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Temperature Dependence of the Dislocation Drag Constant in Antimony

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The over-damped resonance of dislocation segments is investigated in antimony single crystals in the frequency range 7.5 to 172.5 MHz in order to obtain information on the dislocation drag constant. The dislocation part of the losses is separated by two methods, namely by subtracting the internal friction of a crystal before straining from that of the strained crystal and by calculating the value analytically. The dislocation density is determined by etch pit counting. The data of the two methods show a good agreement. The frequency dependence of the internal friction due to dislocation motion is well described by the frequency profile given by the Granato-Lücke theory for an exponential length distribution of dislocation loops. The drag constant is obtained and increases from 6×10^{-5} dyn s/cm² at 100 K to 9×10^{-5} dyn s/cm² at 300 K. The results correspond to a theoretical analysis of the dislocation drag near the Debye temperature due to the joint action of the phonon wind and "slow" phonon relaxation. Temperature dependences are obtained for the dislocation segment lengths in the deformed and non-deformed crystals.

С целью получения информации о коэффициенте демпфирования дислокации в монокристаллах сурьмы исследовался задемпфированный резонанс дислокационных сегментов в интервале частот 7,5 до 172,5 MHz. Дислокационная компонента выделялась двумя методами: путем вычитания из внутреннего трения деформированного кристалла величины внутреннего трения до деформации, а также аналитическим путем. Плотность дислокаций определялась по фигурам травления. Данные, полученные обоими методами, согласуются между собою. Частотная зависимость дислокационного внутреннего трения хорошо описывается частотным профилем, предсказываемым теорией Гранато и Люкке для случая экспоненциального распределения петель по длинам. Получена величина коэффициента демпфирования дислокаций, которая росла от 6×10^{-5} dyn s/cm² при 100 K до 9×10^{-5} dyn s/cm² при 300 K. Результаты согласуются с теоретическим рассмотрением демпфирования дислокаций в окрестности дебаевской температуры в рамках суперпозиции механизмов релаксации „медленных" фононов и фононного ветра. Получена температурная зависимость длины дислокационного сегмента в деформированном и недеформированном монокристаллах.

1. Introduction

To comprehend the plastic properties of crystals one needs a knowledge of the characteristics of dislocation dynamics, among which the drag constant B is of greatest importance. The reason is that the value of B completely determines the fast motion of the dislocation over local barriers and also has a substantial influence on thermally activated surmounting of stronger barriers [1]. The drag on dislocation motion results from the dissipation of energy via many different channels of which the most important are the ones related with the phonon sub-

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system of the crystal elementary excitations (and with the electron subsystem at low temperatures). The microscopic mechanisms of the drag can be identified through investigations of the temperature dependence of the drag constant in each specific range of temperatures. Measurements of the over-damped dislocation resonance are especially promising.

The present paper describes investigations of that part of the dislocation drag for which the phonon subsystem of excitations might be responsible. Single crystals of antimony were chosen as the object. The contribution of some phonon mechanisms to the drag constant is essential below the Debye temperature θ, while others can manifest themselves at temperatures above θ. That is why we have chosen the range 100 to 300 K containing the Debye point which is 200 K in the case of antimony. This interval happened to be technically advantageous, too.

2. Experimental

The measurements were carried out by the ultrasonic pulse-echo technique in the frequency range between 7.5 and 172.5 MHz. Longitudinal waves were excited in the sample with an X-cut quartz plate, 8 mm in diameter, with the fundamental frequency 7.5 MHz. The experimental equipment was earlier described in detail in [2].

All the experiments were carried out with antimony single crystals of the same orientation, grown from a 99.995% pure material. The crystals were grown on a seed in a sectional graphite crucible placed in a 2×10^{-2} Torr vacuum [3]. The as-grown single crystals were of dimensions $8 \times 36 \times 110$ mm^3. The single crystal was cleaved in liquid nitrogen along the cleavage plane (111) and then the spark cutting technique was employed to prepare $8 \times 18 \times 18$ mm^3 samples. The (111) plane served as a lateral surface of the 8×18 mm^2 specimen. The 8 mm edge was along $[1\bar{1}0]$. This crystallographic axis coincided with the direction of sound propagation. The as-grown surfaces normal to $[1\bar{1}0]$, 18×18 mm^2 in size, were the working surfaces on which the quartz plates were cemented. The careful preparation of the moulds allowed one to obtain the as-grown working surfaces parallel to within ± 2 μm/cm. To reduce the non-parallelity, the samples were lapped on an iron lapping plate until they were parallel to ± 0.3 μm/cm.

