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X-ray Studies of Polycrystalline Metals Deformed by Rolling. I. The Examination of the Harder Metals, Copper, Nickel and Iron

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The recently developed X-ray microbeam technique has been applied to the study of polycrystalline metals deformed by rolling. In this way it is possible to obtain information both about the mode of deformation of the material and about the nature of the cold worked state. Similarities in the behaviour of soft and hard metals are found and the results are discussed in terms of current theories of metallic strength.

This paper contains only the results of the investigations of typical hard metals, copper, nickel and iron. It is found that a limiting particle size is reached when these metals are heavily rolled, and that the particles are distorted.

1. Introduction

The results obtained in numerous previous researches in this field have not always been satisfactory, since they have been subject to an ambiguity of interpretation which has not been removed by recent experimental refinements. The authors have attempted to resolve some of the difficulties by applying the X-ray microbeam technique to the problem; this and other papers to follow describe the results obtained.

A survey of the behaviour of polycrystalline metals, deformed at room temperature by rolling, has been carried out to determine as far as possible the mechanism by which deformation occurs and the structure of the heavily cold-worked material. Metals of varying crystal structures and mechanical properties have been studied in an attempt to find unifying factors in their behaviour. The results of these experiments are presented in three papers. The first deals with the work on the harder metals copper, nickel and iron, and includes a short discussion of the results; the second paper contains a similar treatment of the experiments on the softer metals tin, cadmium, zinc and lead. In the final paper the overall picture of the deformation and structure of cold-worked metals is discussed and some dislocation mechanisms are examined.

X-ray back-reflexion methods have been used in these researches, and a transmission method was also used to provide additional information in the case of copper. X-ray cameras using beams of diameter ~ 1 mm., and the recently developed X-ray microbeam technique (Hirsch & Kellar, 1951; Gay *et al.*, 1951), have both been employed; the latter method is capable of yielding much more unambiguous information than has been obtained hitherto. A comprehensive study of the intermediate metal, aluminium, has been made by Hirsch & Kellar (1952) and by Hirsch (1952*a*, *b*) and the interpretation of microbeam photographs obtained in the present experiments is based largely on this earlier work.

2. Experimental methods and interpretation

If the deformed metal contains small discrete particles, a 'spotty ring' back-reflexion pattern can be obtained by irradiating a small number of grains. The mean particle volume may be found from the number of spots around a Debye–Scherrer ring by a modification of the method of Stephen & Barnes, while some deductions may be made about the distortion of the particles from the observed size and shape of the diffraction spots (cf. Hirsch & Kellar, 1952; Hirsch, 1952*a*). The size of the X-ray beam required to give spotty diffraction rings for a particular specimen depends primarily on the mean volume and distortion of the particles irradiated. In ordinary back-reflexion cameras the beam diameter is fixed at about 1 mm.; the lower limit of grain size which can be measured using such cameras and the method of counting spots is about 15–20 μ, but particles of much smaller dimensions can be measured using the smaller beam diameter of the microbeam technique.

A full account of this technique in back-reflexion work has already been published (Hirsch & Kellar, 1951; Gay *et al.*, 1951; Hirsch & Kellar, 1952); a similar experimental procedure has been used throughout this work. The lower limit of the volume of material which can be irradiated (and hence the lower limit of the particle size which can be measured) is determined primarily by consideration of the exposure time necessary to give a strong diffraction pattern; capillaries down to 17 μ in diameter have been used in the back-reflexion work with a corresponding exposure time of about 55 hr. using the high-intensity X-ray generator. This restricts the lower limit of particle size measurable by back-reflexion methods to about

1 μ in favourable cases. A microbeam transmission method has been used to decrease this lower limit; the theory of the interpretation of microbeam transmission photographs will be given elsewhere; only the relevant experimental results are quoted here.

For the harder metals it is necessary to use the microbeam technique even at the smallest deformations, since the decrease in particle size is initially very rapid with increasing deformation. It has not been possible to achieve resolution of the back-reflexion diffraction rings from heavily worked specimens of the harder metals even when the smallest microbeam experimentally available is used. However, even at the heaviest deformations continuous back-reflexion rings are not obtained on microbeam photographs, but the pattern consists of a number of arcs. It is possible to set an upper limit to the particle size in the material from measurements of the angular extent of these arcs, as will now be shown.

In this treatment it is assumed that each of the unresolved arcs found on photographs of heavily rolled specimens is formed by the superposition of the reflexions from one original grain; for this grain the normals to the reflecting planes cover a range of orientation due to the deformation the grain has suffered. The first stage is the calculation of the fraction of the mosaics within the deformed grain which are in a reflecting position.

