Anomalous Ultrasonic Velocity Changes in Deformation and Irradiation Induced Dislocation Pinning

R. TRUELL AND A. GRANATO*

Metals Research Laboratory, Brown University, Providence, Rhode Island, U.S.A.

The effect of dislocation pinning is usually that of reducing the damping and increasing the velocity (or modulus) for ultrasonic waves. However, dislocation theory predicts that at high enough frequencies (or at large loop lengths or high phonon densities), an anomalous effect can occur. That is, the ultrasonic velocity can decrease with pinning. This effect is discussed and examples are shown for slightly deformed pure aluminum and sodium chloride single crystals.

Introduction

Dislocation theory predicts an unusual velocity behavior for ultrasonic velocity changes at high frequencies. Normally, the effect of pinning dislocations in crystals is to increase the velocity of ultrasonic waves (or elastic modulus) in the crystal. Pinning reduces the motion of the dislocations, and therefore the anelastic strain, so that only the purely elastic strain is measured in crystals with completely pinned down dislocation. However, it is possible, under conditions to be discussed here, for the velocity or modulus to decrease with pinning of disloca-The latter effect will be termed tions. anomalous. It is important both because an experimental check of this unusual prediction can be regarded as one of the strongest checks available to the theory, and because of the possible application of this effect to the study of deformation and irradiation processes.

The results of the relevant theory have already been used by Hikata, Chick, Elbaum and Truell¹⁾ to account for the anomalous velocity change observed during deformation of aluminum single crystals. Since that time the physical nature of the cause of the effect has become clear, and a detailed and extensive examination has been made of the rather special conditions required to obtain the anomalous effect. At the same time a more complete mathematical description of the expected pinning behavior has been made including velocity calculations having quantitative velocity at high frequencies. A comparison of this theory is made with the experimental results available to date.

Physical Nature of the Anomalous Effect

The origin of the effect of dislocations on the elastic modulus and velocity may be understood simply with the help of Fig. 1. In Fig. 1A, the straight line segment along the x-axis between x=0 and x=l represents a dislocation loop of length l which is straight when no stress is applied. When a stress is



Fig. 1 Displacement of a dislocation under stress (Schematic)

- a. Curve (1): Dislocation displacement amplitude at low damping (small phonon density) Curve (2): Dislocation displacement amplitude for high damping
- b. Out of phase and in phase component of dislocation displacement for high damping
- c. Change in Fig. 1B when pinning point is added at center of dislocation loop (Pinning increases area of in phase component thereby decreasing elastic modulus and increasing velocity of ultrasonic wave).

^{*} Permanent Address: Physics Department, University of Illinois, Urbana, Illinois, U.S.A.

applied, the dislocation loop bows out, but it is held back by the pinning points at the ends, as in curve (1) of Fig. 1A. If the stress is sinusoidal, with frequency v, the displacement y(x) is sinusoidal, with curve (1) representing the amplitude of the dislocation However, if the medium in displacement. which the dislocation segment vibrates is viscous, there will be a drag on the dislocation tending to oppose the motion. In real crystals, the viscosity is thought to be caused by the phonon gas through which the dislocation is being driven.²⁾

At low frequencies, the velocity of the dislocation is small, and the viscous drag may be neglected. In this case, the motion of the dislocation is limited only by the tension of the dislocation line, and a curve such as (1)of Fig. 1A is obtained. At high frequencies, the velocity of points of the dislocation line remote from the pinning points is large, so that the viscous force is more important than the tension force. The amplitude of motion then becomes viscosity-limited, and is less than that obtained for low frequencies. Now, the dislocation segment oscillates over most of its length as a rigid rod, except at the end points, where the displacement is zero. The same transition in behavior from curve (1) to curve (2) occurs if the ultrasonic or driving frequency is held fixed and the loop length is increased, or if the frequency and loop length are held fixed and the viscosity is increased (for example, by increasing the temperature and therefore increasing the density of lattice vibration phonons).