Temperatures from the 1.3 to 100 K range were measured with a GaAs thermometer providing an accuracy of ± 0.01 K. The voltage across the thermometer produced by a stabilized current was measured with a microvolt/microammeter. Between 100 and 300 K a copper–constantan thermocouple was employed whose thermo-e.m.f. was measured with the same device. The temperature was recorded continuously with an automatic recording potentiometer. A given temperature could be maintained and controlled with an electronic mechanical semiautomatic system to an accuracy of 0.1 K.

The dislocation density was determined from etch pit patterns on the cleavage plane (111). The etchant was a solution of $FeCl_3$ in methylalcohol. After etching the samples were rinsed with concentrated HCl and distilled water [4]. Fig. 1a and b represent the etch pit patterns obtained with non-deformed and deformed specimens. The etch pits were distributed randomly; hence, to improve the accuracy of the dislocation density determination, the number of the etch pits was counted and averaged over a large number of microscope visual fields (not less than 50).

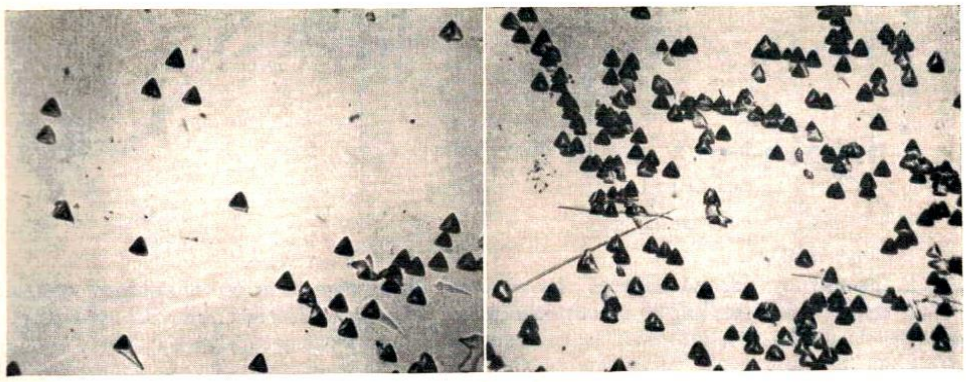

Fig. 1. Dislocation etch pits on the cleavage plane (111) of antimony single crystals (77×). a) Unstrained crystal; b) strained crystal

3. Experimental Results

3.1 Separation of the dislocation part of ultrasound attenuation

The absorption of the ultrasonic signal was measured at different frequencies. The value obtained consists of dislocation and non-dislocation parts. As is known, the background losses are of the same order of magnitude as those due to dislocation motion, hence the problem of their separation is of principle importance.

Several methods are known for separating the dislocation part of the attenuation. According to one of them, the non-dislocation (background) component is assumed to coincide with the attenuation in samples subjected to heavy irradiation (10^8 to 10^{10} R) by neutrons or γ-rays. As a result, dislocations are pinned to radiation-induced crystal defects [5 to 11]. The drawback of the method is that it can be applied only to a number of materials and in a limited temperature range [9, 12, 13]. Another method is to take for the background the attenuation in annealed specimens [14 to 18] or in specimens before strain treatment [19]. Still another technique of determining the dislocation part of ultrasound absorption consists in subtracting the total contribution of non-dislocation mechanisms evaluated analytically [12, 20 to 22].

The authors have tried all the three techniques. We had to abandon the first method since non-deformed samples subjected to γ-radiation from a CO^{60} source (temperature 350 K, radiation dose up to 10^8 Rad) showed no absorption values below those of annealed crystals. Two samples that were strained to 0.1% showed a reduction in the amount of attenuation through the entire frequency range after they were subjected to a dose of (1 to 3) $\times$ 10^8 Rad; yet the values obtained were not below the attenuation observed in the crystals before straining.