If a beam of monochromatic X-rays, of divergence $d\theta$, is incident on the specimen, a reflexion can take place only if the normals to the reflecting planes lie between two cones of semi-angle $(\frac{1}{2}\pi-\theta)$ and $(\frac{1}{2}\pi-\theta+d\theta+\Delta)$, where Δ is a term which takes into account the wavelength spread of the radiation and the possible range of reflecting angles due to distortion or small particle size; θ is the Bragg angle for the reflexion. In Fig. 1 the shaded cap represents the

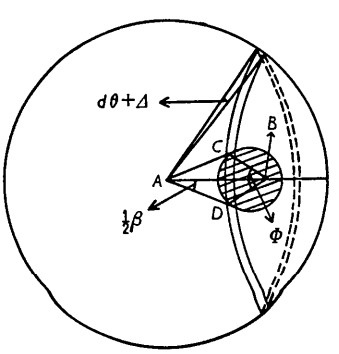

Fig. 1. Diagram illustrating the calculation of the fraction of reflecting particles within a deformed original grain.

distribution of normals of the particular reflecting planes within the deformed original grain. The area of intersection (CD) of this cap with the two limiting cones represents the particles in reflecting positions. From the geometry of the figure it follows that the fraction of the particles within the deformed grain which can reflect is

$$\frac{\cos\theta(d\theta+\Delta)\varphi}{4\pi\sin^2\frac{1}{4}\beta}.$$

Here β is taken as the width at half maximum of the angular distribution curve of the normals in the deformed grain, and φ is the angle subtended by the extreme reflecting normals (CD) at the centre of the circular section cut by the cones.

Now the volume irradiated is given by the product of the area of cross section of the beam (A) and the depth of penetration of the X-rays (l); but the effective volume contributing to the reflexion along a particular arc will be V, the volume of an original grain. This is true provided the dimensions of the incident beam are large enough to cover an original grain and the depth of penetration is of the order of the original grain size; these conditions are usually satisfied.*

Thus the number of diffraction spots (N_A) which are found on the arc corresponding to this deformed grain is given by

$$N_A=\frac{V}{v}\frac{\cos\theta(d\theta+\Delta)\varphi}{4\pi\sin^2\frac{1}{4}\beta}, \qquad (1)$$

where v is the mean volume of the mosaics within the deformed grain.

Since the incident and reflected beams and the normal to the reflecting planes all lie in the same plane, $\varphi=\gamma=$ total length of the arc on the photograph. β, in general, is unknown since the section of the spherical cap (Fig. 1), representing the normals to the deformed grain cut by the cones defining a reflexion, is not equal to the maximum half-width of the distribution curve. However, on the average, any section is connected with the maximum section by a factor of order unity and hence β can be found from the formula for angular misorientation (cf. Hirsch & Kellar, 1952) which may be written

$$\sin\tfrac{1}{2}\beta=\cos\theta\sin\tfrac{1}{2}\gamma. \qquad (2)$$

Equation (1) can then be transformed into

$$N_A=\frac{V}{v}\frac{(d\theta+\Delta)\gamma\cos\theta}{2\pi[1-(1-\cos^2\theta\sin^2\frac{1}{2}\gamma)^{\frac{1}{2}}]}$$
$$\doteq\frac{V}{v}\frac{(d\theta+\Delta)\gamma}{\pi\cos\theta\sin^2\frac{1}{2}\gamma}. \qquad (3)$$

This relation leads to the direct calculation of the mean particle volume from the number of spots on a given arc since all the variables can be evaluated except v and Δ. V, the original grain volume, can be determined for the annealed material by the standard method of counting spots. The term Δ is usually small;

* When very fine beams are used to examine material of coarse initial grain size, A is equal to the area of cross section of the beam and, since l is not known, the double-exposure method must be employed.

for large values of $d\theta$ it may be neglected, though an estimate can be made from the shapes of the diffraction spots (Hirsch, 1950).

This method must be considered to be more strictly applicable in the present circumstances than the modification of the Stephen & Barnes method since the latter assumes that the diffracting particles are randomly orientated; experimentally it is found that the mosaics have a preferred orientation and that the diffraction spots are clustered into arcs. However only small errors result from the use of the Stephen & Barnes treatment provided that the spotty arcs are fairly evenly distributed around the ring. This can be demonstrated by the use of both formulae in the evaluation of the particle size from a photograph, showing a single spotty arc, of material of initial grain size which satisfies the above condition.

The results of the foregoing discussion may now be applied to the evaluation of the upper limit of particle size from the measured extent of the unresolved arcs. Equation (3) gives the number of spots along an arc of given angle γ. Each of these spots has a tangential length (S_T) on the film. For complete continuity all the spots on the arc must overlap; that is, the length of the arc must be less than the product of the number of spots on the arc and their tangential breadths. Then

$$R_0 \tan 2\theta . \gamma < N_A S_T ,$$

where R_0 is the specimen–film distance.

