The Effect of Stress on Internal Friction in Polycrystalline Copper

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The internal friction of polycrystalline specimens of oxygen-free copper, subjected to compressive stress, increases and then decreases with continuously increasing stress. A complementary behavior of Young's modulus is found. Thus, while the internal friction increases 100 percent, Young's modulus decreases about 6 percent for a stress of 90 kg/cm^2. An increase of stress to 160 kg/cm^2 leaves a remnant increase of only 40 percent in the internal friction and a remnant decrease in Young's modulus of 4 percent. This effect is nearly independent of the temperature at which the stress is applied. Although the Taylor dislocation of plasticity is capable of explaining qualitatively the data obtained at any one temperature, no simple explanation of the temperature independence of the effect is available at the present time.

RECENT theoretical studies of phenomena associated with slip in single crystals have been based principally on a model proposed independently by Taylor,[1] and by Orowan[2] who suggested that the various irreversible processes occurring in stressed crystals originate in microscopic lattice irregularities which are termed dislocations. Numerous proposals[3] have been made concerning the nature and behavior of these dislocations, whose movement through the lattice is believed to be responsible for creep and strain hardening.

Recently, Read[4] reported some measurements of internal friction in single crystals. His results indicate that the irreversible work performed during the vibration of a single crystal is generated by the mechanism responsible for plastic flow. This relation is immediately suggested by the fact that the decrement of the vibrations is inversely proportional to the frequency of vibration. Further, Read finds that an increase in the amplitude of vibration augments the internal friction. He suggests that the explanation of the phenomena is as follows. The increase in amplitude of vibration generates dislocations whose motion contributes to the internal friction. The increase of internal friction accompanying application of an external stress supports this contention. The internal friction returns to a lower value after annealing, and, in fact, the more careful the anneal, the lower the internal friction. If it be assumed that dislocations can diffuse out of the lattice during the annealing process, it is reasonable to conclude that all internal friction in single crystals arises from the presence of dislocations. This statement must be qualified, of course, if the specimens are ferromagnetic or if the geometry is such that the vibration is neither adiabatic nor isothermal.

The object of this paper is to report some measurements on the increase in internal friction at different temperatures in polycrystalline copper due to the application of various external compressions. The range of stress here studied is about four times that covered by Read.[5] Such data should yield information concerning the ease with which dislocations may be generated at various temperatures.

SPECIMENS

The study of polycrystalline specimens has the disadvantage that a certain amount of internal friction must arise from the irreversible flow of heat currents between variously oriented grains to neutralize the temperature gradients established by thermoelastic effects. Zener[6] has treated this type of internal friction both experimentally and theoretically and shown that the contribution of such heat currents to the internal friction is small if the time for the heat wave to flow across a grain is either very short (isothermal

[1] G. I. Taylor, Proc. Roy. Soc. **A145**, 362, 388, 405 (1934).
[2] E. Orowan, Zeits. f. Physik **89**, 614, 634 (1934).
[3] For a comprehensive review of plasticity and correlated subjects, the reader should consult F. Seitz and T. A. Read, J. App. Phys. **12**, 100 (1941).
[4] T. A. Read, Phys. Rev. **58**, 371 (1940).
[5] T. A. Read, Phys. Rev. **59**, 477A (1941).
[6] C. Zener, Phys. Rev. **52**, 230 (1937).

extreme) or very long (adiabatic extreme) compared to the period of vibration. The grain size of the specimens herein described is estimated visually after etching the copper with a concentrated solution of nitric and acetic acids. The linear dimensions of the grains are of the order of magnitude of several millimeters, and the contribution to the internal friction due to their random orientation is sufficiently small to permit a determination of internal friction due to the generation of dislocations by an external load.

The advantage of using polycrystalline specimens in this work is afforded by the availability of extremely pure polycrystalline samples of copper. This is important since it is well known that strain hardening is very structure sensitive, particularly with regard to the addition of small amounts of a foreign substance. The materials used in this work were prepared in the research laboratories of the American Smelting and Refining Company, in Barber, New Jersey, under the supervision of Dr. A. J. Phillips. The most refined chemical and spectroscopic analyses fail to reveal any impurities. In most cases, this indicates that the impurity in question is less than 0.0001 percent, and, though it is impossible to prove the oxygen content is less than 0.001 percent, other evidence indicates the oxygen content is also less than 0.0001 percent. In view of the extreme purity of the samples, it was decided to work on polycrystalline samples despite the disadvantage noted above, since it was felt that remelting would undoubtedly introduce impurities. The specimens therefore were machined directly from the supplied stock which consisted of a rolled rod $\frac{3}{8}''$ in diameter. These specimens are right cylinders approximately 3.2 cm in length and 0.476 cm in diameter.

