DISLOCATION DENSITY AND FLOW STRESS OF COPPER*

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Measurements of the dislocation density by a chemical etching technique in high-purity copper single crystals were related to the applied flow stress and yield stress during easy glide deformation at room temperature. The results were interpreted by the model of Seeger for pure metals in which the flow stress consists of a temperature, strain rate dependent component arising from the thermally activated motion of glide dislocations through a forest, and a dislocation density dependent component arising from the elastic stress interactions of glide dislocations. The activation energy calculated from the forest mechanism was 0.9 eV, consistent with the formation energy of jogs due to screw dislocation intersection. The flow stress was found to increase parabolically with dislocation density in accord with the Seeger model. Observations of mechanical polygonization after plastic stress were also made.

INTRODUCTION

Recently, chemical etching techniques have been developed which reveal dislocation sites in copper by forming pits on oriented crystal surfaces.(1–3) In the present investigation, changes in dislocation density and distribution were observed during single glide (Stage I) deformation using an etching method which developed pits on (111) oriented faces of high-purity copper crystals with the triangular edges of the pits parallel to the (110) glide directions in the crystal. Previous work has shown that there is essentially a 1:1 correspondence between dislocations and etch pits with this technique.(2) Measurements of the increase in dislocation density were related to the shear flow stress and shear strain for the initial stage of tensile plastic stressing using crystals suitably oriented to provide a single operative glide system. Changes in the strain rate of the tensile loading were used to determine the dependence of the yield stress upon glide dislocation interactions and a thermally activated flow mechanism.

EXPERIMENTAL METHODS

The crystals used in the present investigation were supplied by Metals Research, Ltd., England in the form of bars 5–7 in. long with a square cross-section 3 in. on a side. They were of Johnson-Matthey high-purity copper with an estimated purity of 99.999 per cent. The crystals were prepared by seeded growth to have a set of surfaces within 2° of a {111} orientation and a tensile axis approximately 7° from a [110] pole within the basic stereographic triangle. With this tensile orientation, a single glide system would be operative in the shear strain range under examination.

Before etching, the crystals were carefully annealed in nitrogen for 150 hr using a thermal cycling method which periodically varied the annealing temperature between 1050°C and 750°C. Thermal cycling had previously been reported more effective than isothermal
annealing in reducing the dislocation density in copper crystals. After annealing, the crystals were electropolished in a 60 per cent ortho phosphoric acid-40 per cent water solution with a cell voltage of 1 V, followed by a water rinse and then immersion in the etchant for 4-30 sec. The etchant used was a modification of etch solutions reported by Lovell and Wernick and Livingston with the composition 0.5 ml liquid bromine, 25 ml glacial acetic acid, 30 ml HCl, and 125 ml water. The etching time was found to vary in inverse proportion to bromine content. After etching, the crystals were rinsed in alcohol and dried in a warm air stream.

Stressing of the crystals was accomplished with an Instron tensile machine using strain rates of $0.6 \times 10^{-5} \text{ sec}^{-1}$ and $1.2 \times 10^{-4} \text{ sec}^{-1}$. The corresponding strain was measured by two Tatnall electrical resistance strain gages (type TXI-32A) mounted axially on opposite nonoriented surfaces of the crystal and connected in series. The strain gages were carefully bonded about 2 in. from the center of the crystal in order to permit immersion of the central section in the electropolishing and etching solutions. Preliminary loading measurements with this method showed that the difference in strain recorded by the two gages due to bending of the crystals in the loading fixture was less than $\pm 5 \times 10^{-6}$ in/in. Since the yield stress indicated by the gage technique was less than that detected by cross-head extension, the effect of the bonded gages on the flow characteristics of the crystals appeared to be negligible. Strain and load signals were fed into the Instron X-Y chart drive amplifier to obtain an automatically plotted tensile stress-strain curve. The maximum resolution of the apparatus was approximately 1 y/in$^2$ (0.707 g/mm$^2$) stress and $5 \times 10^{-6}$ in/in. strain. The resolved yield stress, defined as the minimum flow stress to produce a detectable strain of $5 \times 10^{-6}$, was obtained graphically from the stress-strain curve.