To employ the second method of separating the dislocation part of attenuation, the initial dislocation density was determined first and frequency dependences of the attenuation rate were measured at fixed temperatures decreasing from 300 to 100 K. Then the temperature was lowered to 1.4 K and the temperature dependence of $\alpha(T)$ was measured at two fixed frequencies, viz. 7.5 and 97.5 MHz. The $\alpha(T)$ curves were also measured when the sample was heated back to room temperature. At 300 K the sample was strained in compression to small residual deformations (not exceeding 0.2%). The measurements were

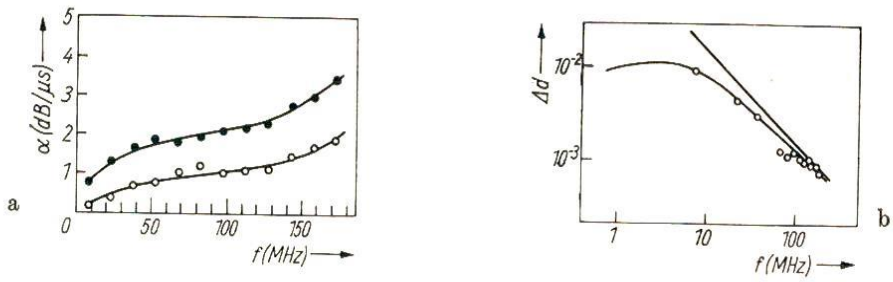

Fig. 2. Frequency dependences of the a) total absorption rate (○ unstrained crystal; ● strained crystal) and of the b) dislocation part of the decrement at $T = 155$ K

repeated for the deformed sample. The difference in attenuation between the deformed sample and the same crystal before straining was assumed to be dislocation contribution. It was related to the difference in the density of dislocations observed in the deformed (7.7×10^4 cm^{-2}) and non-deformed ($3.2 \times \times 10^4$ cm^{-2}) samples. Frequency dependences of the total ultrasound absorption in both non-deformed and deformed samples and of the dislocation decrement, Δd, are shown in Fig. 2a and 2b. It is easy to see from Fig. 2b that the experimental frequency dependence of Δd conforms to the theoretical curve calculated for the exponential length distribution of dislocation loops (solid line in Fig. 2b [23, 24]).

The dislocation density of strained crystals was determined after the frequency measurements in the entire range of temperatures had been completed, hence it was necessary to make sure that the density did not change with temperature. To that end temperature dependences of the attenuation were taken with decreasing sample temperature from 300 to 1.4 K (direct run) and on heating the samples from 1.4 back to 300 K (reverse run). The results are presented in Fig. 3. As can be seen, the direct and the reverse runs of the temperature dependence do not coincide which suggests changes of the dislocation segment mean length and of the dislocation density. Direct measurements of the dislocation density before and after the helium temperature experiment have also shown the value to increase by 30 to 100%. The increased density of dislocations results from the deformation of the sample by the quartz transducer cemented to the crystal, due to the difference in their thermal expansion constants [8, 9]. Hence, the second method of separating the background and dislocation losses is not applicable in the temperature range 1.4 to 300 K. However, between 100 and 300 K no multiplication of dislocations was observed and the temperature dependences of α on cooling and heating did coincide. Thus, the second and the third methods of loss separation can be confronted in the range 100 to 300 K.

