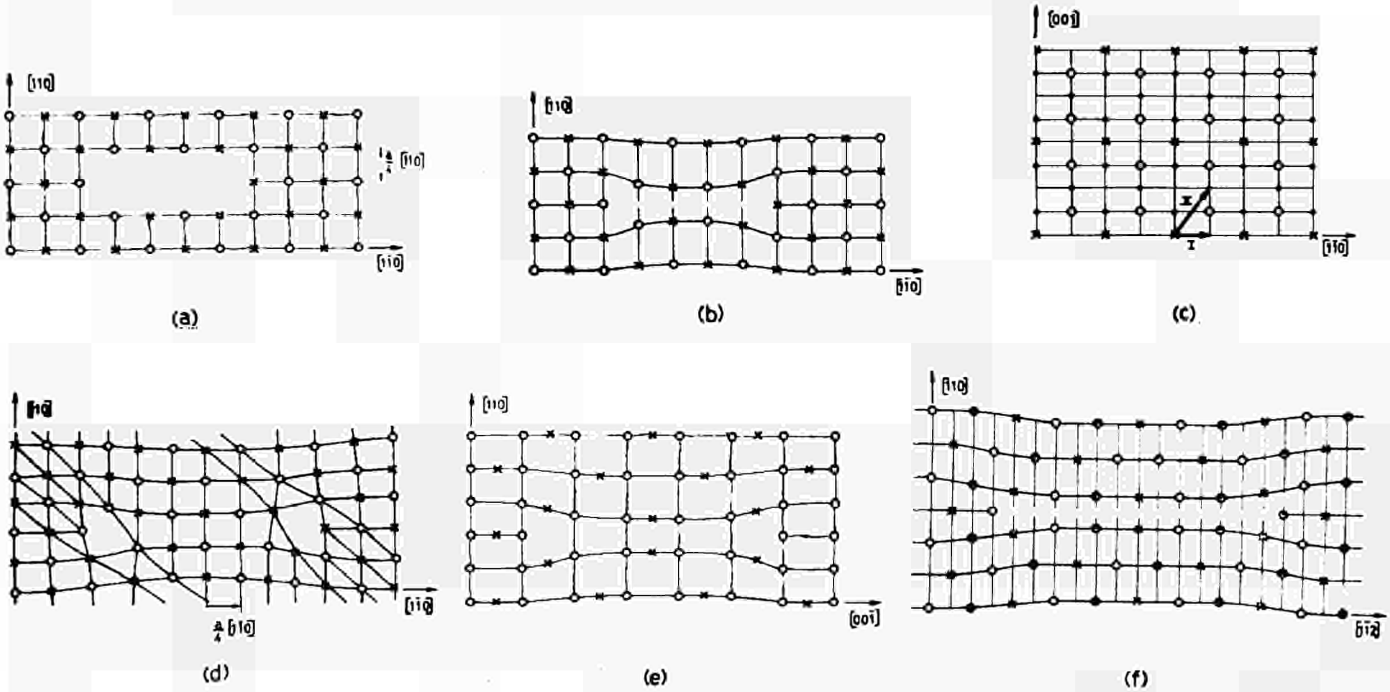
$$\frac{a}{4}[110]+\frac{a}{4}[1\bar{1}2]\rightarrow\frac{a}{2}[101]$$

or

$$\frac{a}{4}[110]+\frac{a}{4}[\bar{1}12]\rightarrow\frac{a}{2}[011]$$

as can be seen from Figs. 4(c) and 4(g).

FIG. 3. Absence of precipitates near a small angle boundary decorated by bigger precipitations.