**RESULTS**

Shear stress-strain curves

Copper crystals were plastically stressed at room temperature within the single glide stage of the stress-strain curve at strain rates of $0.6 \times 10^{-5} \text{ sec}^{-1}$ and $1.2 \times 10^{-4} \text{ sec}^{-1}$ to a maximum shear strain of 2.5 per cent. At various strain levels, deformation was interrupted and the stress unloaded in order to measure dislocation density. A typical tensile repeated loading curve is shown in Fig. 1 with resolved shear stress-strain values computed for the operative glide plane. The resolved stresses of interest are the flow stress $\tau$, defined as the maximum shear stress prior to unloading, and the yield stress $\tau_y$ obtained upon reloading. Figure 2 shows a plot of the flow stress and the yield stress obtained from tensile curves as a function of residual shear strain and strain rate, the points indicating interruptions in the loading curve. It is apparent from Figs. 1 and 2 that strain hardening occurred during single glide extension, an initially high hardening coefficient $\theta = d\tau/d\varepsilon$ approaching a steady value of about 1 kg/mm$^2$ as deformation progressed.

Although individual dislocation motion has been found in copper with extremely small applied stress pulses, macrosopic plastic flow was not detected below resolved shear stresses of 19 g/mm$^2$ ($\varepsilon_1 = 0.6 \times 10^{-5} \text{ sec}^{-1}$) and 24 g/mm$^2$ ($\varepsilon_2 = 1.2 \times 10^{-4} \text{ sec}^{-1}$) for annealed crystals. These yield stresses were well

![Fig. 1. Resolved shear stress-strain curves obtained with interrupted deformation of copper crystal.](image-url)
below the established range for the critical resolved
shear stress, but were in good agreement with the
minimum shear stress of 18–20 g/mm² required for dis-
location multiplication recently observed by Young.\(^{(4)}\)
Figure 2 shows that the yield stress measured upon
reloading increased with plastic strain in a manner
similar to the flow stress. The relationship between
the flow stress and the yield stress, shown in Fig. 3,
can be expressed in the form:
\[
\frac{\tau_y - \tau_0}{\tau} = 0.33
\] (1)
where \(\tau_0\) is the value of \(\tau\) for the undeformed crystal
\((\sim 20 \text{ g/mm}^2)\). For initial increments of plastic stress,
it was apparent that \(\tau_y\) varied linearly with \(\tau\) and was
independent of strain rate; however, continued pre-
stressing above 150 g/mm² resulted in smaller increases
in the yield stress.

**Dislocation density**

Etching of undeformed crystals revealed that most
of the dislocations were arrayed in a well defined sub-
boundary structure with an average density of about
10⁵ cm⁻². The etch pit count or “intersection density”
was multiplied by a factor of 2 in order to obtain the
ture length density.\(^{(6)}\) It was found that application
of shear stresses below the measured yield stress pro-
duced no observable change in the dislocation density.

Values of the dislocation density \(N\) for plastically
stressed crystals are plotted as a function of the shear
flow stress $\tau$ and yield stress $\tau_y$ in Fig. 4 for both strain rates. It was found that $\tau$ and $\tau_y$ varied nearly linearly with $N^{1/2}$ in the initial stages of deformation, obeying the relations:

$$
\tau_y = \tau_0(\varepsilon) + A N^{1/2}
$$
$$
\tau = \tau_0'(\varepsilon) + A' N^{1/2}
$$

(2)

where the hardening coefficients had the values: $A = 1.7$ g/cm, $A' = 3.2$ g/cm and were independent of the strain rate. Similar results for the variation of $\tau$ with $N^{1/2}$ and $\dot{\varepsilon}$ have been reported for NaCl crystals with a value of 7.2 g/cm obtained for the constant $A'$. The limiting flow stress $\tau_0$, determined by extrapolation to $N = 0$ and defined as a strain rate sensitive frictional stress, varied from 14-15 g/mm$^2$ at $\dot{\varepsilon}_1 = 0.6 \times 10^{-5}$ sec$^{-1}$ to about 20 g/mm$^2$ at $\dot{\varepsilon}_2 = 1.2 \times 10^{-4}$ sec$^{-1}$ for both stress values. The minimum flow stresses for an undeformed crystal ($N = 10^6$ cm$^{-2}$) calculated from equation (2) were 17 g/mm$^2$ and 22 g/mm$^2$ for the two strain rates above, in good agreement with the observed values.

