

## Generation of Ultrasonic Second and Third Harmonics due to Dislocations. II\*

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Experiments on the generation of third harmonics due to dislocations were carried out on aluminum single crystals. The amplitude of the third harmonics and the attenuation of the fundamental wave were measured as a function of impurity content, static bias stress, plastic deformation, and amplitude of the fundamental wave. The results of these experiments are compared with the predicted behavior. Good qualitative agreement is found between theory and experiment.

### INTRODUCTION

IN Paper I,<sup>1</sup> a theoretical consideration on the generation of ultrasonic harmonics has been presented. Based on the dislocation-string model, the treatment takes into account higher order terms in the expansion of potential energy due to the dislocation bowing out under the influence of applied oscillatory stresses. When the higher order terms are retained in the dislocation equation of motion, the solution contains odd multiples of the fundamental frequency, and if a static stress term is superimposed on the applied oscillatory stress, even multiples also appear. Consequently, the actual amplitudes of the second, third, and higher harmonics are obtained through the solution of the wave equation for the medium. One of the conclusions deduced from this treatment is that the third harmonic lends itself to the study of the interactions between dislocations and point defects more directly than does the second harmonic. Furthermore, the interpretation of the results may be simplified by the fact that for all cases of interest in this work the lattice contribution to the third harmonic can be neglected in comparison with the dislocation contribution. Following this reasoning, measurements of the third-harmonic amplitudes generated in aluminum single crystals of various impurity content are carried out.

The expression for the amplitude of the third harmonic [expression (41) in Ref. 1] may be rewritten in the following way:

$$A_3 = KNA_{10}^3 f_1(L_0) f_2(\alpha, x), \quad (1)$$

where  $K = 12\rho\omega^2 b^4 \Omega R^3 CC' / kA^4$ , the loop-length factor  $f_1(L_0) = 1/S_0^{3/4} T_0^{1/2} L_0^4$ , the attenuation factor  $f_2(\alpha, x) = e^{-3\alpha_1 x} - e^{-\alpha_3 x} / \alpha_3 - 3\alpha_1$ ;  $N$  is the dislocation density, and  $A_{10}$  is the stress amplitude of the fundamental wave at  $x=0$ . In the above expressions  $\rho$  is the density of the material,  $\omega$  the frequency of the fundamental,  $b$  the Burgers vector,  $\Omega$  a factor converting the shear strain to the longitudinal strain, and  $R$  the resolving shear

factor. Furthermore

$$C = W_e(1 + m \cos^2\theta - 2m \sin^2\theta),$$

$$C' = \frac{3(1 + 3m \cos^2\theta - 4m \sin^2\theta)}{2(1 + \cos^2\theta - 2m \sin^2\theta)},$$

in which  $W_e$  is the line energy of an edge dislocation per unit length,  $\theta$  the angle between Burgers vector and the dislocation line under zero stress, and (with  $W_s$  equal to the line energy of a screw dislocation per unit length)  $m = (W_e - W_s) / W_e$ . Finally,  $k$  is the wave vector;  $A$  is the effective mass of dislocation per unit length;

$$S_0 = (\omega_0^2 - \omega^2)^2 + (\omega d)^2, \quad \text{where } \omega_0 = (\pi/L_0)(C/A)^{1/2};$$

$L_0$  is the effective loop length of dislocation;  $d = B/A$  with  $B$  the damping coefficient of dislocation motion;

$$T_0 = \{\omega_0^2 - (3\omega)^2\}^2 + (3\omega d)^2;$$

$\alpha_1$  is the attenuation coefficient of the fundamental wave;  $\alpha_3$  is the attenuation coefficient of the third harmonic; and  $x$  is the distance the wave has traveled.

The experimental scheme used here to test the validity of the expression (1) is to vary each factor appearing in the expression and observe the corresponding change in  $A_3$ .

The factor  $N$  (dislocation density) may be varied by deforming the sample plastically.

The factor  $A_{10}$  (amplitude of the fundamental wave) can be varied simply by inserting an attenuator in the driving circuit.

The factor  $f_1(L_0)$  may be varied in two ways: (1) by investigating specimens of different impurity content, because the effective dislocation loop length of an annealed crystal is primarily determined by the impurity content; (2) by applying a small static bias stress to the specimen. The bias stress should be small enough so that no multiplication of dislocations takes place. With this procedure, some of the dislocations in the crystal will break away from the pinning points, resulting in an increase in the effective loop length, without changing the dislocation density.

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<sup>1</sup> A. Hikata and C. Elbaum, Phys. Rev. 144, 469 (1966).

The factor  $f_2(\alpha, x)$  may be tested in the experimental system used (i.e., the pulse-echo method<sup>2</sup>) by observing the echo pattern of the third harmonic resulting from the multiple reflections.

In the following, the experimental results conducted along the scheme outlined above are presented.

### EXPERIMENTAL TECHNIQUE

The general technique for the detection and measurement of ultrasonic harmonics was discussed in Refs. 2 and 3, and will not be repeated here.

However, some changes have been made in the instrumentation; in particular, we now use a high-power rf pulse generator with which it is possible to obtain an output of 1400 V peak to peak, corresponding to a peak power of 5 kW into 50  $\Omega$ . This unit uses the ceramic tetrode tube type 4CN15 in an electron coupled oscillator circuit. The oscillator tank circuit is coupled to the control grid and cathode in a Hartley configuration. An 800-V dc modulation pulse is applied to the screen which is normally biased at -100 V. The plate circuit is energized from a 4000-V dc supply. Coupling from the plate circuit to the 50- $\Omega$  load is accomplished with a  $\pi$  network to provide optimum impedance transformation.