Curve (2), however, represents the envelope of the amplitude of the motion. At any given instant therefore the shape of the dislocation displacement will not appear as in curve (2). The reason for this is that the phase of all points along the line is not the same. Those points near the pinning points have a small maximum displacement and therefore a small velocity so that the viscous drag is small, and they are always in phase with the applied stress. On the other hand, points near the middle of the segment are approximately 90 degrees in phase behind the stress for large viscous drags. As a result, the parts of the motion represented by curve (2) which are out of phase and in phase with the applied stress are as shown in Fig. 1B. If the loop

length l is long, then the motion is out of phase (*i.e.* lags behind by nearly 90 degrees) over most of the loop length. The curve shown in Fig. 1B also represents schematically the shape of the dislocation segment at two times differing by one quarter of the period of the motion.

In ultrasonic measurements, the out of phase and the in phase parts of the motion are measured separately. The out of phase motion leads to attenuation and the in phase motion leads to velocity changes of ultrasonic waves. The area under the curve of the in phase part of the motion is proportional to the anelastic strain contributed by the dislocation. The dislocation contribution to the strain is given by

$$\varepsilon_{\rm dis} = \frac{1}{l} \int_0^l by'(x) dx$$

where y'(x) is the real part of y(x), b is the Burgers vector, and ε_{dis} can thus be thought of simply as the area under the real part of the dislocation displacement. The modulus change of the specimen is given by

$$(M_0 - M(\omega))/M_0 = \varepsilon_{\rm dis}/\varepsilon_{\rm el},$$

where $M(\omega)$ is the modulus measured at frequency ω , and M_0 is the modulus measured at infinite frequency. $\varepsilon_{\rm dis}$ is the dislocation contribution to the total strain and $\varepsilon_{\rm el}$ is the elastic strain. Since the velocity of an ultrasonic wave is given by

$$v = \sqrt{\frac{M}{
ho}}; \; rac{\Delta v}{v_0} = rac{v_0 - v(\omega)}{v_0} = rac{1}{2} \; rac{\Delta M}{M_0},$$

where ρ is the density of the solid. This then represents the velocity or modulus decrease of an ultrasonic wave in a crystal containing dislocations. As the frequency is increased, this area decreases and the ultrasonic velocity therefore increases, as the dislocation is no longer able to follow the rapidly varying external stress. Such a dispersion has been observed in NaCl between 10 and 100 megacycles/sec by Granato, deKlerk and Truell.³⁾

Experimentally it is usually easier to vary the average loop length or resonant frequency ν_0 than it is to vary the measurement frequency ν , as for example in irradiation or deformation and recovery experiments. If instead of varying the stress wave frequency with fixed loop length, one varies the loop length with fixed frequency, the anomalous

velocity effect appears. Fig. 1C shows how the displacements of Fig. 1B change when a pinning point is added. At low frequencies, when a pinning point is added, the area of the in phase component of the displacement is decreased, and so the ultrasonic velocity increases. i.e. $\Delta v/v_0$ decreases and $v(\omega)$ increases. However, at high frequencies, addition of a pinning point forces the part of the dislocation near the newly added pinning point to be in phase with the applied stress so that the in phase component of displacement pops out, increasing the area, and decreasing the ultrasonic velocity. It is clear from Fig. 1C that the effect is in general a small one, since at high frequencies, only a small part of the displacement is in phase with the external stress. If more pinning points are added, the in phase component eventually becomes pinned out completely. As this limit is approached the pinning points get close enough together to interact, giving the normal velocity effect because the more pinning point regions that are added the more tension limited segments are added until a limit is reached. Also, from Fig. 1C, one sees that adding pinning points to very long dislocation loops has little effect on the attenuation when the motion over most of the length of the loop is viscous drag limited. Eventually, when the average loop length is made small enough, the attenuation decreases with pinning.