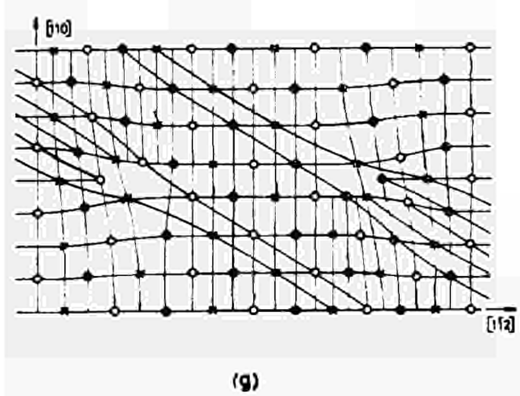


Fig. 4. Geometry of the formation of vacancy loops in {110} planes of UO_2. x uranium, o oxygen. Part of the atoms shown in the plane of the drawings actually lie above or below this plane. (a) View of (110) planes with part of one (110) plane removed as seen along [001] direction. (b) Collapse of the cavity without shift in the (110) plane causes an unstable high energy fault; view along [001] direction. (c) Single (110) plane viewed in direction [110]. Two possible shift vectors $(a/4)[1\bar{1}0]$ (I) and $(a/4)[1\bar{1}2]$ (II) are indicated for diminishing or removing the fault of (b). Positions of ions in the next (110) plane are indicated by black dots. (d) The crystal part beneath the fault of (b) is shifted by $(a/4)[1\bar{1}0]$ thus removing part of the fault. The remaining stacking fault can be recognized in (e). (e) The fault of (d) viewed along $[1\bar{1}0]$ direction. The order of the oxygen sublattice is restored, but in the uranium sublattice an irregularity remains across the plane of the fault. (f) The high-energy fault of (b) viewed along the $[1\bar{1}\bar{1}]$ direction. (g) Shifting the lattice under the fault plane by the vector $(a/4)[1\bar{1}2]$ completely removes the fault and leaves a dislocation loop with Burgers vector $(a/2)[101]$ in the (110) plane. This loop can change its plane into (101) by prismatic glide thus further diminishing the energy of the loop.

The kind of loop that will form will depend on the magnitude of the stacking fault energy. In the latter case prismatic glide is possible. There will be a tendency for the loops to change orientation since their Burgers vector forms an angle of 60° with the normal to the loop plane (110); the new plane will again be of type {110}, namely (101) or (011), respectively, and the loop will be prismatic and square-shaped.

The second type of loop was encountered only in a very few instances in a foil prepared from an annealed crystal which was irradiated afterwards with nearly 10^{15} *nvt* (thermal). The diameter of these loops is greater than in the first type, namely, 2.000 to 2.500 Å. They are of circular shape and sometimes show a strange contrast, Fig. 5. It is supposed that they result from fission damage by a mechanism not yet understood.

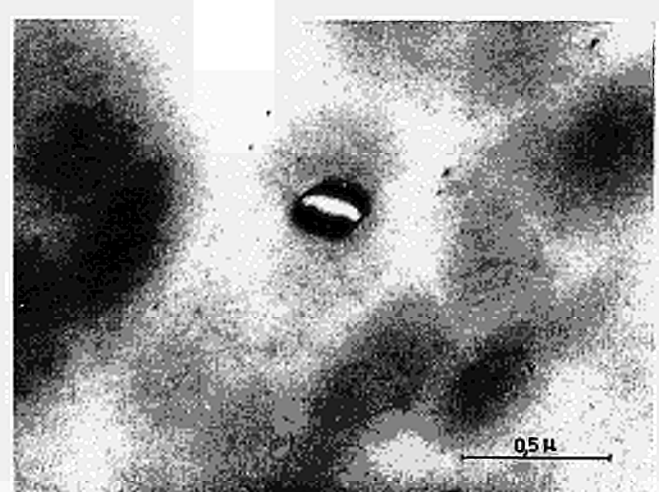


Fig. 5. Large single loop in crystal foil, irradiated up to 10^{15} *nvt* (thermal).

C. Dislocations

No systematic study of dislocations is available as yet, only preliminary observations made on as-grown

FIG. 6. Dislocations lying steeply inclined to the plane of the foil. Area near a crack which has been produced in a thick part of the specimen by manipulating the electron beam.

FIG. 7. Dislocations lying more or less parallel to the plane of the foil. Area near another crack. In thicker parts of this specimen more regular dislocations patterns have been observed.

crystals are discussed. Such crystals contain, in general, a very small density of dislocations.