In order to estimate the actual stress amplitude in the specimen, a capacitive pick-up method described elsewhere<sup>4</sup> was used. It is found that at full driving amplitude,<sup>5</sup> the stress at the driven end of the specimen is about  $\pm 10^6$  dyn/cm<sup>2</sup>.

Aluminum single crystals have been prepared with resistivity ratios varying over a wide range. Zone-refined aluminum of several degrees of purity was used in a modified Bridgman technique to grow specimens whose axes lie along several directions of high symmetry. The electrical resistivity ratio, defined as  $R = \rho_{300^\circ\text{K}} / \rho_{4.2^\circ\text{K}}$ , was measured using an eddy-current technique, which is a modification of that described by Bean *et al.*<sup>6</sup> The size of all the specimens was  $0.96 \times 0.96 \times 12.7$  cm. Details of deforming the specimens in an Instron testing machine are described elsewhere.<sup>7</sup> The application of the static bias stress is achieved by hanging small weights on a grip attached to the lower end of the specimen, as shown in Fig. 1.

<sup>2</sup> A. Hikata, B. B. Chick, and C. Elbaum, *J. Appl. Phys.* **36**, 229 (1965).

<sup>3</sup> U. S. Air Force Report No. AFML-TR-65-56, 1965 (unpublished).

<sup>4</sup> U. S. Air Force Report No. AFML-TR-65-56, Part II, 1966 (unpublished).

<sup>5</sup> By full amplitude is meant that the driving voltage for the transducer is maximum without electrical discharge in the vicinity of the transducer. This voltage depends on the quality of the bond between the transducer and the specimen and on the atmosphere in which the experiment is carried out; when the experiment is done in air, the moisture content is an important factor.

<sup>6</sup> C. P. Bean, R. W. DeBlois, and L. B. Nesbitt, *J. Appl. Phys.* **30**, 1976 (1959).

<sup>7</sup> A. Hikata, B. B. Chick, C. Elbaum, and R. Truell, *Acta Met.* **10**, 423 (1962).

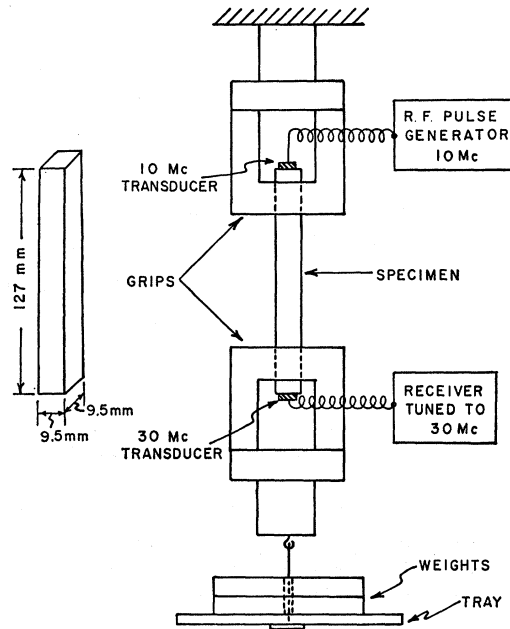


Fig. 1. Experimental arrangement (schematic).

Also indicated in Fig. 1 is the experimental arrangement, in which a 10-Mc/sec longitudinal pulsed signal is introduced into one end of the specimen by means of an appropriate quartz transducer. The 30-Mc/sec output is detected at the opposite end of the specimen with another transducer. With this arrangement it is possible to measure simultaneously the amplitude of the third harmonic and the attenuation of the fundamental wave.

### EXPERIMENTAL RESULTS AND DISCUSSION

In the following, the measured changes in the amplitude of the third harmonic are plotted in logarithmic scale, as a function of the variable under study, and normalized to their initial values (values at zero-bias stress, for example). Thus the factor  $K$  (including the dislocation density  $N$ ) in the expression (1) cancels, and the change in the amplitude of the harmonic is given by

$$\Delta A_3 = 20 \log_{10} \frac{A_3}{A_{30}} = 20 \log_{10} \frac{f_1(L_0)}{[f_1(L_0)]_0} + 20 \log_{10} \frac{f_2(\alpha, x)}{[f_2(\alpha, x)]_0} \quad (\text{dB}), \quad (2)$$

where  $A_{30}$  is the initial amplitude of the third harmonic. When  $|3\alpha_1 x - \alpha_3 x| \ll 1$ ,  $f_2(\alpha, x)$  can be expanded and the second term in expression (2) can be approximated by

$$-26x \times \Delta\alpha_1,$$

where  $\Delta\alpha_1$  is the measured change in the attenuation of the fundamental wave corresponding to the change  $\Delta A_3$ . Throughout this article, amplitude change of the