In this way it is seen that for long enough loop lengths (or large enough frequencies or large phonon densities or temperatures), velocity or modulus measurements are sensitive to conditions near the pinning points while attenuation (or decrement) measurements are sensitive to the viscous drag, or specific damping caused by dislocation-phonon scattering. Under the specific conditions just mentioned, velocity measurements are especially appropriate for investigating dislocationpinning point interactions while attenuation measurements are more relevant for dislocation-phonon interactions.

Velocity and Attenuation Relations

A quantitative theory for the attenuation and velocity of ultrasonic waves in crystals containing dislocations has been given by Granato and Lücke.⁴⁾ The model is based upon the analogy between the motion of a dislocation loop and that of a vibrating string proposed by Koehler.⁵⁾ The result are given in the form of infinite series. The first term of the series gives a sufficiently accurate description of the attenuation. Because of the simplicity of this attenuation result, the effect of a distribution of loop lengths on the result is also not difficult to obtain. For velocity, the first term of the series is alsosufficiently accurate for frequencies up to about that for which the inflection point appears in the dispersion curve. As mentioned at the end of Section I for higher frequencies. (where the velocity reduction $\Delta v/v_0$ is small) the first term of the series is not quantitatively accurate, so that it is more difficult to take into account the effect of the distribution.* However, calculations show that the first term of the series still gives all the qualitative results, including the anomalous velocity effect. The effect of a distribution of loop lengths has not been considered. Simple analytical results, giving explicitly the effects. of the various variables, will be used for the present discussion.

For the purpose of studying the effects of pinning or unpinning of dislocations it is convenient to express the changes in attenuation and velocity in terms of loop length or resonant frequency ω_0 . We have chosen to use the variable $y=\omega_0/\omega$ (see reference 1) where ω_0 is the resonant frequency of the dislocation loops and ω is the frequency at which measurements are taken. As the loop length is decreased by pinning, y increases since the resonant frequency increases. Expressed in this way, the attenuation becomes

$$\alpha(y) = K \frac{d}{\omega^2} \left\{ \frac{1}{(y^2 - 1)^2 + (d/\omega)^2} \right\} \quad (1)$$

and the velocity change is given by

$$\frac{dv}{v_0} = \left(\frac{v_0 - v(\omega)}{v_0}\right) = H \frac{1}{\omega^2} \left\{\frac{y^2 - 1}{(y^2 - 1)^2 + (d/\omega)^2}\right\} (2),$$
$$d = B/A$$

where

* For the case of very low frequencies an approximation has been used in which the inertia of a dislocation has been neglected to obtain numerical results for the normalized decrement and modulus change as a function of normalized frequency.⁶⁾

$$K = \left[\Omega \, 8.68 \times 10^{-6} \left(\frac{4G \, b^2}{\pi^2} \right) \frac{\Lambda}{A} \right]$$
$$H = \left[\Omega \left(\frac{4G \, b^2}{\pi^2} \right) \frac{\Lambda}{A} \right]$$

G is the elastic shear modulus, Ω is the orientation factor, b is the Burgers vector, and Λ is the dislocation density. The factor 8.68×10^{-6} is needed to give the attenuation in the commonly used units of decibels per

microsecond.

In the above, the parameter d is a measure of the damping. These curves for various frequencies of interest are plotted in Fig. 2 for an assumed value of the damping constant d of 7.4×10^9 sec⁻¹.

For a given measurement frequency, as the resonant frequency of the dislocation loops is increased from y=1, the attenuation decreases





from a maximum value, while the velocity at first decreases (anomalous effect), passes through a minimum, and then increases The velocity curve has a (normal effect). minimum at $y_m^2 = 1 + d/\omega$, at which point the attenuation is half the maximum value. This is also the point at which the decrement is a maximum. One can plot, on the velocity curves of Fig. 2, values of y for an assumed initial resonant frequency. For example, if $\nu_0 = 300 \text{ mc/sec}$ is assumed, the velocity curve for $\nu = 30 \text{ mc/sec}$ would have y = 10 as the value of y at the start of irradiation. By drawing a plot of the locus of all points for each assumed resonant frequency, one finds a set of curves such as those shown in Fig. 2 for 150, 200, and 300 mc/sec.