Attempts were therefore made to deform crystals in the microscope, by thermal shock. In regions which are thick enough, a sudden increase in the electron-beam

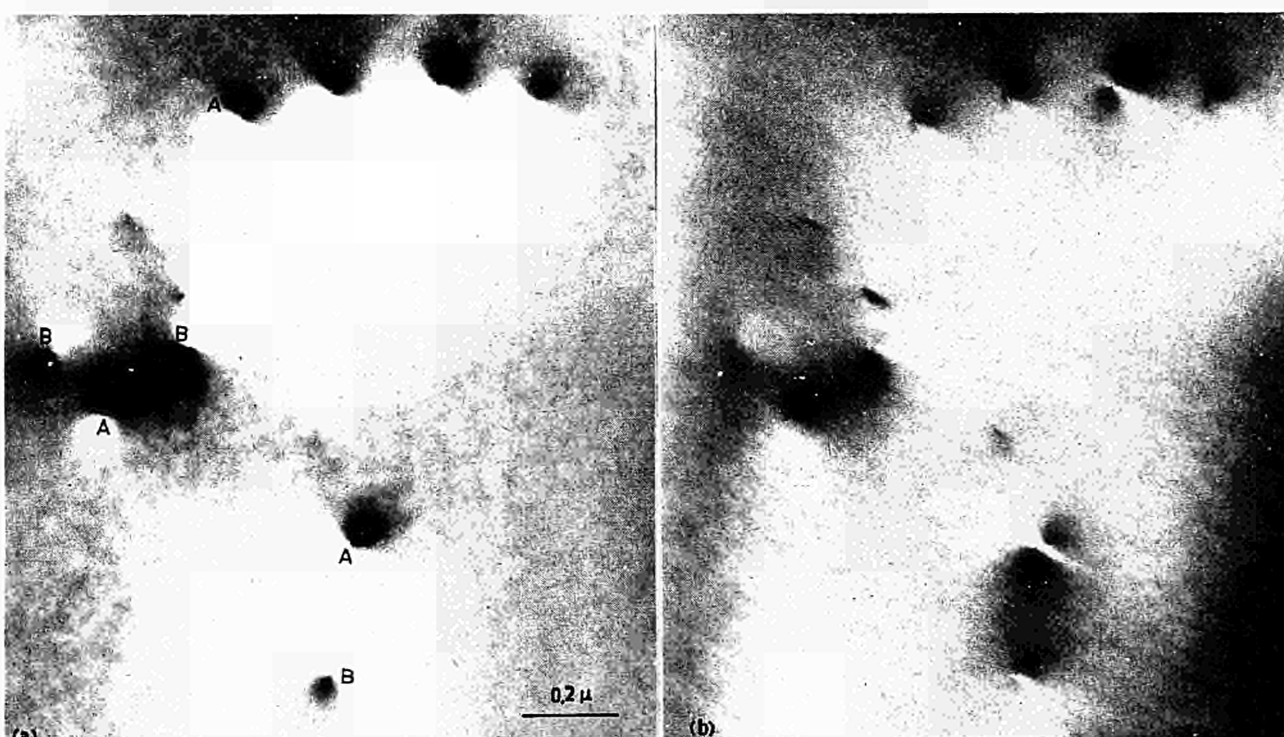


FIG. 8. Dislocations seen end-on in an area which had been thinned with the electron beam by evaporating part of the UO_2. (a) Bright field image. Dislocations denoted by A have opposite Burgers vectors with respect to those denoted by B. (b) Dark field image with $\mathbf{g}=[331]$ (two-beam case). Some of the dislocations show noncomplementary contrast in the dark field due to anomalous absorption.