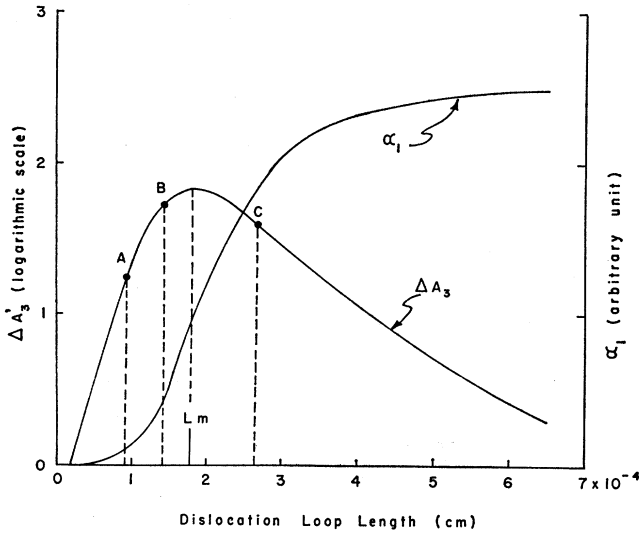


FIG. 2. Theoretical curve for the amplitude of the third harmonic and the attenuation of the fundamental wave for edge dislocations as a function of dislocation loop length.

third harmonic  $\Delta A_3'$  is defined by

$$\Delta A_3' = \Delta A_3 + 26 \cdot x \cdot \Delta \alpha_1 = 20 \log_{10} \frac{f_1(L_0)}{[f_1(L_0)]_0} \quad (\text{dB}).$$

**Effect of Loop Length: Impurity Content and Bias Stress**

As shown in Ref. 1, it is expected that the dislocations with smaller  $C$  values (edge dislocations) will generate larger harmonics, if other factors are identical. With this expectation, the calculated amplitude of the third harmonic due to edge dislocations is plotted in logarithmic scale as a function of dislocation loop length as shown in Fig. 2. In that figure the corresponding attenuation is also shown. It is seen that the third-harmonic amplitude has a maximum at a loop length of  $L_m \sim 2 \times 10^{-4}$  cm, if one uses the numerical values quoted in Ref. 1 for the factors  $A, B, C,$  and  $\omega$ . As mentioned in the Introduction, the effective loop length of an annealed specimen is determined primarily by impurity content, and a small bias stress causes an increase in the loop length. Thus, if the loop length in the sample before the application of a bias stress (initial loop length) lies somewhat below  $L_m$ , for example having a value given by the point  $A$  indicated in Fig. 2 (which may correspond to the loop length in a relatively impure specimen), then the amplitude of the third harmonic will increase initially with increasing bias stress. If the initial loop length in the sample is less than but close to  $L_m$  (point  $B$  for example in Fig. 2), which may correspond to the loop length in a specimen of intermediate purity, the amplitude of the third harmonic will go through a maximum as the bias stress increases. Finally, if the initial loop length is larger than  $L_m$ , point  $C$  for example (which may correspond to the loop

length in a specimen of highest purity used in these experiments), the amplitude of the third harmonic will decrease monotonically with increasing bias stress.

The experimental results shown in Fig. 3 are those for three samples of resistivity ratios 3100, 750, and 300 ( $\langle 100 \rangle$  axial orientation), subjected to a bias stress ranging from 0 to  $1.25 \times 10^6$  dyn/cm<sup>2</sup>. It is readily seen that the behavior of the third-harmonic amplitude follows the above prediction; i.e., in the impure specimen ( $R=300$ ) the amplitude of the third harmonic simply increase with bias stress; in the intermediate specimen ( $R=750$ ) the amplitude goes through a maximum; and in the zone refined specimen ( $R=3100$ ) the amplitude simply decreases with increasing bias stress.

In each of the above-mentioned tests, the attenuation of the fundamental wave as a function of bias stress was also measured, and the results are likewise shown in Fig. 3. It appears that for samples in the annealed state, the amplitude of the third harmonic is a much more sensitive guide to the changes in dislocation loop length that occur under the influence of applied bias stress than is the attenuation. This observation is consistent with model; in annealed specimens considered here the dislocation density  $N$  is low and  $\Delta \alpha_1$ , which is proportional to  $N$ , will be small. On the other hand  $\Delta A_3$  is independent of  $N$  [see Eq. (2)] and is a sensitive function of  $L_0$ , hence of bias stress.

**Effect of Plastic Deformation**

Figure 4 shows the effect of a bias stress on  $A_3$  and  $\alpha_1$  (hereafter called "the bias stress test") for the specimen

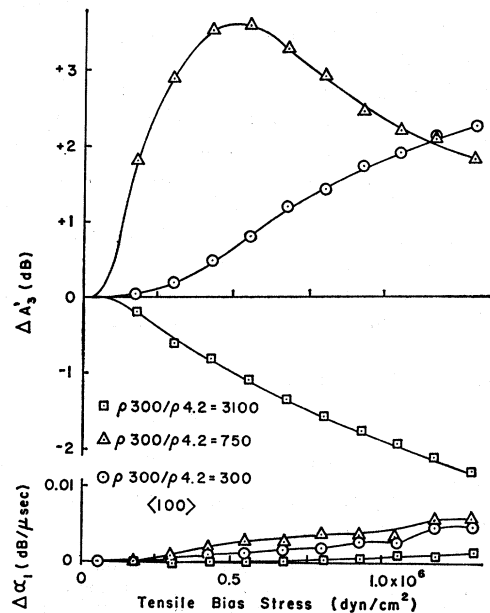


FIG. 3. Effect of impurity content on the third-harmonic amplitude  $\Delta A_3'$  and the attenuation of the fundamental wave  $\Delta \alpha_1$  as a function of bias stress for the specimens of  $\langle 100 \rangle$  axial orientation.

with  $R=30$  ( $\langle 100 \rangle$  axial orientation), before and after deformation in tension to a stress of  $6.7 \times 10^7$  dyn/cm<sup>2</sup>. In both cases  $A_3$  increases with increasing bias stress, but the amount of increase in the deformed state is much smaller than that in the annealed state, and is approaching a limiting value. The difference in the attenuation change  $\Delta\alpha_1$  for the annealed and deformed state is also significant; less than 0.01 dB/ $\mu$ sec and 0.06 dB/ $\mu$ sec, respectively.