Substituting for N_A from above,

$$v < \frac{V(d\theta+\Delta)}{\pi \cos\theta \sin^2 \tfrac{1}{2}\gamma \tan 2\theta} \cdot \frac{S_T}{R_0} . \qquad (4)$$

From this inequality the upper limit of v can be found if a value can be assigned to S_T; all other variables are calculable. It is impossible, of course, to make a direct estimate of S_T; however, the radial broadening (S_R) of the arc is always measurable (provided that the α-doublet is satisfactorily resolved) and an approximate value of S_T can be calculated from the measured value of S_R, if it is assumed that the mosaics are equiaxed (Hirsch, 1952*a*). Experimental evidence shows that this assumption is generally valid (see § 4(*a*), (iii), later).

The value of the upper limit to the particle size calculated from the inequality can be made smaller by decreasing the initial grain size, V. If V is decreased, however, it is also necessary to decrease the beam diameter; otherwise a large number of grains in which there is widely misoriented material will be irradiated. This will cause overlapping of the arcs; the Debye–Scherrer ring will be complete and no estimate of γ is possible. Thus this method, too, is limited by consideration of exposure time; under favourable conditions it may be possible to reduce the upper limit of particle size measurable to about 7×10^{-5} cm., but this has not been achieved in the present experiments.

Both the method of counting spots and the method using the condition for the continuity of the arcs have been used for the determination of particle size in the back-reflexion work on the harder metals. The measurement of angular misorientation has been carried out using the relationship (2) between β and γ. Further important data may be obtained from the measurement of the size and shape of the spots on the microbeam photographs.

Hirsch (1952*a*) has made an analysis of the dependence of the shapes of spots on microbeam photographs on the size, shape and distortion of the diffracting elements and the experimental conditions, for example, the divergence of the X-ray beam. He has shown that it is possible to obtain reasonable estimates of the physical broadenings of the spots due to the shape and distortion of the diffracting particles. His interpretation and terminology have been followed throughout. From the reciprocal-lattice treatment, it follows that the tangential (S_T) and radial (S_R) measured widths of the diffraction spots can be related to three angular functions $d\varphi_1$, $d\varphi_2$ and $d\varphi_3$ which are associated with the physical broadening of the spots, due to distortion or small-particle-size effects. $d\varphi_1$ and $d\varphi_2$ are related to the angular radial breadth of the spot, whilst $d\varphi_3$ is related to the angular tangential breadth of the spot. These three functions are particularly convenient for the interpretation of the physical breadths in terms of particle size or distortion effects, and all discussions of spot shape in this paper are made in terms of them.

There are then, three main measurements which can be made from the microbeam photographs: the grain size, the angular misorientations, and the physical broadening of the diffraction spots. The results of these measurements for copper, nickel and iron are presented in § 4.

3. Preparation of specimens

All the specimens were in the form of bars; the initial annealed grain size of the metals was about 20–30 μ. The deformation was carried out by rolling; the percentage deformation quoted is the ratio of the thickness measured after deformation to the original thickness of the material.

The copper specimens were of 99·99% purity (Johnson, Matthey Ltd, J.M. 30, 31). After rolling, the specimens were etched with a solution of one part hydrochloric acid and one part water, saturated with ferric chloride, until the surface layers ($\sim$ 0·1–0·2 mm.) had been removed.

The nickel specimens were of 99·6% purity and were obtained from Henry Wiggin Ltd. After deformation they were etched with dilute nitric acid to a similar depth.

The iron specimens were of 99·99% purity (Johnson, Matthey Ltd, J.M. 845). After rolling they were etched with a 2% solution of alcohol in nitric acid to remove the surface layers. For these specimens at

small deformations it is particularly important that the surface layers be removed if consistent results are to be obtained.

4. Experimental results

(*a*) *Copper*

(i) *Particle size.*—The undeformed, annealed specimens of copper were of fairly large grain size, so that a spotty ring pattern was obtained using an ordinary back-reflexion camera. For small deformations normal back-reflexion photographs show that the pattern spreads out into arcs which form almost continuous Debye–Scherrer rings; at heavier deformations the rings become continuous and quite diffuse.

At small deformations, by the use of an X-ray microbeam of the appropriate diameter, it is possible to resolve the diffraction pattern into short arcs around the Debye–Scherrer ring. In these arcs there is a definite substructure, which is often resolved into spots linked by a continuous background. Sometimes, however, the spots are not so well resolved within the arcs and only ill-defined intensity variations can be seen. As the deformation is increased the angular extent of the arcs increases, whilst the resolution of the structure within the arcs decreases. At heavier deformations the arcs are of uniform intensity and there is no resolution even when using a very small microbeam.