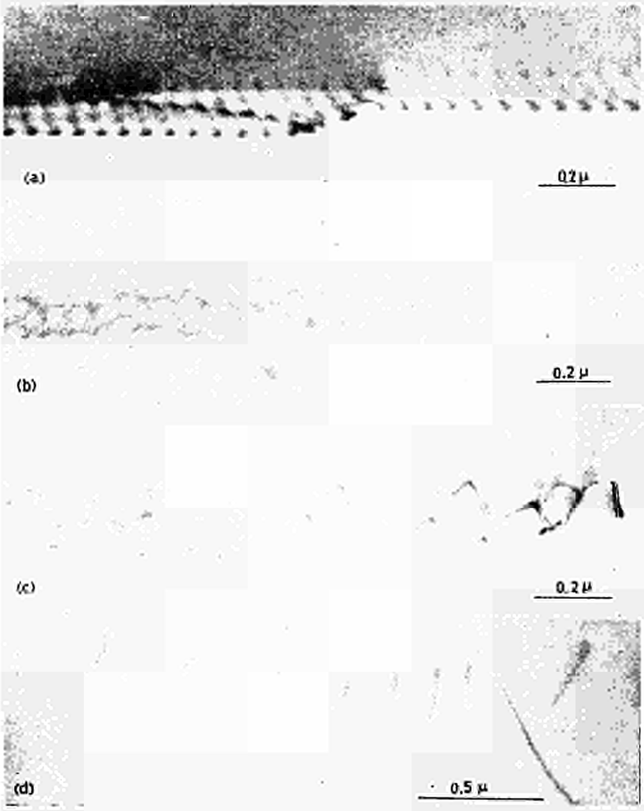


FIG. 9. Several small-angle boundaries found in annealed crystals. (a) Tilt boundary containing some dislocations of different Burgers vectors. (b) Twist boundary due to interaction of two sets of dislocations with different Burgers in the same lattice plane. (c) Boundary similar to the one shown in (b) but less complete developed. One set of dislocations shows double contrast. (d) The boundary shown in (c) ending in loosely assembled single dislocations.

current is sometimes sufficient to create cracks. In the vicinity of these cracks irregular dislocation patterns are often produced. (See Figs. 6 and 7.) In thin regions this treatment does not produce dislocations. In a few cases, dislocations perpendicular to the foil plane resulted from a sudden cooling of regions which had previously been thinned by heating with the electron beam and subliming away some material. The pattern of precipitates (or loops) previously present in such an area is completely wiped out by this procedure [Fig. 8(a), (b)]. From this pair of photographs it is clear that the bright (a) and the dark field images (b) are not complementary, as a result of anomalous absorption. From the asymmetry the sign of the dislocations can be determined.

Dislocations in annealing configurations have been found in crystals that were heated during 1 h at 1500°C under hydrogen. This treatment also resulted in the removal of the square-shaped precipitates (or loops) described in the previous paragraph.

The stress relaxation on anneal results in the formation of networks, e.g., as Figs. 9(a), (b), and (c), or in single dislocations more loosely lined up as in Fig. 9(d).

D. Glide Elements, Burgers Vectors

The deformation modes of UO_2 single crystals prepared by fusion have been studied already by Rapperport and Huntress[6] using optical surface inspection of glide traces and x-ray diffraction. The results were in agreement with the slip systems to be expected in crystals of the CaF_2 structure. The slip direction is $\langle 110\rangle$ with the most important glide plane $\{100\}$[7] and the second important glide plane $\{110\}$. In the present investigation it could be deduced directly that the Burgers vector is parallel to $\langle 110\rangle$. The following contrast conditions were found for two different dislocations:

	Diffraction vector g In contrast	Diffraction vector g Out of contrast	Approx. image plane
Dislocation 1	$[\bar{2}24]$	$[\bar{6}22]$	$(\bar{1}\bar{5}2)$
Dislocation 2	$[\bar{1}33]$	$[2\bar{4}2]$	(620) and (210), respectively.

Using the criterion $\bar{g}\cdot\bar{b}=0$ for the absence of contrast, it can be concluded that the observed contrast effects are consistent with a $(a/2)\langle 110\rangle$ vector.

E. Fission Damage

The first experiments with annealed crystals were oriented towards a determination of the minimum neutron dose that would result in observable track formation. After an integrated flux of about 3.5×10^{12} thermal neutrons no unambiguous evidence for the presence of tracks could be found, presumably because their density on the scale of the electron microscope is too small. A specimen which had a dose of 10^{15} *nvt* showed clearly many tracks and black dots. This specimen was irradiated in two steps, partly before the final thinning and partly as a thinned foil. (Fig. 10). The area shown has a thickness of about 1000 Å as deduced from the spacing of Kossel–Möllensted fringes.[8,9]

From the dose and the thickness of the foil one can estimate that about 50 fission events should have taken place in an area $2.5\times 2.5\ \mu^2$ which is shown as a field 6×6 cm^2 on the photographic negative at magnification 24 000. In the best negatives nearly 20 tracks could be counted in such a field. One of the reasons for this discrepancy between the actual and the registered density of fission events is due to the contrast properties of fission tracks that is discussed below.