Figure 5 shows a summary of the results of bias stress tests for the specimen with  $R=1200$  and a  $\langle 111 \rangle$  axial orientation. The specimen was deformed successively to  $1.5 \times 10^7$ ,  $2.2 \times 10^7$ ,  $3.2 \times 10^7$ , and  $6.3 \times 10^7$  dyn/cm<sup>2</sup>. After each deformation, the bias-stress tests were conducted. For this specimen, there is a maximum in  $\Delta A_3'$  curve in the annealed state. As the amount of plastic deformation increases, the position of the maximum in  $\Delta A_3'$  curve shifts toward smaller bias stresses and finally disappears. The corresponding attenuation changes of the fundamental wave are also shown in the figure. The change  $\Delta\alpha_1$  increases with increasing plastic deformation. After  $6.3 \times 10^7$  dyn/cm<sup>2</sup> deformation, the specimen was annealed at 550°C for 3 h and then tested with the bias stress. The results are marked "reannealed," and show almost complete recovery in the characteristics of both  $A_3$  and  $\alpha_1$ .

The results obtained on the specimen with  $R=3100$  and a  $\langle 100 \rangle$  orientation are shown in Fig. 6. In this case the maximum in  $\Delta A_3'$  curve did not appear even in the annealed state. The amplitude simply decreases with

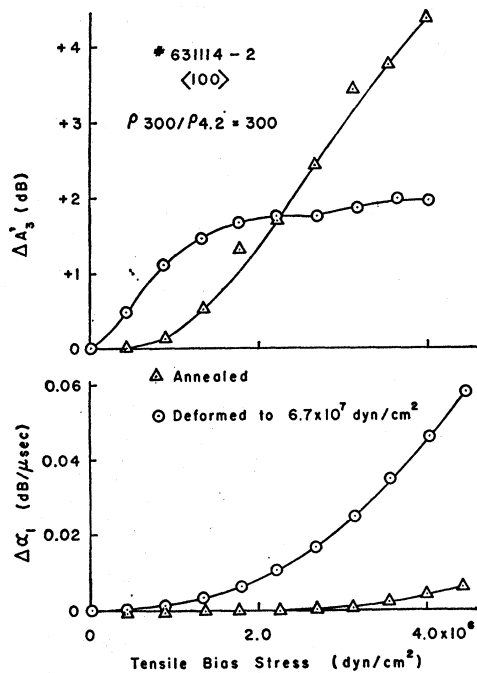


FIG. 4. Effect of plastic deformation on the amplitude change of the third harmonic  $\Delta A_3'$  and the attenuation change of the fundamental wave  $\Delta\alpha_1$  as a function of bias stress for the specimen of  $\langle 100 \rangle$  axial orientation and of  $\rho_{300}/\rho_{4.2}=300$ .

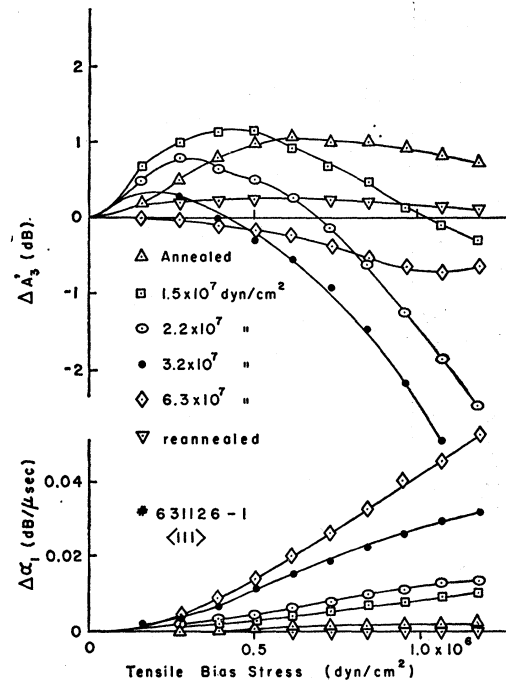


FIG. 5. Effect of plastic deformation on the amplitude change of the third harmonic  $\Delta A_3'$  and the attenuation change of the fundamental wave  $\Delta\alpha_1$  as a function of bias stress for the specimen of  $\langle 111 \rangle$  axial orientation and of  $\rho_{300}/\rho_{4.2}=1200$ .

increasing bias stress, and the amount of the decrease for a given bias stress increased after deformation.

If one compares these results with the theoretical curve (Fig. 2), it appears that the effective loop length becomes successively longer as the amount of deformation increases. This apparent increase in loop length with increased plastic deformation is discussed below in qualitative terms.