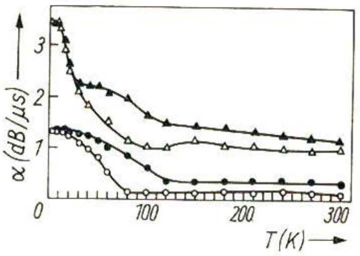

Fig. 3. Temperature dependences of the ultrasound absorption at sample cooling (○, △) and heating (●, ▲) ○, ● 7.5 MHz; △, ▲ 97.5 MHz

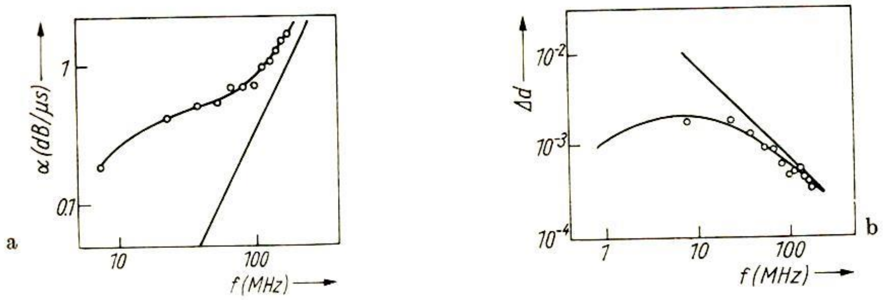

Fig. 4. Separation of the dislocation part of the decrement from the total absorption rate at $T = 260$ K. a) ○ Experimental frequency dependence of the total ultrasound absorption. Straight line: background losses due to the mechanisms which determine the quadratic frequency dependence of α (i.e. thermoelastic and phonon viscosity losses). b) Frequency dependence of the logarithmic decrement corresponding to the normalized frequency profile for the case of exponentially distributed dislocation loop lengths

For separating the dislocation part of the absorption by the third method, the frequency dependence of α was measured in unstrained samples. At higher frequencies the major contribution to the non-dislocation losses is known to be due to thermo-elastic mechanisms [25] and phonon viscosity [26] while at lower frequencies diffraction losses dominate. Contributions of other phenomena can be neglected [27]. The diffraction losses were calculated in the isotropic approximation [28, 29]. An important feature of the mechanisms described in [25, 26] is the quadratic frequency dependence of the amount of attenuation. If the frequency dependence of the ultrasound absorption is plotted on a lg–lg scale (see Fig. 4a), the contribution due to the mechanisms [25, 26] should appear as a straight line of slope 2. On the other hand, the dislocation losses are of overdamped resonance nature [30] which is well described by the normalized frequency dependence corresponding to the case of exponentially distributed dislocation loop lengths [24]. Thus, we simply have to match the position of the straight line of slope 2 in such a way that its difference from the measured values of α would yield on a lg Δd versus lg f plot the normalized frequency profile (Fig. 4b). The plausibility of this method for separating the dislocation part of attenuation was confirmed experimentally in [12, 21, 25]. In the present work the dislocation losses were determined by this method for five antimony samples. Hereafter we will compare the dislocation components of the attenuation rate as determined by the second and third methods.

3.2 *Effect of temperature on the frequency-dependent losses due to dislocation motion*

Fig. 5a and b show the frequency dependences of the dislocation decrement for three values of temperature in the range 100 to 300 K for deformed and non-deformed samples. Presented on the same graphs are also the high-frequency asymptotic curves for the decrement rate, reflecting its behaviour at frequencies much higher than maximum [30]. In this temperature range the maximum value of the rate Δ_m increases with growing temperature, the position of the maximum shifts towards lower frequencies and so do the high-frequency asymptotic curves.

According to the Granato-Lücke theory [23], the maximum value of the decrement Δ_m, the frequency of the maximum absorption f_m calculated for the

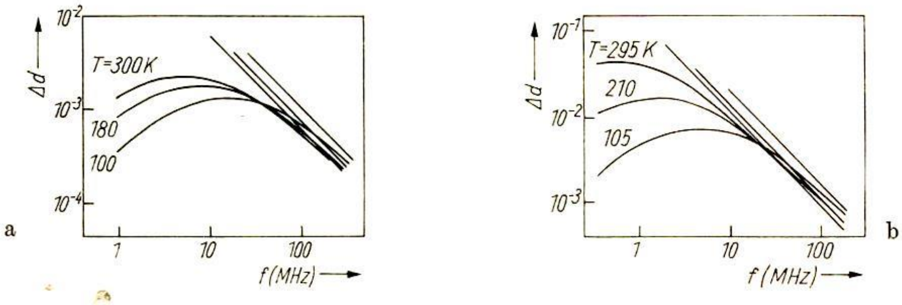

Fig. 5. Effect of temperature on the frequency dependence of the dislocation part of the decrement, Δd, in an a) unstrained crystal (background losses have been separated analytically) and b) in a strained crystal (the attenuation rate of the crystal before straining has been assumed as background)