From this short account of the changes observed on the back-reflexion photographs with increasing deformation of the specimen, it is clear that the method of determining particle size by counting spots around a diffraction ring is very limited in its application; the results for deformations 2·9% and 7·3% are given in Table 1. An estimate of the upper limit (1·7 μ) of particle size for a heavily (49%) deformed specimen may be found from the length of the unresolved arc on the photograph by the method described earlier. Further, a lower limit (0·03 μ) to the particle size for this specimen may be obtained by attributing the whole of the experimental line breadth to the small-particle-size effect (see § 5). It should be emphasized that this new method of determination demonstrates that the particle size in heavily cold rolled copper is less than 1·7 μ, independently of any assumptions as to the cause of line broadening in worked metals.

Table 1. *Mean grain size of deformed copper and nickel specimens*

Copper

Deformation (%)	Mean grain size (cm. $\times 10^{-4}$)
0	24·6
2·9	4·2
7·3	2·8
49·0	$0{\cdot}03 < t < 1{\cdot}7$
Rolled foil ($\sim$ 50% deformation)	$0{\cdot}6 \pm 0{\cdot}1$ (by transmission)

Nickel

Deformation (%)	Mean grain size (cm. $\times 10^{-4}$)
0	21·0
3·5	4·8
6·6	2·8
33·0	$0{\cdot}04 < t < 1{\cdot}9$

This result has been confirmed for the case of heavily rolled copper foil by the use of a microbeam transmission technique. It has been explained in an earlier section that the non-resolution of the arcs obtained on microbeam back-reflexion photographs is due to the overlapping of the separate reflexions from individual mosaics, and that the use of beams of diameters less than about 20 μ to obtain separate reflexions from particles of size less than 1 μ is limited by considerations of exposure time. However, by the use of transmission methods the difficulty of exposure time may be overcome for crystallites of size greater than about 10^{-5} cm., whilst the breadth of individual reflexions is also reduced at the lower Bragg angles. The latter effect may not lead to resolution, however, since the angle between reflexions on the Debye–Scherrer ring from crystallites inclined at small angles to one another also decreases at lower Bragg angles (see equation (2)). A full account of the microbeam transmission method will be published elsewhere.

Transmission photographs of copper foils 6 μ thick have been obtained using beams of diameter down to 8 μ. These foils were produced from annealed sheets of pure copper, 12 μ thick, by 'sandwich' rolling between sheets of commercial aluminium, and thus were in a highly worked state. The thickness was determined by weighing, and the foils were etched slightly before examination.

The microbeam transmission photographs show spotty rings (Fig. 3) and the particle size may be determined as $6 \times 10^{-5} \pm 1 \times 10^{-5}$ cm. It may be argued that this particle size is not likely to be the same as that found in the bulk material, at similar deformations, where the grains may be more constrained. However, examination of the foils by microbeam back-reflexion methods gives a pattern exactly similar to that obtained from the bulk material.

(ii) *Misorientations.*—As has been explained above, a measure of the total range of angles covered, after deformation, by material belonging to one original grain, is given by the angular extent of the diffraction arc on the photograph. Further measurements may be made using the same procedure to determine the relative misorientation of the two particles contributing to adjacent reflexions; this is not necessarily the angle between adjacent mosaics within the deformed grain.

Table 2 shows a series of values for copper at various stages of deformation; in this table, the bracketed value is uncertain because of the difficulty of experimental measurement. From this table it is seen

Table 2. *Misorientations in deformed grains*

Deformation (%)	Total angular misorientation within one original grain (°)	Relative misorientation of two grains contributing to adjacent reflexions (′)
Copper		
2·9	1½	15
7·3	2	18
10·8	3½	(25)
19·6	6	—
28·9	8	—
49·0	15	—
Nickel		
3·5	1	10
6·6	3½	15
10·6	5	—
33·0	7½	—
Iron		
1·0	4½	23
7·8	6½	23
22·0	6½	30
38·0	12½	31
57·0	14	27
80·0	19½	—

that the total angular spread in orientation of the mosaics increases with increasing deformation; the experimental data for the relative misorientations are not sufficient to allow any conclusion to be drawn.

It is interesting to note that even at the heaviest deformation, a complete Debye–Scherrer ring is not obtained; discrete arcs are still present on the photograph although no structure can be distinguished within them. This effect is not due to preferred orientation, for when a similar photograph is taken of a different area of the specimen the relative positions of the arcs are quite different.

(iii) *Measurement of spot shapes.*—On photographs from lightly deformed specimens the spots on the Debye–Scherrer rings have appreciable breadths. At small deformations it is possible to measure values for both the radial and tangential spot widths averaged around the diffraction rings. At heavier deformations, it is possible to measure only the radial breadths, provided that the α_1 and α_2 reflexions are resolved; this condition is satisfied on all the photographs. Although for some specimens the reflexions have not been completely resolved all around the ring, it has still been possible to measure the spot shapes where resolution does occur. Thus, although it may not be possible to obtain a value for the particle size from a spot count, it may still be possible to obtain a value for the tangential spot broadening.