Dislocations in a crystal are known to form networks, and the onset of macroscopic plastic deformation of the crystal is primarily determined by the network loop length  $L_n$ . The network loop length  $L_n$  is further subdivided into  $L_0$  by pinning due to point defects, such as impurities and vacancies. The loop length which determines the interaction with ultrasonic stress waves is expected to be  $L_0$ ;  $L_0$  will be affected by a small static bias stress as well as by the oscillatory stress itself, if the latter is of sufficient amplitude. In the annealed state the dislocation density is small so that the network length  $L_n$  is expected to be large. In this case, however, the distribution of impurities between the bulk of the crystal and the dislocations is expected to be near equilibrium. For all the concentrations dealt with in this work, therefore, sufficient numbers of impurities are likely to have migrated to dislocations, thus reducing  $L_0$  considerably. When the crystal is deformed plastically, the dislocation density will be increased, resulting in a decrease in network loop length  $L_n$ , especially in specimens whose axes lie along directions

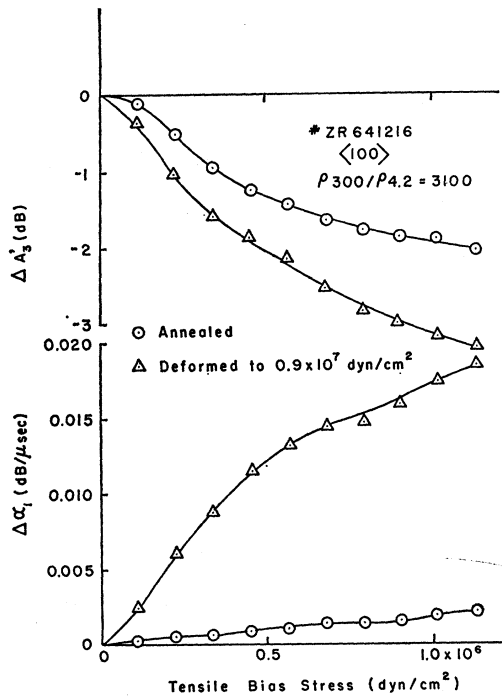


FIG. 6. Effect of plastic deformation on the amplitude change of the third harmonic  $\Delta A_3'$  and the attenuation change of the fundamental wave  $\Delta \alpha_1$  as a function of bias stress for the specimen of  $\langle 100 \rangle$  axial orientation and of  $\rho_{300}/\rho_{4.2} = 3100$ .

of high symmetry, for which several slip systems operate simultaneously. The macroscopic yield stress in such a specimen will be larger than that of an annealed specimen, because of the difference in network loop length  $L_n$ . On the other hand, dislocations created by the deformation will lie, on the average, in regions of low impurity content, because the concentration of impurities in the bulk is smaller than at the dislocations originally, and at room temperature redistribution of the impurities is not likely to occur appreciably during the course of these experiments. Thus, it is plausible that the loop length  $L_0$  responsible for the interaction with the ultrasonic waves becomes longer, on the average, with the increasing amounts of plastic deformation studied here.

In the case of the attenuation change  $\Delta \alpha_1$ , the dislocation density  $N$  appears explicitly in the comparison between deformed and undeformed crystals. Therefore, the successive increase in  $\Delta \alpha_1$ , with the increasing plastic deformation, for a given bias stress, can be interpreted in terms of a combined effect of the dislocation density increase and the loop-length increase.

#### Amplitude Dependence of the Third Harmonic and of the Attenuation

Equation (1) predicts that if there is no change in dislocation loop length  $L_0$  the factors  $f_1(L_0)$  and  $f_2(\alpha, x)$  remain constant, and the third-harmonic amplitude

should obey the third-power law, i.e.,  $A_3$  is proportional to  $A_1^3$ .

In Fig. 7, the variation of the third-harmonic amplitude and the attenuation of the fundamental wave, both as a function of the amplitude of the fundamental wave, are compared before and after deformation for the specimen of  $R=300$  and  $\langle 100 \rangle$  axial orientation. The behavior of the third harmonic in the annealed

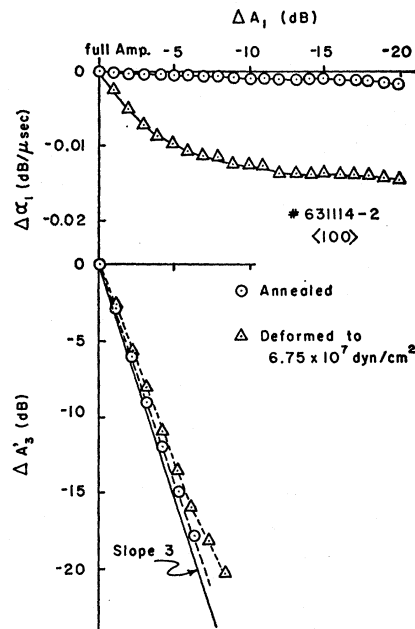


FIG. 7. Amplitude of the third harmonic  $\Delta A_3'$  and the attenuation of the fundamental wave  $\Delta \alpha_1$  as a function of the amplitude of the fundamental wave for the specimen of  $\langle 100 \rangle$  axial orientation and of  $\rho_{300}/\rho_{4.2} = 300$ .

state is such that the experimental points fall practically on the line of slope 3, while there is a slight deviation from the slope-3 condition where the measurements are made after deformation. The attenuation change in the annealed state was about 0.002 dB/ $\mu$ sec when  $A_1$  was decreased by 20 dB from the full amplitude. After deformation to a stress of  $6.75 \times 10^7$  dyn/cm<sup>2</sup>, a change of 20 dB in  $A_1$  was accompanied by a change of 0.014 dB/ $\mu$ sec.