Method

Internal friction is most conveniently described in terms of the decrement of the vibrations, which is defined as the ratio of the energy dissipated per cycle of vibration to twice the vibrational energy of the specimen. This quantity may be ascertained by observing the variation with frequency of the impedance of a composite piezoelectric oscillator constructed by cementing a rod of cylinder material to a suitably cut quartz crystal. Since the electric network equivalent to the piezoelectric oscillator is a capacitance C in parallel with a series circuit comprising an inductance L, capacitance K, and resistance R, the admittance A at a frequency $\Delta\omega$ from the resonant frequency may be written

$$A=\left\{\frac{1-4LC\omega\Delta\omega}{R^2+4L^2(\Delta\omega)^2}+\omega^2C^2\right\}^{\frac{1}{2}}, \tag{1}$$

where ω is the angular frequency. An analysis[7] of the admittance as a function of frequency reveals that, at constant voltage, the resulting current possesses a maximum and minimum value and frequencies near resonance. If one denotes the maximum and minimum observed currents by I_1 and I_2, respectively, and the frequencies at which they occur by f_1 and f_2, respectively, the resonant frequency f_0 is calculated from the relation:

$$f_0=f_1+(f_2-f_1)\frac{I_2}{I_1+I_2}. \tag{2}$$

The decrement of the composite oscillator δ is given in terms of the foregoing quantities by:

$$\delta=2\pi\{(f_2-f_0)(f_0-f_1)\}^{\frac{1}{2}}/f_0. \tag{3}$$

The decrement of the specimen Δ_1 may be calculated from a knowledge of δ and the decrement of the quartz crystal alone Δ_2, determined in the same manner as δ, with the aid of the relation:

$$\delta=\frac{m_1\Delta_1+m_2\Delta_2}{m_1+m_2}, \tag{4}$$

where m_1 and m_2 are the masses of the specimen and the quartz crystal, respectively.

When the resonance curve is very narrow, a precise determination of the quantity (f_2-f_0) appearing in Eq. (3) is difficult and the errors in δ are large. The value of δ is then best determined by an alternating-current bridge after the manner of Cooke and Brown.[8] When the resonance curve is broad, the precision of δ suffers due to the difficulty of locating f_1 and f_2 accurately. Fortunately, however, in this experiment we are interested in the *increase* in internal friction upon the application of an external stress. In other

[7] S. Siegel and S. L. Quimby, Phys. Rev. **49**, 663 (1936).
[8] W. T. Cooke and W. F. Brown, Phys. Rev. **50**, 1158 (1936).

words, relative values of internal friction are of primary interest. These may be determined by the simple method described below for all values of δ to a precision of about 2 percent.

If, in Eq. (1), we regard ω^2C^2 as constant over the width of a resonance curve, we find simply:

$$A_{\max}-A_{\min}=1/R, \tag{5}$$

where R, the resistance of the oscillator, is related to the decrement by the relation:[9]

$$R=\tfrac{1}{4}K(m_1+m_2)\delta f_0. \tag{6}$$

If the amplitude of the voltage applied to the quartz crystal is held constant, we obtain immediately from Eqs. (4), (5), and (6) the relations:

$$\frac{\delta'}{\delta''}=\frac{I_1''-I_2''}{I_1'-I_2'}, \tag{7}$$

$$\frac{\Delta_1'}{\Delta_1''}=\frac{I_1''-I_2''}{I_1'-I_2'}\left\{\frac{(i_1-i_2)-(I_1'-I_2')}{(i_1-i_2)-(I_1''-I_2'')}\right\}, \tag{8}$$

where I_1 and I_2 are the maximum and minimum currents, respectively, drawn by the composite oscillator, and i_1 and i_2 are the maximum and minimum values of the current drawn by the quartz crystal alone under the action of the applied voltage. The prime and double prime quantities refer to values observed in two different measurements on the same specimen. It is to be noted that the quantity in the brackets in Eq. (8) is, in general, a small correction term, since the contribution of the quartz crystal to the decrement of the composite oscillator is usually ten to a thousand times smaller than the contribution of the specimen.