Values of the $d\varphi$'s may be calculated from these measurements of spot shapes. Fig. 2 shows these values plotted as a function of the deformation for both lines 19 and 20 for copper. The broken curves indicate the trends of the measurements; the physical breadths increase with increasing deformation.

These measurements have been made by averaging the spot shapes around the diffraction ring. It can be shown (Hirsch, 1952*a*) that if $d\varphi_1/d\varphi_2 \sim \tan\theta$, from the averaged measurements, then the average reciprocal volume has spherical symmetry. The values of

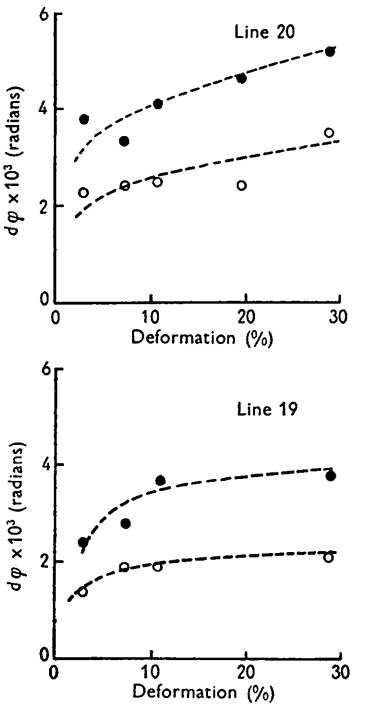

Fig. 2. Values of $d\varphi$'s, determined from spot-shape measurements on microbeam photographs of deformed copper specimens, plotted as a function of the amount of deformation. Points ○ represent values of $d\varphi_2 = d\varphi_3$; points ● represent values of $d\varphi_1$. The broken lines indicate expected trends in the values.

$d\varphi_1$ and $d\varphi_2$ in the figure show that the ratio is not much less than 2; the value of tan θ under the present arrangement is 2·5, and so it appears that the average reciprocal volume is roughly spherically symmetrical.

This does not imply that this distribution surrounds the reciprocal point for a given reflexion. To determine the distortion of a particular particle it is necessary to find the physical broadening from measurements on an individual spot. In this case $d\varphi_2$ may not be put equal to $d\varphi_3$ and in general the complete determination of the magnitudes of the $d\varphi$'s is not possible (Hirsch, 1952*a*).

The results in Table 3 show that the values of the breadths of typical spots are very close to the averaged values; hence the asymmetry of individual distributions around reciprocal-lattice points does not seem to be very large, i.e. the particles are probably equiaxed.

(iv) *Recovery.*—The specimens of copper were examined nine months after rolling to investigate any possible recovery effects. The photographs show that no detectable change in the metal structure has occurred in this time. The values of the $d\varphi$'s measured from photographs taken nine months after rolling do

Table 3. *Values of $d\varphi$ for typical spots for several copper specimens**

Deformation (%)	Values of $d\varphi$'s (radians $\times 10^{-3}$)	Line 19; Cu $K\alpha$ radiation
2·9	$d\varphi_3 = 1{\cdot}8$; $d\varphi_1 + d\varphi_2 = 4{\cdot}2$	Typical
	$d\varphi_3 = 1{\cdot}4$; $d\varphi_1 + d\varphi_2 = 3{\cdot}8$	Average
7·3	$d\varphi_3 = 1{\cdot}7$; $d\varphi_1 + d\varphi_2 = 4{\cdot}6$	Typical
	$d\varphi_3 = 1{\cdot}9$; $d\varphi_1 + d\varphi_2 = 4{\cdot}7$	Average
10·8	$d\varphi_3 = 1{\cdot}7$; $d\varphi_1 + d\varphi_2 = 5{\cdot}0$	Typical
	$d\varphi_3 = 1{\cdot}9$; $d\varphi_1 + d\varphi_2 = 5{\cdot}6$	Average

* For these measurements $d\varphi_2 \neq d\varphi_3$ (see § 4*a* (iii)).

not show any significant consistent decrease; the variations that occur may be attributed to the inaccuracy of the experimental measurements.

The non-occurrence of self recovery in solid specimens of copper has been noted by Megaw & Stokes (1945) using conventional line-broadening measurements.

(*b*) *Nickel*

(i) *Particle size.*—The features shown on photographs from nickel specimens are very similar to those of corresponding photographs from copper specimens.