Similar experiments were performed on the  $R=1200$  sample whose long axis coincides with the  $\langle 111 \rangle$  direction. The results are shown in Fig. 8. On this specimen data were recorded after it was slightly handled while being put in the grips, and after deformation to  $1.5 \times 10^7$  dyn/cm<sup>2</sup> and to  $2.2 \times 10^7$  dyn/cm<sup>2</sup>. The change in attenuation as a function of  $A_1$  becomes successively larger after each of these stages, finally changing by 0.1 dB/ $\mu$ sec after the last deformation. The amplitude of the third harmonic deviates slightly from slope 3 after annealing, and continues to deviate more with handling and with deformation.

The same general behavior (see Fig. 9) is also observed in the  $\langle 100 \rangle$  sample with  $R=3100$ : A striking

change in  $\alpha_1$  after the sample is deformed to  $0.9 \times 10^7$  dyn/cm<sup>2</sup> is observed when  $A_1$  is decreased by 20 dB. In the case of the amplitude of the third harmonic there is some deviation from slope 3 in the annealed state, and drastic deviation after deformation. Whenever the departure from the third-power law is observed, the corresponding attenuation of the fundamental wave is found to be amplitude-dependent.

As shown in Fig. 2, the factor  $f_1(L_0)$  has a maximum when plotted as a function of loop length  $L_0$ . If the loop length changes with the ultrasonic amplitude at all, the change must be in the direction of an increase in loop length with increasing amplitude. If the loop length of the specimen happens to be in the range  $L_0 > L_m$  (overdamped region<sup>1</sup>) the deviation from the third-power law may be explained through the effect

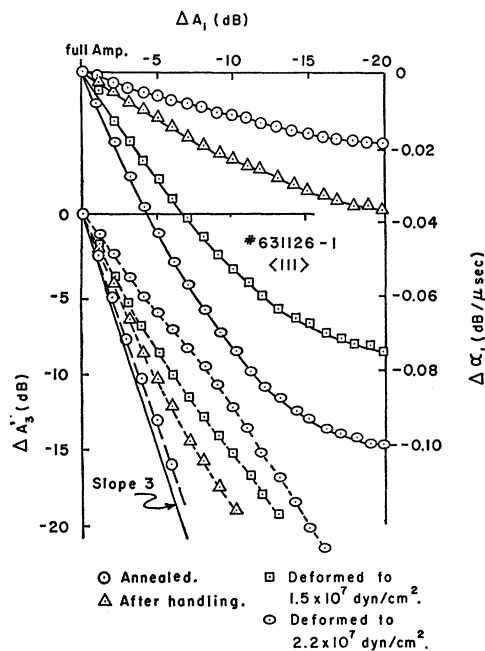


FIG. 8. Amplitude of the third harmonic  $\Delta A_3'$  and the attenuation of the fundamental wave  $\Delta \alpha_1$  as a function of the amplitude of the fundamental wave for the specimen of  $\langle 111 \rangle$  axial orientation and of  $\rho_{300}/\rho_{4,2} = 1200$ .

of the factor  $f_1(L_0)$ . Whether the loop length is larger than  $L_m$  or not, can be tested easily by applying a small bias stress to the specimen. If the bias stress reduces the amplitude of the third harmonic, one can assume that the loop length is in the overdamped region. On the other hand, if the third-harmonic amplitude increases with the bias stress, the loop length is in the underdamped region.<sup>1</sup> There is, however, experimental evidence that specimens whose dislocation loop length appears to be in the underdamped region (through the bias stress test) also show the deviation from the third-power law, as can be seen in Figs. 7 and 8. This experimental result strongly suggests that there must be a mechanism different from the simple string oscillation

playing a role in generating the third harmonic. The deviations from the third-power law observed here correspond to a slower rate of increase of  $A_3$  with increasing  $A_1$ . Any additional mechanism must, therefore, produce a decrease in the initial amplitude, i.e., the new third harmonic has to be of opposite phase to that generated by the first mechanism. The key to the search of a second mechanism seems to lie in the amplitude-dependent attenuation. There are four readily recognized reasons for the attenuation to change with amplitude: (1) change in dislocation density, (2) change in dislocation loop length without breakaway, (3) generation of harmonics, (4) hysteresis loss due to breakaway. Since the amplitude used in this investigation is small (in the order of  $10^6$  dyn/cm<sup>2</sup>) dislocation multiplication is not likely to occur, and (1) can be ruled out.<sup>8,9</sup> The mechanism for (2) is the side shift of pinning points along the dislocation line. The position of mobile pinning points along dislocation lines can be determined by minimizing the Helmholtz free energy of the system "dislocations plus pinning points," with respect to the location of the pinning points.<sup>10,11</sup> These positions are influenced by applied stresses. However, the ultrasonic frequencies, pulse width, and repetition

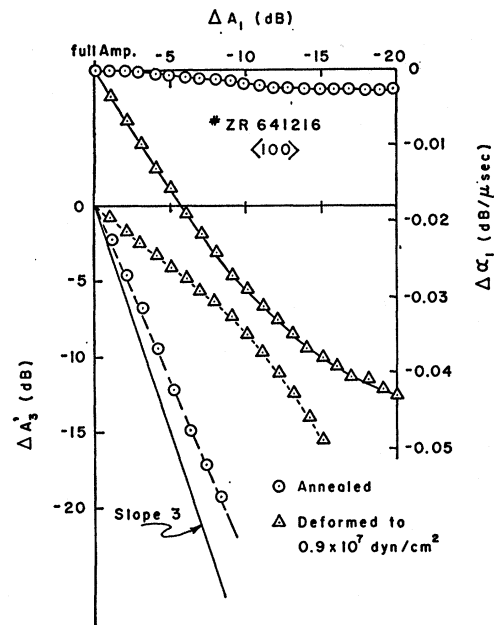


FIG. 9. Amplitude of the third harmonic  $\Delta A_3'$  and the attenuation of the fundamental wave  $\Delta \alpha_1$  as a function of the amplitude of the fundamental wave for the specimen  $\langle 100 \rangle$  axial orientation and of  $\rho_{300}/\rho_{4,2} = 3100$ .