The track lengths are of course mostly limited by intersection with the surface. The longest tracks observed had a length of about 1.5 μ. These long tracks have a nonuniform contrast, the contrast disappears

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[6] E. J. Rapperport and A. M. Huntress, NMI-1242 (August 1960).
[7] S. Amelinckx, Nuovo Cimento Suppl. **7**, No. 2, 569 (1958).
[8] W. Kossel and G. Möllenstedt, Naturwissenschaften **26**, 660 (1938); Ann. Phys. **36**, 113 (1939).
[9] R. Siems, P. Delavignette, and S. Amelinckx, Phys. Stat. Solid **2**, 421 (1962).

once or twice along this length, as, for instance, in Figs. 10 and 11.

A number of shorter tracks seems to start in a big dot as shown clearly in Fig. 12. Many tracks are registered only as faint straight lines whereas others are decorated by more or less regularly spaced dots. The spacing between these dots is found to vary in most cases between about 300 and 800 Å, see for instance Fig. 10. This is the range of distances which should be expected between high-energy collisions along the fission fragment path when the moving atom has at least lost half of its initial energy.[10] It is suggested that these dots represent disordered regions of the displacement spike type. Clearly, further investigation is necessary to decide on the origin of the single dots at the beginning of the visible tracks and on the many isolated dots also showing up in most pictures.

FIG. 10. Aspect of crystal foil 1000 Å thick after irradiation with nearly $10^{15}nvt$ (thermal). Nondecorated and decorated fission tracks, black dots, and short segments of dislocations are visible.

To date only very few cases have been observed where the spacing of several dots diminishes along a visible track as should be expected from the decreasing mean free path of the fission fragment when it looses its energy near the end of its path; see Fig. 13.

In a few cases bent tracks were observed. These surely must be attributed to large-angle scattering events which, according to theory,[10] should occur

[10] J. A. Brinkman, "Fission Damage in Metals," Summer Course on Radiation Damage in Solids, Ispra (September 1960).

FIG. 11. Varying contrast of fission tracks due to different orientation of tracks with respect to diffraction vector **g**. *g* is parallel to the line of the scale indicator. The track denoted BB lies nearly parallel to **g** and, therefore, is almost out of contrast. Tracks roughly at right angles to **g** show strong contrast.

FIG. 12. A bent fission track, short tracks starting in a big black dot, and short straight segments of dislocation in irradiated UO_2.

FIG. 13. Interaction between fission tracks and dislocations. At AA a fission fragment probably has cut through two dislocations of the small angle boundary. At B a track decorated by dots showing decreasing distances can be seen.

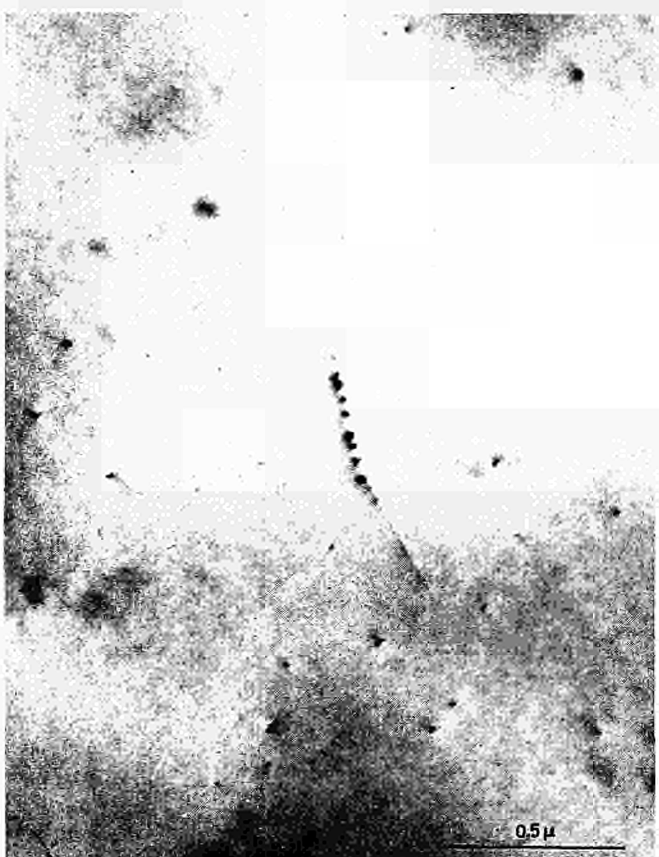

FIG. 14. A bent fission track decorated by dots.

seldom. Examples are shown in Fig. 12 and Fig. 14.