The photographs shown in Fig. 4 were taken with an ordinary back-reflexion camera. Fig. 4(*a*) shows the random arrangement of spots around the Debye–Scherrer rings, obtained from an annealed nickel specimen. After 6·6% deformation the diffraction arcs have spread out until almost continuous rings are obtained (Fig. 4(*b*)). With heavy deformations (33%) the Debye–Scherrer rings on the photograph are continuous and quite diffuse (Fig. 4(*c*)).

Figs. 5(*a*) and 5(*b*), of the 6·6% and 33% rolled specimens respectively, were taken using the appropriate microbeam diameters. In Fig. 5(*a*) is seen the resolution into spotty rings of the arcs shown in Fig. 4(*b*); in Fig. 5(*b*) the discrete unresolved arcs from the heavily rolled specimen are shown.

With nickel, as with copper, it is possible to determine the mean grain volume only at small values of the deformation. Numerical values are given in Table 1 together with the limits of particle size in the heavily deformed nickel specimen.

(ii) *Misorientations.*—The overall misorientation of the material within a deformed grain increases with increasing deformation (Table 2).

(iii) *Measurement of spot shapes.*—The calculated values of the $d\varphi$'s for lines 19 and 20 for Cu $K\alpha$ radiation are shown in Fig. 6 plotted against deformation. The expected increase in the values at heavier deformations is not apparent for line 19; this may be due to the difficulties of experimental measurement.

(*c*) *Iron*

(i) *Particle size.*—The specimens were examined using Fe $K\alpha$ radiation (filtered through manganese dioxide). The photographs are somewhat different from those obtained from copper and nickel in that structure is clearly visible within the diffraction arcs, in photographs taken with a microbeam of the appropriate diameter, at deformations as large as 57%; even at 80% deformation spots may be seen at the *ends* of the arcs. At these heavy deformations a faint continuous background is found on the photographs; it extends over a large portion of the ring and is weak by comparison with the arcs. By counting the resolved spots found in the arcs, a variation of particle size with deformation is found (Fig. 7); it appears that a limiting value is reached at about 1–2 μ.

Calculation of the limits of grain size from the unresolved arcs for an 80% deformed specimen gives

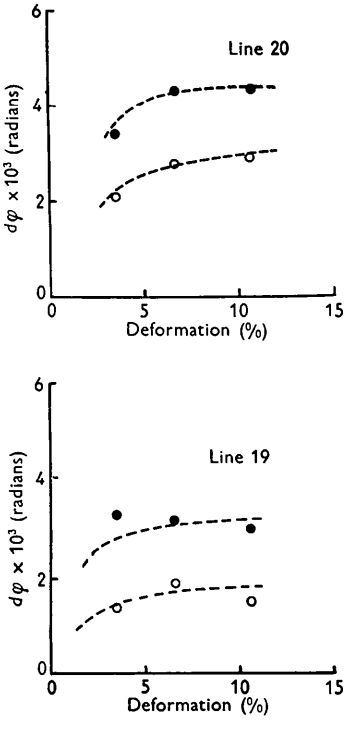

Fig. 6. Values of $d\varphi$'s, determined from spot-shape measurements on microbeam photographs of deformed nickel specimens, plotted as a function of the amount of deformation. Points ○ represent values of $d\varphi_2 = d\varphi_3$; points ● represent values of $d\varphi_1$. The broken lines indicate the expected trends in the values.

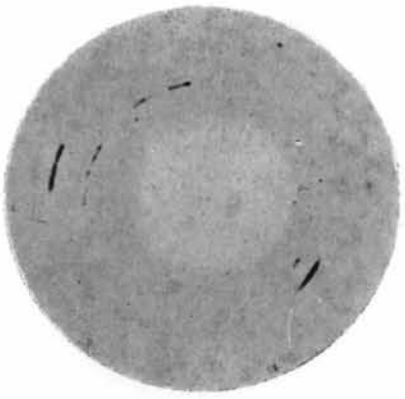

Fig. 3. X-ray microbeam transmission picture of rolled copper foil (thickness 6 μ) taken with a beam of diameter 17 μ using Cu $K\alpha$ radiation; lines 111 and 200.

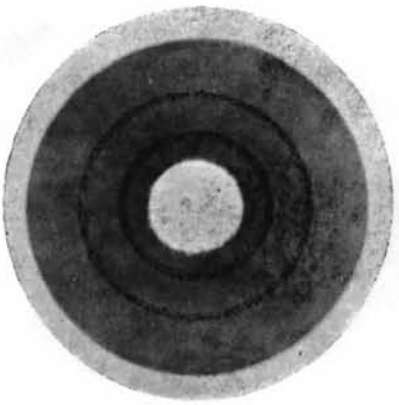

Fig. 4 (*a*).