exponential length distribution of dislocation loops, and the decrement at frequencies much higher than f_m, respectively, are given by the relations

$$\Delta_m = 2.2\Omega\Delta_0\Lambda L^2 , \tag{1}$$

$$f_m = \frac{0.084\pi C}{2L^2B} , \tag{2}$$

and

$$\Delta_\infty = \frac{4\Omega Gb^2\Lambda}{\pi^2 Bf} , \tag{3}$$

where Ω is an orientation-dependent factor, $\Delta_0 = 8Gb^2/\pi^3C$, G is the shear modulus, b the amount of the Burgers vector, C the line tension (evaluated as $2Gb^2/\pi(1-\nu)$), ν Poisson's ratio (equal to 0.251 [32]), Λ the dislocation density, L the mean length of the dislocation loop, and B the drag constant.

Since the dislocations in the glide plane {111} cannot be revealed in antimony by the etch pit technique, the sample orientation was chosen so that the ultrasonic wave gave no shear stress component in the {111} plane, the major contribution to the dislocation absorption of ultrasound being provided by the system $\{1\bar{1}1\}\langle 101\rangle$ [31]. The orientation-dependent factor Ω was calculated for that system with help of [30, 33]. The corresponding magnitude of the Burgers vector is 8.06×10^{-8} cm [31, 34]. The temperature dependence of elastic moduli from 300 to 873 K can be found in [35]; we have extrapolated them to 100 K. Having determined Δ_∞ from the high-frequency asymptotic behaviour and Λ from independent etch pit counts, one can obtain the drag constant B from (3). The value can also be found from (1) and (2). Fig. 6 shows the temperature dependence of B as obtained from Δ_∞. One of the $B(T)$ curves was provided by the second of the above methods for separating the dislocation losses. Another one is the result of measurements carried out on the same sample before straining applying the analytical method. The third curve was obtained from data on another sample, also through the use of the analytical method. As is seen in Fig. 6, the three curves are in good agreement which supports the analytical method of separating the dislocation part of absorption in the case of antimony.

The drag constant B changes with temperature between 0.6×10^{-4} dyn s/cm² (at 100 K) and 0.9×10^{-4} dyn s/cm² (at 300 K).

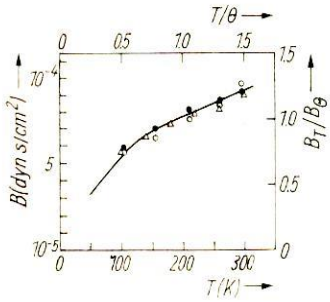

Fig. 6

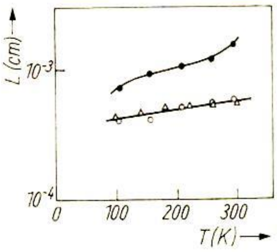

Fig. 7

Fig. 6. Temperature dependences of the drag constant as obtained via the analytical method of background separation (△, ○) and with the assumption of unstrained crystal losses as the background (●). The solid line is a theoretical curve [36]; B_θ denotes the drag constant value at the Debye temperature

Fig. 7. Temperature dependences of the mean effective dislocation segment length in unstrained (○, △) and strained (●) crystals

Fig. 7 represents the temperature dependence of the mean effective dislocation segment length L. The value was estimated from (2) for deformed as well as non-deformed crystals.