Figure 5 shows that the increase in dislocation density $\Delta N$ as a function of shear strain $\varepsilon$ can be expressed by the relation:

$$
\Delta N = B + C \log \varepsilon
$$

(3)

where $B = 17.5 \times 10^6$ cm$^{-2}$ and $C = 3.9 \times 10^6$ cm$^{-2}$ for the single glide range $4 \times 10^{-8} < \varepsilon < 10^{-2}$. It was found that equation (3) was relatively insensitive to variation of the strain rate.

**Dislocation distribution**

In undeformed crystals, “grown-in” dislocations were chiefly arrayed in well defined boundaries outlining subgrains with an average size of 0.2-1.0 mm. New dislocations generated by application of shear stresses above the resolved yield stress resulted in the
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Fig. 5. Variation of the shear strain with the increase in dislocation density.

formation of increasingly dense arrays superimposed on the original subgranular structure as deformation progressed. Figure 6 showing a representative dislocation distribution after 0.24 per cent shear strain indicates the long range alignment parallel to the trace of the active glide plane found in copper, similar to previous results. In the initial stages of plastic flow, network formation occurred more readily in certain localized areas, producing high-density clusters which persisted to some extent throughout single glide deformation.

In addition to the primary alignment parallel to the slip plane trace, short range dislocation alignments normal to the trace were also observed after shear strains above $10^{-4}$ as shown in Fig. 7. These glide-induced alignments, previously reported by Livingston for copper, can be considered analogous to the edge dislocation arrays perpendicular to the primary

Fig. 6. Dislocation structure after $2.4 \times 10^{-3}$ shear strain with $\dot{\varepsilon} = 0.6 \times 10^{-5}$ sec$^{-1}$; $N = 7 \times 10^4$ cm$^{-2}$. 320 X
glide trace occurring in thermal polygonization. Similar mechanically polygonized arrays have been observed in LiF\(^{10}\) and zinc\(^{10}\) crystals deformed at room temperature.

**DISCUSSION**

Of basic interest is the behavior of the flow stress as a function of dislocation density. Assuming that contributions to the shear stress arise from the elastic interaction between glide dislocations and from the intersection of mobile dislocations with sessiles penetrating the glide plane (i.e. "dislocation forest" mechanism), current theory predicts:\(^{10}\)

\[
\tau = \bar{\alpha} G b N^{1/2} + \tau_s(T, \dot{\varepsilon}, N') \tag{4}
\]

where \(b\) is Burger’s vector, \(G\) is the appropriate shear modulus for the glide system, \(\bar{\alpha}\) is a constant of order 0.2 sensitive to dislocation distribution and \(\tau_s\) is a friction stress dependent on temperature \(T\), strain rate \(\dot{\varepsilon}\) and the forest density \(N'\). Comparison of the experimental results expressed in equation (2) with equation (4) requires that \(A = \bar{\alpha} G b\) and \(\tau_0 = \tau_s\). Taking \(\bar{\alpha} = 0.2\), \(b = 2.5 \times 10^{-8}\) cm and \(G = 4.4 \times 10^6\) g/mm\(^2\), a value of 2.2 g/cm was obtained in reasonable agreement with the experimental coefficient \(A = 1.7\) g/cm, \(A' = 3.2\) g/cm.

The disparity between the strain hardening coefficients for the flow stress \(\tau\) and the yield stress upon reloading \(\tau_y\) was significant, the ratio \(A'/A\) being about 1.9. The smaller hardening rate measured for \(\tau_y\) may be attributed to the occurrence of partial recovery (work-softening) after unloading the stress. Dislocations rearrangements such as the mechanical polygonization observed after deformation would act to relieve stress fields on the active glide planes, thus lowering the stress required for flow upon subsequent reloading. An estimation of the maximum attractive interaction stress between edge dislocations on parallel glide planes in a polygonized array may be obtained from the relationship \(\tau = 6G b/8\pi(1 - \nu)l\) where \(r\) is Poisson’s ratio and \(l\) is the spacing between dislocations normal to the glide planes. After a shear strain of \(2.4 \times 10^{-3}\), \(l\) was about 3 \(\mu\) as shown in Fig. 7; \(l\) was about 4 \(\mu\) with initial polygonization at \(\varepsilon = 10^{-4}\). Hence, the maximum attractive stress was about \(18\) g/cm\(^2\) at the onset of polygonization. Since dislocation motion has been observed with applied stresses well below this value, it is apparent that polygonization can occur readily at room temperature in copper.