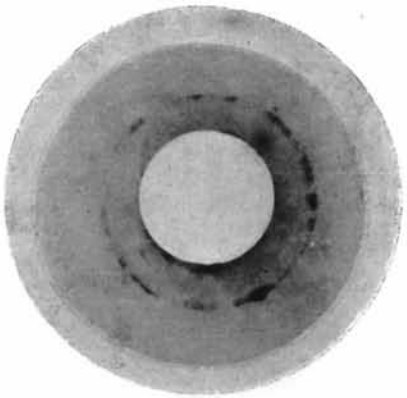

Fig. 5 (*a*).

Fig. 4 (*b*).

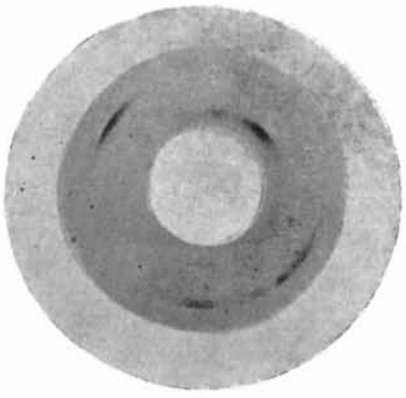

Fig. 5 (*b*).

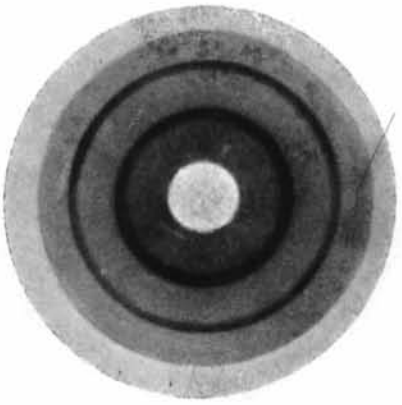

Fig. 4 (*c*).

Fig. 5. (*a*) Microbeam back-reflexion photograph of same specimen from which Fig. 4(*b*) was taken; beam diameter 100 μ, lines 19 and 20, Cu $K\alpha$ radiation. (*b*) Microbeam back-reflexion photograph of same specimen from which Fig. 4(*c*) was taken; beam diameter 30 μ, lines 19 and 20, Cu $K\alpha$ radiation.

Fig. 4. (*a*) Ordinary back-reflexion photograph of annealed nickel specimen (grain size 21·0 μ); lines 19 and 20, Cu $K\alpha$ radiation. (*b*) Ordinary back-reflexion photograph of 6·6% rolled specimen of nickel; lines 19 and 20, Cu $K\alpha$ radiation. (*c*) Ordinary back-reflexion photograph of 33·0% rolled specimen of nickel; lines 19 and 20, Cu $K\alpha$ radiation.

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these as 1·0 μ and 0·02 μ. The upper limit is probably slightly too low owing to the omission of the term Δ in equation (4); in this case the angle Δ over which a particle can reflect is unknown.

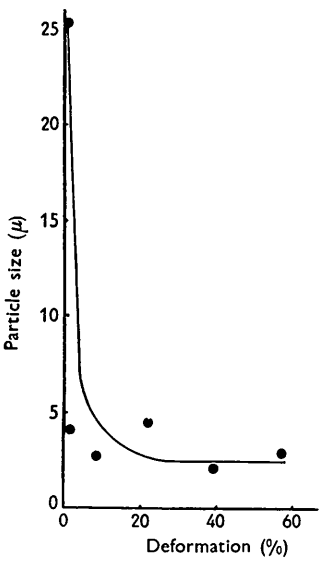

Fig. 7. Variation of particle size with deformation for iron.

(ii) *Misorientations.*—Values of the total and relative misorientations are given in Table 2; at comparable deformations they are very similar to those for copper and nickel.

(iii) *Measurement of spot shapes.*—The values of the $d\varphi$'s are shown in Fig. 8 plotted against the deformation. The increase in $d\varphi_3$ with deformation is clearly shown although, owing to scarcity of data, a similar trend for $d\varphi_1$ is not so marked. Further, it appears, that the ratio $d\varphi_1/d\varphi_2$ is quite different from tan θ ($\sim 3{\cdot}2$), in contrast to the results for copper and nickel.

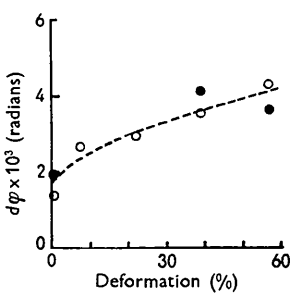

Fig. 8. Values of $d\varphi$'s, obtained from spot-shape measurements on microbeam photographs of deformed iron specimens, plotted as a function of the amount of deformation. Points ○ represent values of $d\varphi_2 = d\varphi_3$; points ● represent values of $d\varphi_1$. The curve indicates a trend in the values of $d\varphi_2 = d\varphi_3$ only.