Even these lightly irradiated foils seem to contain more dislocations than unirradiated ones, suggesting that dislocations might be introduced by the irradiation; see Figs. 10, 11, and 12. There is a visible interaction between a fission track and dislocations in Fig. 15. In Fig. 13 a fission fragment atom probably has cut through two dislocations of a small angle boundary.

F. Contrast Formation at Fission Tracks

The contrast of the tracks depends sensitively on the orientation of the foil with respect to the electron beam, tilting of the foils may cause the disappearance

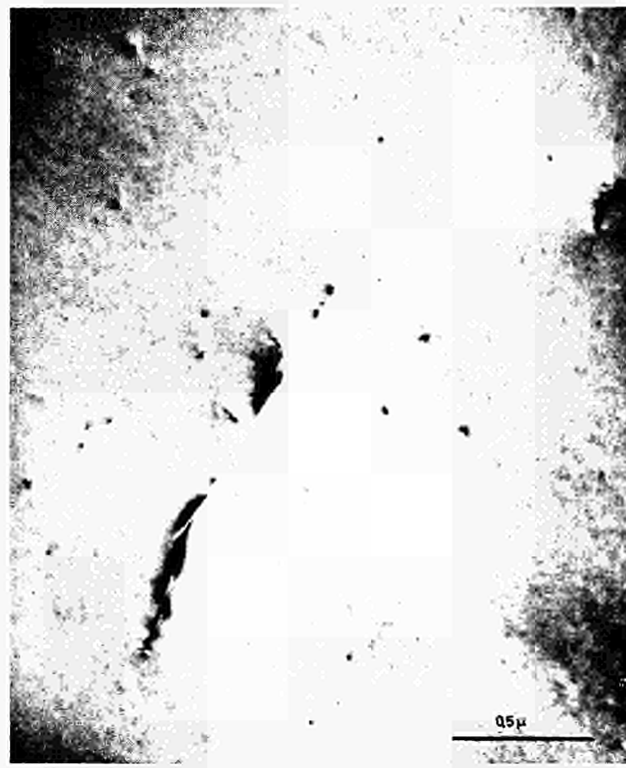

FIG. 15. Interaction between fission tracks and dislocations. The fission fragment takes its course partly along the cores of the two dislocation lines.

of tracks, as already mentioned by Whapham and Makin.[2] To investigate this phenomenon more closely, several areas showing fission tracks have been photographed with different diffraction vectors operating effectively in two beam cases. Larger fission tracks, i.e., those that are not too far from parallel to the plane of the foil, are found to be in good contrast if they are roughly at about right angles to the diffraction vector **g** operating. If the angle between **g** and the track becomes smaller (especially smaller than about 20°) the contrast becomes weaker and practically vanished if **g** and the track are parallel. In this case the track may only be revealed if it is decorated by dots. An example is given in Fig. 11 where the diffraction