4. Discussion

In the temperature range under discussion an important role in damping the dislocation motion is played by phonon mechanisms. Among such phonon effects as combination scattering, phonon viscosity, phonon wind, flutter effect, and thermo-elastic losses at temperatures close to θ, the phonon wind mechanism stands out. Recently another efficient channel for the dissipation of the energy of moving dislocations has been discovered, namely the "slow" phonon relaxation [36]. At the temperatures of our interest the joint action of these two mechanisms can be the dominant factor in the dislocation drag. A reasonable comparison of experimental results with theory [36] apparently can consist in confronting the temperature run of B as provided by experiment with the theoretical prediction. This can be conveniently done in terms of dimensionless variables $B(T)/B(\theta)$ versus T/θ. According to [36] we have

$$\frac{B(T)}{B(\theta)} = \frac{f_1(T/\theta)}{f_1(1)}\,[1 - \delta f_2(1)] + \delta\,\frac{l}{l_0}\,f_2\left(\frac{T}{\theta_M}\right) \tag{4}$$

with

$$f_1\left(\frac{T}{\theta}\right) = \left(\frac{T}{\theta}\right)^5 \int\limits_0^{\theta/T} \frac{dt\, e^t t^5}{(e^t - 1)^2}\, \frac{\operatorname{arctg} \beta \dfrac{T}{\theta}\, t}{\beta \dfrac{T}{\theta}\, t}$$

and

$$\beta = 2k_D r_0\,.$$

Here r_0 is the effective radius of the dislocation core and k_D the Debye limit of the phonon spectrum;

$$f_2\left(\frac{T}{\theta}\right) = \frac{\theta}{T}\,\frac{\exp(\theta/T)}{[\exp(\theta/T) - 1]^2},$$

l is the phonon path length, δ a dimensionless parameter which can be found from experimental data by extrapolating the high-temperature asymptotic behaviour of $B(T)/B(\theta)$ as a function of T/θ to zero temperature, and θ_M is a phenomenological parameter of "slow" phonons. The ratio l/l_θ was estimated from $(c(\theta)/c(T)) \times (\theta/T)$ where c denotes the heat capacity. As can be seen from Fig. 6, the theoretical curve (solid line) agrees with the experimental data at $\delta = 0.67$. Thus, the temperature run of B can be interpreted in terms of a superposition of the phonon wind and the "slow" phonon relaxation mechanisms. The magnitude of δ, namely $\delta = 0.67$, indicates that the relaxation of "slow" phonons is in this case an essential mechanism in the dislocation drag. The importance of this mechanism has been already pointed out by other authors [37, 38]. Hence, the dislocation dynamics should be sensitive to the form of the crystal phonon spectrum.

Although the temperature run of B is of importance, it is also necessary to compare the absolute values predicted by the theory and those given by the experiment. According to [36], the contribution of the phonon wind and the relaxation of "slow" phonons to the dislocation drag is described by the relation

$$B = \left[4 + \left(\frac{|n|}{G} - 6\right)^2\right] \frac{\hbar}{b^3} \left(\frac{k_D b}{2\pi}\right)^5 \left[f_1\left(\frac{T}{\theta}\right) + \lambda_\theta \frac{l}{l_\theta} f_2\left(\frac{T}{\theta_M}\right)\right], \qquad (5)$$

where n is the Murnaghan modulus and

$$\lambda_\theta = \frac{\delta f_1(1)}{1 - \delta f_2(1)}.$$

Assuming for antimony $|n|/G = 10$ one can obtain from (5) absolute values of B which are close to the experimental data, e.g., at 300 K the equation yields 9×10^{-5} dyn s/cm².

To find out the role of other mechanisms of dislocation drag one needs data on the behaviour of B in a wider range of temperatures, particularly in the low-temperature region (1.3 to 100 K).

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References

[1] Yu. Z. Estrin, phys. stat. sol. (a) **26**, K161 (1974).
[2] M. V. Zinovev, P. P. Pal-Val, V. Ya. Platkov, L. N. Polunina, and A. G. Ryzhakov, Proc. Phys. Tech. Inst. Low Temperatures, Vol. 32, Kharkov 1974 (p. 88).
[3] L. A. Maslova, S. V. Tsivinskii, and B. N. Alexandrov, Kristall und Technik **8**, 629 (1973).
[4] L. M. Soyfer, Zashchita metallov (Metal Protection) **1**, 29 (1965).
[5] G. A. Alers and D. O. Thompson, J. appl. Phys. **32**, 283 (1961).
[6] R. M. Stern and A. V. Granato, Acta metall. **10**, 358 (1962).