5. Physical interpretation

In all discussions of this problem it is assumed that cold-worked metals contain regions of coherently diffracting material. Some previous workers (Stokes, Pascoe & Lipson, 1943) have held the view that these 'particles' are distorted, and that this distortion is the primary cause of the line broadening observed with cold-worked metals; others (Wood, 1939) have suggested that the broadening is due largely to the effect of the small size of the particles which are perfect. Many experiments have been conducted to test these two hypotheses but no conclusive evidence has been presented.

In the present experiments it has been possible to measure both the mean particle volume and the broadening of individual diffraction spots for certain specimens. For these specimens it is possible, by considering limiting shapes of the particles, e.g. equiaxed, rods, or lamellae, to predict the relative importance of the two types of broadening, i.e. to determine whether the particles are distorted or not (cf. Hirsch, 1952*b*). Applying this treatment, it may be shown that for any specimen for which the mean grain volume is known, distortion broadening is certainly present; however, in some cases, the particles may be in the form of lamellae so that the broadenings in one direction may be due to small-size effects. If it is assumed that all the broadening is due to elastic stresses, then the range of stresses within the particles, calculated from the measurements of broadening, is of the order of the yield strength of the material; a similar result has been obtained by Megaw & Stokes (1945).

For specimens for which it is not possible to obtain unequivocal measurements of the mean grain volume, these arguments cannot be used. Nevertheless, it is to be expected, from the results on the lightly worked specimens, that the particles within the heavily rolled specimens are distorted. The evidence of the transmission experiments on the rolled copper foil supports this interpretation of the experimental data for the bulk specimens, for the particle size in this specimen is about 6×10^{-5} cm. in contrast to the value of 3×10^{-6} cm. which it is necessary to assume if all the line broadening is to be explained by small-size effects alone.

These experiments suggest, therefore, that distorted particles of size greater than 10^{-5} cm. are formed within the cold-worked metals during or immediately after rolling. Further, they give some indication of the method of formation of these particles. The angular spread of the diffraction arcs shows that considerable angular misorientations must be associated with the deformed original grains; these are formed when the original grains suffer plastic curvature. However, the intensity variations within the arcs indicate that there are regions of low distortion (particles) separated by regions of higher distortion. A discussion of possible mechanisms for the formation of such regions is given in the third paper of this series. No direct evidence is obtained of the existence of such particles for the heavily rolled bulk specimens, but it is probable that

the process is fundamental and occurs to some extent at all stages of deformation.

No evidence of spontaneous recrystallisation after rolling was found for any specimen, nor was there any evidence of self-recovery in copper specimens. Thus the texture formed in these metals during or immediately after deformation at room temperature is quite stable, in sharp contrast to the behaviour of the softer metals which are to be discussed in the next paper.

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X-ray Studies of Polycrystalline Metals Deformed by Rolling. II. Examination of the Softer Metals, Tin, Zinc, Lead and Cadmium

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The examination of the softer metals, tin, zinc, lead and cadmium after deformation by rolling, is described. X-ray back-reflexion methods, employing both beams of normal diameter and micro-beams, have been used.

In the initial stages of deformation a mosaic structure is found within the deformed grains of the metal and the size of the mosaics reaches a lower limiting value, dependent on the particular metal. At heavy deformations these metals spontaneously recrystallize. The effects upon the mosaic structure of annealing at low temperatures and of recovery at room temperature have been studied. Some evidence is presented concerning the nature of recrystallization nuclei.

1. Introduction

Most of the previous investigations of the structure of cold-worked metals have been concerned with the behaviour of the harder metals of high melting point; little attention has been devoted to the deformation of those melting at lower temperatures (~300–400° C.). The experiments to be described in this paper are complementary to those described for the harder metals in a previous paper (Gay & Kelly, 1953).

In these experiments, the soft metals selected for examination were cadmium and zinc, representatives of the hexagonal metals; the tetragonal modification of tin; and lead, a face-centred cubic metal. It is found that the behaviour of these metals at heavy deformation differs from that of the harder metals, in that they exhibit spontaneous recrystallization. However, the experiments to be described here have been mainly concerned with the textures of the materials before the onset of recrystallization. The authors have attempted to determine the processes by which deformation, recovery and recrystallization occur, and to this end three main groups of experiments have been carried out.

First, the metals have been examined immediately after deformation for various degrees of reduction (cf. § 3). Then the changes which occurred, with time after deformation, within the same volume of a particular specimen were observed, and finally the effects of annealing at low temperatures were studied. Complete investigations of this type have been carried out for the m als tin, zinc and lead, but for cadmium such detailed studies have not been undertaken since it appears that its behaviour is similar to that of the other metals.

In this paper, the results of the experiments and a