<sup>8</sup> For all the specimens used, the macroscopic yield stress is about  $10^7$  dyn/cm<sup>2</sup>.

<sup>9</sup> After deformation, a certain amount of time was allowed to elapse before the experiment was performed again. Usually we waited until  $A_3$  and  $\alpha_1$  did not change measurably in a time equal to the time it takes to perform the experiment. In most cases this was about 2 h.

<sup>10</sup> G. Alefeld, *Phil. Mag.* **11**, 809 (1965).

<sup>11</sup> C. L. Bauer, *Phil. Mag.* **11**, 827 (1965).

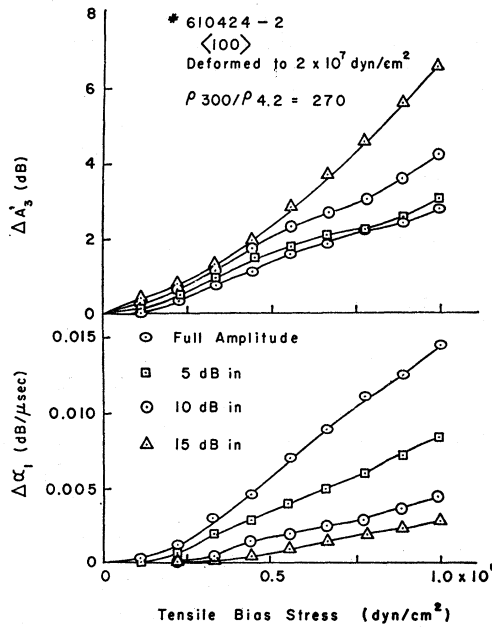


FIG. 10. Effect of the amplitude of the fundamental wave on the amplitude change of the third harmonic  $\Delta A_3$  and the attenuation of the fundamental wave as a function of bias stress for the specimen of  $\langle 100 \rangle$  axial orientation and of  $\rho_{300}/\rho_{4.2} = 270$ .

rate used here are not consistent with the time required, under stress, for a shift of pinning points to occur. It is, therefore, expected that the alternating stress (the wave) will not affect the distribution of loop lengths. Thus, (2) can also be eliminated. Generation of harmonics means that part of the energy of the fundamental wave is transferred to the harmonics, thus increasing the attenuation of the fundamental wave. Since the harmonic amplitude does not vary linearly with the fundamental wave amplitude, the attenuation must depend on the amplitude. Since the ratio of amplitudes of the harmonics (including the second harmonic) to that of the fundamental wave observed in the present experiments are in the order of  $10^{-2}$ , the attenuation change due to the energy transfer from the fundamental to harmonics is very small and cannot account for the observed attenuation change. Therefore, reason (3) can be ruled out.

Hysteresis loss due to breakaway was proposed as a mechanism for the amplitude-dependent attenuation, first by Koehler,<sup>12</sup> and elaborated later by Granato and Lucke.<sup>13</sup> According to the latter analysis, a dislocation breaks away from pinning points as stress increases (in the increasing stress quarter cycle), causing additional dislocation strain. In the decreasing stress quarter cycle, the dislocation follows a different path from that of the increasing stress cycle, thus producing a hysteresis in the stress-strain relation. Therefore, there are at least two dislocation contributions to the attenuation; one

<sup>12</sup> J. S. Koehler, *Imperfections in Nearly Perfect Crystals* (John Wiley & Sons, Inc., New York, 1952).

<sup>13</sup> A. Granato and K. Lucke, *J. Appl. Phys.* **27**, 583 (1956).

is due to the damping considered before, and the other is the hysteresis loss mentioned here. The damping loss is considered to be independent of the amplitude while the hysteresis loss is amplitude-dependent. The dependence on amplitude comes from the fact that the stress-strain relation of the breakaway process is nonlinear. Harmonic generation will, therefore, also result from the breakaway process. While we have not obtained an analytical form for the amplitude dependence of the third harmonic due to the breakaway process, it is clear that, in this case, the stress-strain relation is "softening." As mentioned earlier, the third harmonic generated by the string oscillation mechanism and that generated by the breakaway mechanism should be of opposite phase. This implies that the stress-strain relation for the simple string oscillation is hardening. According to the analysis reported in Ref. 1, the dislocations which give a hardening stress-strain relation are those of a predominantly edge-type. Therefore, the simultaneous operation of both mechanisms is consistent with the experimental observations, if one assumes that the dislocations responsible for the generation of the third harmonic due to the string oscillation mechanism are mostly edge-type dislocations.