The above theory is only applicable, of course, if the resistance of the device employed to measure the current through the quartz crystal is negligible compared to the resistance of the quartz. There seems to be no way of predicting from the geometry and physical constants of the quartz crystal the resistance of the oscillator. In general, the resistances of quartz crystals similar to those used in this work lie between several hundred and several thousand ohms. The resistance of the composite oscillators is a factor of ten to a thousand larger, since the resistance is proportional to the decrement. In order to measure the currents in these experiments, a resistance of 17 ohms is inserted between the crystal and ground, and the voltage developed across this resistor is amplified and measured. In order to ascertain the effect of the resistance on the accuracy of the measurements, the same experiments were repeated with a 50-ohm resistor. No systematic discrepancies between the two sets of data appeared. The alternating voltage applied to the quartz crystal is generated by a transitron oscillator which is decoupled from the crystal by means of two buffer stages of amplification. From a knowledge of the resonant frequency of free longitudinal vibration of the quartz crystal alone, and that of the composite oscillator, the frequency of free vibration of the specimen alone may be computed.[7] Twice the product of this quantity and the length of the specimen yields the velocity of longitudinal waves in the specimen material.

It may further be noted that, provided the crystal and specimens have resonant frequencies differing by less than ten percent, the effect of the adhesive is not discernible, nor is it necessary to have the areas of the crystal and the specimen congruent.

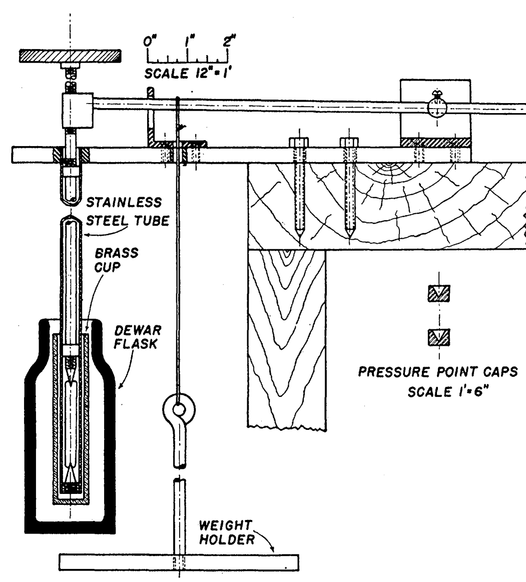

FIG. 1. The apparatus used for subjecting specimen cylinders to compressive stresses at various temperatures.

[9] In Eq. (6), $K=1/b^2\epsilon^2$, where ϵ is the appropriate piezoelectric constant for quartz, and b is the width of the metal electrodes plated on the quartz.

Procedure

The specimens are initially annealed by heating them in hydrogen at atmospheric pressure at a rate of approximately 100°C an hour to 600°C where their temperature is kept constant for several hours. The specimens are then cooled to room temperature over a period of ten hours.

The specimens are cemented to a quartz crystal whose resonant frequency for longitudinal vibrations is about 50 kc, by a thin film of beeswax and rosin under a compressive force of 0.2 kg at a temperature of 60°C. This procedure does not seem to affect the specimens appreciably, inasmuch as reproducible results are obtained after successive adhesions. A further justification of this technique will be discussed below.

The specimens are detached from the quartz crystal and subjected to compressive stresses in the device shown in Fig. 1. This device is designed in a manner calculated to facilitate the application of stresses over a wide range of temperatures. Thus, the specimen is supported at the bottom of a stainless steel tube on a pressure point cap which rests on a pointed screw. Slots cut in the side of the tube permit the insertion of the specimen. The load is transmitted from a mechanical lever system to a similar pressure point cap on the top of the specimen by a rigid steel rod which is threaded so that its length may be adjusted to fit with that of the specimen. Initially, the lever, from which the weights are hung, rests on a knife edge which bears the load until the steel rod is so adjusted in length that it just touches the cap on the top of the specimen. When it is desired to apply the load to the specimen it is merely necessary to advance the screw slightly. The angle of the cone of indenture in the pressure point caps is slightly greater than the angle of the conical points applying the compression. This fact, coupled with the fact that the ends of the specimen are ground accurately perpendicular to the cylinder axis, ensures that the surface traction will be normal to and uniform over the ends of the specimen.