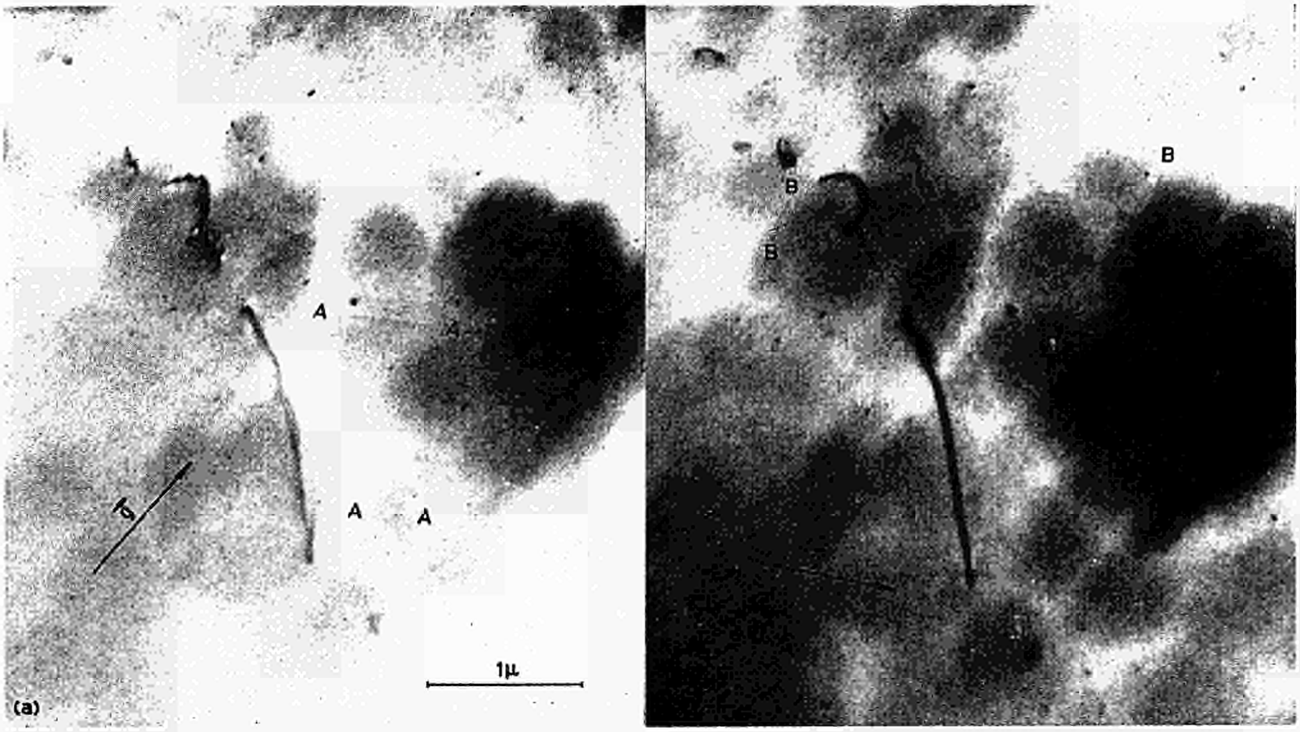

FIG. 16. Visibility of fission tracks due to different vectors. The tracks AA in (a) are not visible in (b) and tracks BB of (b) are not visible in (a). The direction of the diffraction vector operating is indicated in each case.

vector lies parallel to the horizontal edge of the picture. The track denoted by AA is in weak contrast, that denoted by BB is hardly visible, whereas the others making larger angles with **g** are seen very clearly. Figures 16(a) and (b) show that different tracks can be brought into contrast by tilting, i.e., by using different vectors **g**.

Since the contrast depends critically on the foil orientation, it is clear that the main origin of the contrast is by diffraction. A model which appears to be a good basis for the interpretation of the observed features is as follows.

We assume that the core of the track consists of a highly disordered region, and hence that volume expansion (or contraction) has taken place all along the track. We further assume that the material is isotropic and that, therefore, the situation has cylindrical symmetry. A reasonable displacement function for the region outside of the core is

$$R=\epsilon(\bar{r}/r^2), \quad (1)$$

where $\bar{r}$ is oriented radially outwards, but perpendicular to the track; and ϵ is a parameter describing the strength of the expansion (or contraction) along the core. A contraction would be described by $\epsilon<0$ while expansion corresponds to $\epsilon>0$. We consider the geometry shown in Fig. 17. The track is along the y axis; the z axis is perpendicular to the foil. This displacement function has the components

$$\begin{aligned} R_x &= \epsilon x/(x^2+z^2) \\ R_y &= 0 \\ R_z &= \epsilon z/(x^2+z^2). \end{aligned} \quad (2)$$

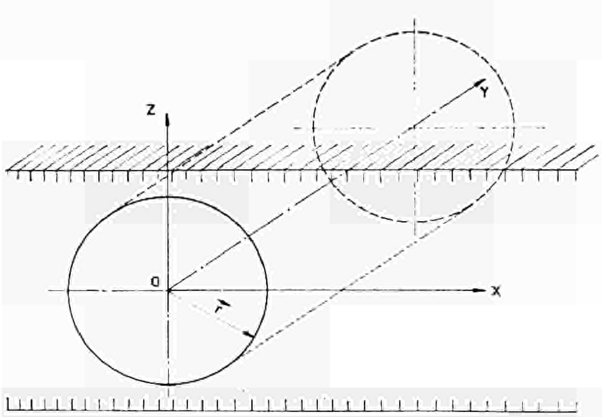

FIG. 17. The coordinate system for the displacement function of a fission track that lies parallel to the surface of the foil.