The intersections of the second set of curves, shown on the velocity diagram with the curves just described, show the values of the velocity at each frequency for a given resonant frequency of the dislocation loops. For example, if the resonant frequency of the dislocation loops is $\nu_0=300$ mc/sec, then for all frequencies below about 20 mc/sec, the velocity is the same (*i.e.* independent of frequency), while for larger frequencies, the velocity is higher. This describes the normal dispersion effect.

If, however, measurements are made at constant frequency with varying loop length or varying ω_0 , the curves show under what conditions the anomalous velocity effect occurs. For example, suppose the resonant frequency of the dislocation loops is 200 mc/sec to start If now the resonant frequency is with. increased to 300 mc/sec by some pinning mechanism, then the velocity increases at 10 and 20 mc/sec, giving the normal effect. At 30 mc/sec, the velocity at first starts out with zero slope and then increases. That is, there is initially no change in velocity in spite of the fact that the attenuation is decreasing rapidly as a function of ω_0 at this frequency. For higher frequencies, the velocity at first decreases with pinning before increasing. It is easy to see that the magnitude of the velocity reversal must be a small one. Suppose, for example, that the velocity change measured at low frequencies is 1% (this corresponds to a 2% change in modulus, and larger changes are seldom seen at megacycle frequencies). Then from Fig. 2, it is evident that the

magnitude of the velocity reversal effect cannot be expected to be greater than about 0.1%. The exact size depends upon how far to the left of the minimum the intersection of curves in Fig. 2 occurs. The effect of the higher order terms in the series expression for the velocity as well as the effect of a distribution of loop lengths, will be to broaden out slightly the minimum and to cause these curves to intersect the abscissa below y=1. This does not change anything qualitatively but has the effect of reducing somewhat the magnitude of the anomalous velocity effect to be expected.

Experimental Observations

From the above discussion necessary conditions for observing the anomalous velocity effect can be established. One needs the resonant frequency to be low initially (i.e. large loop lengths) and velocity measurements sensitive to at least one part in 10⁴. It might be thought that specimens of sufficiently high purity would have long enough loop lengths. Attempts to find this effect in irradiated, undeformed, high purity aluminum and sodium chloride have been unsuccessful. However, the effect has been found in deformed crystals. Evidently light deformation produces loop lengths of sufficient size. Deformation and irradiation (or recovery after deformation) can be expected to give similar but oppositely directed changes. Irradiation or recovery should produce pinning (i. e. increasing y). On the other hand, deformation should produce increased loop lengths for very small deformation (decreasing y), followed by decreasing size of average loop length after large deformation. Although irradiation or recovery experiments should be simpler physically because the dislocation density does not change at the same time that the average loop length does, it seems at present to be necessary to use deformation to produce suitable conditions.

In this section we discuss first the results observed during deformation, and then the simpler case of the changes which occur on recovery.

In Fig. 3, results by Hikata, Chick, Elbaum and Truell,¹⁾ for the velocity, attenuation, and stress as a function of strain are shown for an aluminum single crystal. The direction of polarization of the ultrasonic shear was



Fig. 3. Stress, shear wave attenuation and velocity change as a function of total strain in aluminum. Hikata *et al.*

such that dislocations in the primary glide system were not seen by the ultrasonic wave. As a function of deformation the velocity at first increases a small amount (a few parts in 10^4) before decreasing. Fig. 4 shows this velocity reversal effect for three principal



Fig. 4. Longitudinal wave velocity change $\Delta v/v$ for <100>, <111> and <110> orientation as a function of shear strain. Hikata *et al.*

directions in these specimens. Normally at low frequencies it would be expected that the velocity should decrease with deformation since the dislocations produce an anelastic strain. However, this behavior and magnitude of effect is just what should be expected if small deformation produces large dislocation loop lengths.