The temperature of the specimen during loading is controlled by immersing the end of the stainless steel tube in a suitable temperature bath. In order to control the approximate rate of cooling or heating of the specimen when it is introduced to the bath, a brass cup to surround the end of the tube is provided. In practice, the Dewar flask containing the cryogenic fluid is raised around the cup at such a rate that the thermoelastic forces experienced by the specimen are negligibly small. When the temperature of the specimen has approached sufficiently that of the bath, the flask is raised further until the fluid spills over into the cup and actually comes into direct thermal contact with the specimen. This ensures that the specimen finally attains the temperature of the bath.

After the application of the load, the specimens are allowed to return slowly to room temperature and are removed from the press and reaffixed to the quartz crystal for the purpose of determining the increase in internal friction due to the application of the load. The results obtained seem to be independent of the time of duration of the stress for times greater than one second and less than one hour. No experiments were performed in which the duration of the stress was greater than one hour, and, in general, the duration of the stress was not greater than one minute. Since the recrystallization temperature of copper is about 200°C, the effect of any stress applied below this temperature is probably permanent. The increase in internal friction due to the stress may, therefore, be measured at leisure.

All the results reported in this paper were obtained from measurements on nine specimens. After the curve of increase in internal friction as a function of stress is obtained at one temperature, the specimens are subjected to another cycle of annealing and stressing to obtain similar data at another temperature.

Results

The base value of the decrement of the annealed specimens is approximately 2×10^{-3}. The increase of the decrement over the base value as a function of applied compressive stress is shown in Fig. 2, where the results obtained at 80°K are compared with those obtained at 298°K. The effect of decreasing the temperature is to increase the maximum in the internal friction and to displace it to somewhat higher stress values. In order to ascertain that the larger increase in internal friction observed at liquid-nitrogen temperatures was not due to the existence of thermoelastic stresses, an annealed specimen was quenched directly in liquid nitrogen, removed and warmed to room temperature as rapidly as possible. This drastic treatment produced an increase of only 23 percent in the internal friction; a reasonable rate of cooling such as described above reduced the increase to 2 percent which is within the experimental error.

The average ratio of internal friction after applying a stress of 76.5 kg/cm² to the base value on nine annealed specimens was found to be at

80°K	2.06±0.08,	298°K	1.98±0.08,
193°K	2.02±0.06,	423°K	2.00±0.05.

An attempt to obtain data at temperatures above 200°C was unsuccessful. The data were quite irreproducible due, presumably, to the unpredictable effects of traversing the region of recrystallization. The fact that the data obtained at 150°C agree with those obtained at lower temperatures, however, tends to justify the procedure employed of affixing the specimen to the quartz at a temperature of 60°C.

The average velocity of longitudinal sound waves in the well-annealed specimens at 25°C is found to be 3.23×10^5 cm/sec. This value is considerably lower than that reported by Quimby,[10] who found the velocity of sound to be 3.65×10^5 cm/sec. This discrepancy might conceivably arise from two sources: first, the presence of impurities, although it is extremely doubtful that such a large difference can be explained in this manner; or, secondly, an anisotropic orientation of the crystallites. Annealing the specimens in oxygen produced no appreciable effect on the velocity of sound. In order to investigate more fully the second possibility, a specimen was severely cold-worked by hammering a cylinder into a parallelepiped. After annealing, the velocity of sound in this specimen evinced a normal value, namely 3.67×10^5 cm/sec. As a result of this test, it is concluded that the anomalously low values found for the velocity of sound in specimens employed in these experiments are due to a preferred orientation of the crystallites, arising, presumably, from a rolling or extruding operation used in their preparation.

[10] S. L. Quimby, Phys. Rev. 4, 558 (1925).

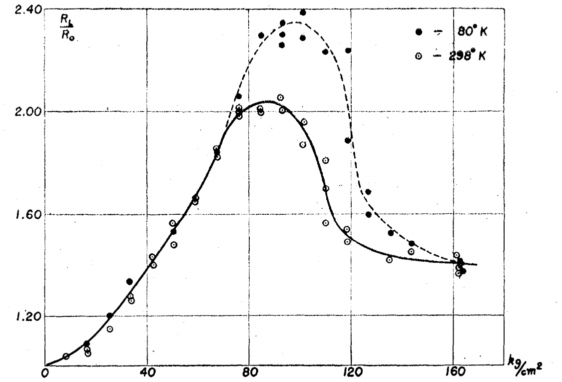

FIG. 2. The relative increase in internal friction as a function of compressive stress. The solid circles refer to data taken on specimens subjected to compression at 80°K. The open circles refer to data taken on specimens subjected to compression at 298°K.