[7] H. INAGAKI, F. A. HULTGREN, and K. LÜCKE, Acta metall. **18**, 713 (1970).
[8] D. LENZ, K. LÜCKE, A. OSTERMANN, and W. A. SIBLEY, J. Acoust. Soc. Amer. **45**, 1374 (1969).
[9] P. WINTERHAGER and K. LÜCKE, J. appl. Phys. **44**, 4855 (1973).
[10] H. AKITA and N. F. FIORE, J. appl. Phys. **44**, 20 (1973).
[11] N. P. KOBELEV and YA. M. SOYFER, Fiz. tverd. Tela **17**, 1516 (1975).
[12] D. LENZ and K. LÜCKE, Z. Metallk. **60**, 375 (1969).
[13] W. A. FATE, J. appl. Phys. **43**, 835 (1972).
[14] O. M. M. MITCHELL, J. appl. Phys. **36**, 1083 (1965).
[15] T. SUZUKI, A. IKUSHIMA, and M. AOKI, Acta metall. **12**, 1231 (1964).
[16] H. INAGAKI, K. SANO, and M. SHIMIZU, Crystal Lattice Defects **4**, 57 (1973).
[17] N. IGATA, R. DOMOTO, and R. R. HASIGUTI, J. Phys. Chem. Solids **31**, 1883 (1970).
[18] R. R. HASIGUTI, N. IGATA, and M. SHIMOTOMAI, Trans. Japan. Inst. Metals **9**, Suppl., 42 (1968).
[19] V. M. ANDRONOV, A. M. PETCHENKO, and V. I. STARTSEV, Acoust. J. (USSR) **21**, 502 (1975).
[20] W. P. MASON and A. ROSENBERG, Phys. Rev. **151**, 434 (1966).
[21] W. P. MASON and A. ROSENBERG, J. appl. Phys. **38**, 1929 (1967).
[22] W. P. MASON and D. MCDONALD, J. appl. Phys. **42**, 1836 (1971).
[23] A. V. GRANATO and K. LÜCKE, in: Physical Acoustics, Ed. W. P. MASON, Vol. **4**, Part A, New York/London 1966.
[24] O. S. OEN, D. K. HOLMES, and M. T. ROBINSON, US AEC Rep. ORNL-3017, 1960 (p. 3); see [22].
[25] K. LÜCKE, J. appl. Phys. **27**, 1433 (1956).
[26] A. AKHIEZER, J. Phys. (USSR) **1**, 277 (1939).
[27] R. TRUELL, C. E. ELBAUM, and B. B. CHICK, Ultrasonic Methods in Solid State Physics, New York/London 1969.
[28] H. SEKI, A. V. GRANATO, and R. J. TRUELL, J. Acoust. Soc. Amer. **28**, 230 (1956).
[29] E. PAPADAKIS, J. Acoust. Soc. Amer. **31**, 150 (1959).
[30] A. V. GRANATO and K. LÜCKE, J. appl. Phys. **27**, 583 (1956).
[31] L. M. SOYFER, Fiz. Metallov i Metallovedenie **19**, 741 (1965).
[32] K. H. SCHRAMM, Z. Metallk. **53**, 729 (1962).
[33] B. LEGROS, These Doct. es Sci. Phys., University P. Sabatier, Toulouse 1972.
[34] C. S. BARRETT, P. CUCKA, and K. HAEFUER, Acta cryst. **16**, 451 (1963).
[35] A. A. VALIEV, YU. A. BURENKOV, S. P. NIKANOROV, and A. V. STEPANOV, Fiz. tverd. Tela **15**, 3098 (1971).
[36] V. I. ALSHITS and V. I. INDENBOM, Uspekhi fiz. Nauk **115**, 3 (1975).
[37] A. M. PETCHENKO and V. I. STARTSEV, Fiz. tverd. Tela **16**, 3655 (1974).
[38] E. V. DARINSKAYA and A. A. URUSOVSKAYA, Fiz. tverd. Tela **17**, 2421 (1975).

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