At high strains, where the dislocation density becomes large, the normal reduction of velocity with deformation is found because the lengths are now much shorter. This small velocity reversal is usually found in high purity aluminum. It should be noted that $\Delta v/v_0 = \{v_0 - v(\omega)\}/v_0$ does not change sign: the quantity $\Delta v/v = \{v_i - v(\omega)\}/v_i$ plotted in Fig. 3 refers not to v_0 , the purely elastic velocity, but to v_i measured before deformation. The value of v_0 , the purely elastic velocity, for a perfect aluminum crystal is not known. Presumably the value of v_0 lies somewhere above any point of the curve. The observed effect is consistent with the expected anomalous effect. No other explanation of the observed behavior has been found.

It might be expected that, if the deformation were stopped in a strain interval where the velocity reversal was taking place, the pinning which would occur during recovery and as a function of recovery time should show the anomalous effect more definitely. This is difficult to arrange at room temperature in aluminum because recovery occurs so quickly. Presumably, recovery occurs as a result of pinning of dislocations by the deformation induced point defects.⁷⁾ However, in NaCl, recovery *is* slower at room temperature, and an example of work in progress by Hikata *et al.*, is shown in Fig. 5.

Here the results of the recovery, under no load, of velocity and attenuation in NaCl after a small deformation are shown. As in the case of aluminum, there is a velocity reversal during deformation. This is not shown here. During recovery, the attenuation decreases from the very beginning, showing that pinning is occurring, but the velocity does not change for at least several minutes. After a time delay, the velocity increases in the normal way. The total velocity change is small (several parts in 10^4). This is the behavior expected if the starting point is near the minimum of the velocity curve in Fig. 2.



Fig. 5. Recovery of ultrasonic attenuation and velocity in slightly deformed NaCl observed with 10 mc/sec <100> compressional waves. Unpublished measurements of Hikata *et al.*

It is believed that the verification of these predictions of the theory constitutes one of the strongest tests available of the validity of the dislocation damping theory. It is expected that the effect can be used with profit in the study of deformation, recovery and irradiation processes. Further work is in progress.

Acknowledgment

The work described here was supported in part through the National Science Foundation and in part United States Air Force through Aeronautical Systems Division, Air Force Systems Command.

References

- 1 A. Hikata, B. Chick, C. Elbaum and R. Truell: Acta Met. **10** (1962) 423.
- 2 R. M. Stern and A. V. Granato: Acta Met. 10 (1962) 358.
- 3 A. V. Granato, J. deKlerk and R. Truell: Phys. Rev. 108 (1957) 895.
- 4 A. V. Granato and K. Lücke: J. Appl. Phys. 27 (1956) 583, 789.
- 5 J.S. Koehler: Imperfections in Nearly Perfect Crystals, Edited by W. Shockley, J.H. Hollomon, R. Maurer and F. Seitz, Ch. 7, J. Wiley and Sons, New York (1952).
- 6 O. S. Oen, D. K. Holmes and M. T. Robinson: Private Communication.
- 7 A. Granato, A. Hikata and K. Lücke: Acta Met. 6 (1958) 470.

DISCUSSION

Klemens, P.G.: Would not the attenuation under the conditions of heavy velocity damping yield a reliable measure of the total length of all the dislocations, and thus of the dislocation density?

Truell, R.: Measurements of attenuation and velocity yield dislocation density and loop length, but one must be very careful because these are "effective values" having to do only with the sections which vibrate and contribute to the losses and velocity change. If dislocations are "well pinned" they would not ordinarily be "seen" by the ultrasonic measurements. In most cases the changes in these quantities, occurring during deformation, irradiation, recovery etc., can be seen and measured rather than their absolute values.

Seeger, A.: A closed expression for the velocity change due to dislocation exist, which is valid for all frequencies. Did you find it useful in speed to use the series?

Truell, R.: In our computations it was just as convenient to use the series since for most frequencies only a few terms are needed. At the higher frequencies (300-400 mc/sec) more terms are needed. The series expressions seem to exhibit the physics of the problem somewhat more than the closed expressions of which I am aware.