The behavior of Young's modulus as a function of the applied stress is complementary to the behavior of the internal friction. Thus in several specimens investigated, Young's modulus decreases with increasing load until at the stresses which produced the maximum increase in internal friction, the value of Young's modulus reaches a minimum from 6 percent to 7 percent lower than the initial value. Application of still larger stresses then causes an increase in Young's modulus until after a stress of 162 kg cm² the value of the modulus is only from 4 percent to 5 percent lower than the initial value in well-annealed specimens.

The effect of oxygen impurity on the generation of dislocations was investigated in the following manner. The specimens were annealed in an atmosphere of air and then etched until a clean copper surface was exposed. The behavior of the internal friction in specimens prepared in this manner as a function of applied compressive stress differs from the behavior of specimens annealed in hydrogen in only one particular. The former specimens evinced a somewhat higher base value of internal friction in the annealed state and the maximum increase in internal friction was somewhat less than that observed in the hydrogen annealed specimens, and furthermore varied from specimen to specimen. The average maximum increase in internal friction in the oxygen annealed specimens was about 60 percent. The decrease in internal friction observed at high stresses was the same in all specimens and equal to that observed in the specimens annealed in hydrogen.

Discussion

Errors in this experiment arise principally from two sources. The first of these is due to the effect of poor adhesion between the specimen and the quartz. Poor adhesion results in an anomalously high value of the decrement. The second source of error is mishandling of the specimen. Thus, when a specimen is dropped a quarter of an inch onto a hard surface, there is an increase in internal friction equivalent to the maximum increase in internal friction produced by the application of external static forces. Errors due to the first source may be corrected by reaffixing the specimen to the quartz crystal in any case in which the value observed for the decrement is suspiciously high. The only precaution which can be adopted to avoid errors from the second source is extreme care in the handling of the specimens to prevent any sudden jarring in their manipulation.

The initial increase in internal friction as a function of applied stress may be interpreted as due to the formation of dislocations which dissipate energy by virtue of irreversible work performed in moving them. The decrease in internal friction at higher stresses may be understood as due to the formation of a more stable array of dislocations. The term "locking" is introduced to describe this condition. Until a more concrete model of locking is proposed, no simple explanation of why more dislocations can be introduced into the lattice at low temperatures and why a higher stress is needed there to produce locking is justifiable until more data on creep at low temperatures is available.

It might be appropriate to mention at this juncture two further experiments, more or less qualitative in nature. A number of rocksalt crystals which had been standing in a desiccator over phosphorus pentoxide for over a year were subjected to a compressive stress of 25 kg/cm^2. The average observed increase in internal friction was about 10 percent. If the specimens were dipped in water for about 10 seconds just prior to or during the application of the stress, the average observed increase in internal friction was about 70 percent. The effect of a strong etch on the polycrystalline specimens of copper, however, was not detectable. This is not surprising in virtue of the fact that only a small part of the total crystal surfaces was exposed to the action of the etch. It is planned to repeat this experiment on single crystals of copper in the future.

In order to investigate the effect of the duration of the forces on the increase in internal friction for very small durations, the following crude experiment was performed. The specimens were rolled from a table top onto a cast iron plate whose height below the table top could be adjusted at various distances. It was found that the maximum increase in internal friction always occurred for distances of less than one inch but that the specimens had to fall about thirty inches before the decrease observed in the internal friction was comparable with that found in the experiments above. Inasmuch as the area of contact when the specimen struck the plate might vary considerably from experiment to experiment, no quantitative conclusions may be drawn. However, these results do suggest that while the formation of dislocations under the action of applied stress is practically instantaneous, duration as well as magnitude of load plays a role in the production of locked dislocations. Since, in some of the specimens which were dropped, increases in internal friction as much as fifty percent greater than any increase under static loads were observed, these experiments suggest that the higher maximum in internal friction and the shift of this maximum to higher stresses at low temperatures are due to a less rapid formation of locked dislocations at low temperatures than at high temperatures for a given stress. More experiments on the effect of rate of loading are being undertaken in an effort to settle this point.

In conclusion, the author desires to express his appreciation to the participants in the Works Progress Administration Project O.P. 65–1–23–2884 for much routine assistance, and to Professor Frederick Seitz and his colleagues Dr. Hillard Huntington and Dr. James Koehler for several stimulating discussions concerning the theoretical aspects of the problem.