#### Effect of Driving Amplitude on Bias-Stress Test

Since the amplitude of the ultrasonic stress wave employed here is sufficient to cause unpinning of dislocations from weak pinning points, the effect of bias stress on  $A_3$  and  $\alpha_1$  should be affected by the driving

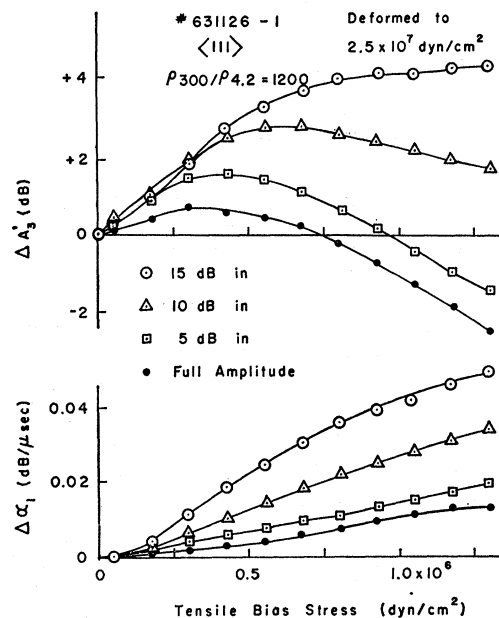


FIG. 11. Effect of the amplitude of the fundamental wave on the amplitude change of the third harmonic  $\Delta A_3$  and the attenuation of the fundamental wave as a function of bias stress for the specimen of  $\langle 111 \rangle$  axial orientation and of  $\rho_{300}/\rho_{4.2} = 1200$ .

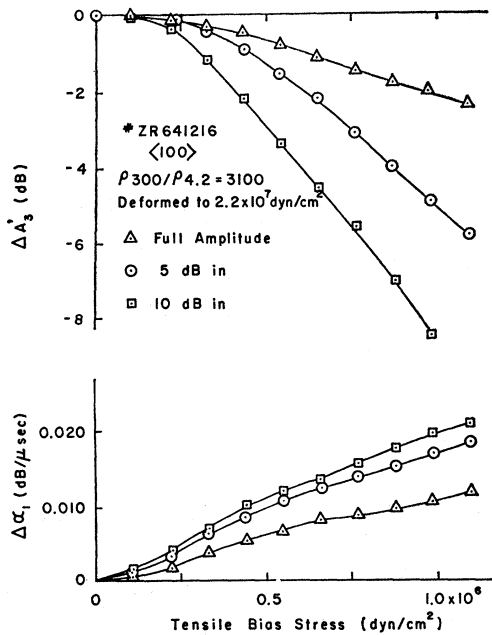


FIG. 12. Effect of the amplitude of the fundamental wave on the amplitude change of the third harmonic  $\Delta A_3'$  and the attenuation of the fundamental wave as a function of bias stress for the specimen of  $\langle 100 \rangle$  axial orientation and of  $\rho_{300}/\rho_{4.2} = 3100$ .

amplitude of the ultrasonic stresses. Figure 10 (for a specimen of  $R=270$ ), Fig. 11 (for a specimen of  $R=1200$ ) and Fig. 12 (for a specimen of  $R=3100$ ) show the experimental results of the bias stress tests conducted with various amplitudes of ultrasonic waves. In these figures, full amplitude has the same meaning as before, "5 dB in" means the driving amplitude is 5 dB less than the full amplitude; and so on. The results are again interpreted in terms of the predicted behavior shown in Fig. 2. In the case of Fig. 10, the change in  $A_3$ ,  $\Delta A_3'$ , increases monotonically with increasing bias stress, and the larger the driving amplitude the smaller

is the change observed in  $\Delta A_3'$ . The loop length  $L_0$  for this specimen is expected to be near point A on the theoretical curve (Fig. 2), and at full amplitude, the loop length approaches  $L_m$ . Since the attenuation change for a given loop-length change is largest for the loop length near  $L_m$  (the slope of the curve  $\alpha_1$  is maximum near  $L_m$ ) the measured attenuation change should be largest for full driving amplitude and should decrease with decreasing driving amplitude, as observed in this case. In the case of Fig. 11,  $\Delta A_3'$  go through a maximum with bias stress, indicating that the loop length for this specimen is near point B (in Fig. 2). As the driving amplitude decreases, the position of the maximum in  $\Delta A_3'$  curve shifts toward larger bias stresses, and at the lowest driving amplitude (15 dB in) it disappears. In contrast to the case of Fig. 10, the larger the driving amplitude the smaller is the change observed in the attenuation. This is consistent with the prediction, because  $d\alpha_1/dL_0$  decreases in the region where  $L_0 > L_m$ . A similar argument applies in the case of Fig. 12, where  $A_3$  decreases with increasing bias stress and  $\alpha_1$  behaves as in the case of Fig. 11. Here the loop length under no bias stress appears to lie in the region  $L_0 > L_m$  and  $L_0$  continues to increase with increasing bias stress.

#### SUMMARY

In conclusion, it is found that the theory presented in Ref. 1 is substantiated by the experimental results on the dependence of the amplitude of the third harmonic  $A_3$  on dislocation loop length. Small amounts of plastic deformation cause an apparent increase of the effective dislocation loop length, as determined from the dependence of the amplitude  $A_3$  and of the attenuation  $\alpha_1$  on bias stress. The dependence of  $A_3$  and of  $\alpha_1$  on the amplitude of the fundamental wave  $A_1$  indicates that the effective dislocation loop length increases with increasing amplitude  $A_1$ ; further study is needed to interpret these results.