It was evident that the friction stress \(\tau_0\), determined by extrapolation to \(N = 0\), provided the major component of the measured flow stress for the undeformed crystal in accord with previous observations on room temperature deformation of copper.\(^{11}\) Considering \(\tau_0\) to be a thermally activated stress arising from the passage of glide dislocations through a sessile forest, the activation energy controlling this mechanism can be calculated from the strain rate dependency of the flow stress:\(^{10,11}\)

\[
\tau_0 = \tau_s = \frac{U_0 - kT \ln [R(N')/\dot{\varepsilon}]}{V} \tag{5}
\]

where \(k\) is Boltzmann’s constant, \(V\) is the activation volume for the process and \(R\) is a function of the forest density \(N'\) considered to be constant during easy glide deformation. For constant temperature and varying strain rate:

\[
\frac{\tau_0(\dot{\varepsilon}_1)}{\tau_0(\dot{\varepsilon}_2)} = \Delta = \frac{U_0 - kT \ln R/\dot{\varepsilon}_1}{U_0 - kT \ln R/\dot{\varepsilon}_2} \tag{6}
\]

Hence:

\[
\frac{U_0}{kT} = \frac{\ln R/\dot{\varepsilon}_1 - \Delta \ln R/\dot{\varepsilon}_2}{1 - \Delta}. \tag{7}
\]

Assuming the rate determining activation process to be the formation of jogs by intersecting dislocation screw components, Seege\(^{11}\) has calculated that \(R\) is of the order of \(10^{-10}\) sec\(^{-1}\). Taking the experimental values \(T = 300^\circ\)K, \(\dot{\varepsilon}_1 = 0.6 \times 10^{-5}\) sec\(^{-1}\), \(\dot{\varepsilon}_2 = 1.2 \times 10^{-4}\) sec\(^{-1}\) and \(\Delta = 0.75\) for undeformed crystals, \(U_0 = 0.9\) eV in reasonable agreement with theoretical considerations of the jog energy.\(^{12}\)

The present results showing the proportionality of the strain rate independent component of the flow stress \(\tau - \tau_0\) to \(N^{1/2}\) are generally consistent with data recently reported by Livings\(^{10}\)ton\(^{16}\) for copper. However, Young\(^{10}\) has reported that the macroscopic yield stress of copper, corresponding to a strain of about \(10^{-4}\), was largely independent of strain rate and dislocation density for crystals with initially low density values. The difference in results may be attributed to
differing definitions of the yield stress, here defined as the minimum flow stress for an initial plastic strain increment of $5 \times 10^{-6}$. The $5$- to 20-fold increase in dislocation density found by Young with “pre-yield” deformation ($\varepsilon < 10^{-4}$) is in good agreement with the 20-fold density increase ($1 \times 10^5$ to $2 \times 10^6 \text{ cm}^{-2}$) shown in Fig. 5 after $10^{-4}$ plastic strain. As noted before, the present yield stress corresponds more closely to Young’s “multiplication stress” above which measurable dislocation generation occurred. The present dislocation density estimates are considerably lower than previous values obtained from X-ray line broadening measurements,\(^{(14)}\) partly attributable to an initially higher dislocation concentration in the latter case. Undoubtedly, errors in the X-ray measurements due to instrumental broadening tend to underestimate the dislocation density, whereas, etch pit counting usually tends to underestimate the density. However, density values obtained by both techniques showed similar exponential increases with initial plastic strain during easy glide deformation.

**SUMMARY**

Measurements of the applied stress and yield stress of copper crystals were made during easy glide tensile deformation as a function of strain rate and dislocation density as determined by an etch pitting technique. The results showed that the stresses could be interpreted by current theory as composed of two contributions; a glide dislocation interaction stress proportional to the square root of the dislocation density, and a thermally activated friction stress arising from the cutting of glide dislocations through a forest. The latter component provides about 75 per cent of the observed yield stress for undeformed crystals. The activation energy for the dislocation forest mechanism was 0.9 eV.

The hardening coefficient of the applied stress was about twice that for the yield stress, indicating that partial recovery occurred after unloading the stress. Recovery of the mechanical properties was accompanied by stress-induced polygonization observed after deformation.

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**REFERENCES**