As a first approximation, we can discuss the contrast on the basis of the kinematical theory[11]; the conclusion will qualitatively be correct. The phase difference introduced by the strain is:

$$\alpha(x,z)=2\pi\bar{g}\cdot\bar{R}=px/x^2+z^2, \quad (3)$$

where $p=2\pi g_x\epsilon$. The assumption was made that the

[11] P. B. Hirsch, A. Howie, and M. J. Whelan, Phil. Trans. Roy. Soc. London **A252**, 499 (1960).

diffraction vector $\mathbf{g}$ is parallel to the foil plane, as is usually the case; g_x is the component of $\mathbf{g}$ along the x axis. If $\mathbf{g}$ is not in the foil plane

$$\alpha = 2\pi\epsilon[(g_x x + g_z z)/(x^2+z^2)]$$

usually

$$g_z \ll g_x.$$

From Eq. (3), the explanation of the experimental evidence for vanishing contrast in fission tracks parallel to $\bar{g}$ follows directly. It is similar to the condition $\bar{g}\cdot\bar{b}=0$ for a dislocation out of contrast. If the track is inclined with respect to the foil plane and the diffraction vector parallel to the foil, there is no total disappearance of contrast, no matter what $\bar{g}$ is. Using the column approximation one finds for the amplitude scattered by the column at $+x$

$$A(x) = \int_{z_1}^{z_2} e^{ip[x/(x^2+z^2)]} e^{2\pi isz} dz. \qquad (4)$$

The quantity s denotes a measure for the deviation from the exact Bragg condition.[11] For the column at $-x$, the argument of the first exponential would change sign. Although the integral has not been calculated it is clear that, in general, $A(x)\neq A(-x)$. If $spx>0$ the two arguments of the exponentials add up and in an amplitude phase diagram[11] we would have a wound-up spiral. For large z the spiral approaches asymptotically to a circle with radius $1/2\pi s$ because

$$\lim_{z\to\infty} [x/(x^2+z^2)] = 0.$$

For $spx<0$, on the other hand, the two arguments of the exponential subtract and we would have an unwound spiral. This means that the intensity of the diffracted beam would be larger at the side of the track for which $spx<0$; this will determine the image side. From the knowledge of the image side and of s, we can therefore deduce the sign of p. If, furthermore, $\bar{g}$ is known, it is clear that we can determine the sign of ϵ, i.e., we can tell whether the track core is expanded or contracted. The necessary experiments are now in preparation.

For small values of s the dynamical theory has to be used. According to Howie and Whelan[12] the amplitudes of transmitted (T) and scattered (S) waves obey the following set of differential equations:

$$dT/dz = (\pi i/t_e)Se^{2\pi i(sz+\alpha)},$$
$$dS/dz = (\pi i/t_e)Te^{-2\pi i(sz+\alpha)}, \qquad (5)$$

where t_e is the extinction distance, s a parameter describing the deviation from the exact Bragg condition, and α is given by $\alpha=\bar{g}\cdot\bar{R}$. In our case $\alpha = g_x\epsilon[x/(x^2+z^2)]$. The initial values are $T=1$ and $S=0$ for $z=0$.

We can now discuss the symmetry properties of the image by the use of a method proposed by Gevers[13] for discussing dislocation images which does not require solving the equation explicitly.

If anomalous absorption is neglected $\tau_e=\infty$ and for $s=0$ the image is symmetrical. This can be shown by the properties of the system (5) for positive and negative values of x. One can deduce for the intensities I_t and I_s, of the transmitted and of the scattered wave, that the relations hold

$$I_t(x)=I_t(-x) \quad \text{and} \quad I_s(x)=I_s(-x).$$

It is further clear that for $x=0$, $\alpha=0$ and therefore I_s and I_t are equal to the background intensities. The image has, therefore, to be double-peaked.

If anomalous absorption is not neglected the image is no longer symmetrical even for $s=0$ and *a fortiori* for $s\neq 0$. This is likely to be true in most cases. Machine calculations for the computation of image profiles are in progress. Oscillating contrast effects similar to those observed for dislocations are to be expected.

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[12] A. Howie and M. J. Whelan, Proc. Roy. Soc. (London) A263, 217 (1961).

[13] R. Gevers, Phys. Stat. Solidi (to be published).