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Ultrasonic study of α-complex peaks in single crystals and polycrystals of pure molybdenum at megahertz frequencies

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Abstract. Measurements of the temperature dependence of the ultrasonic attenuation at 5, 10 and 28 MHz in molybdenum single crystals of 99.995% purity, oriented along ⟨111⟩, ⟨110⟩ and ⟨100⟩, have provided a detailed study of the α complex of dislocation relaxation peaks following compressive stressing at room temperature. At 5 MHz the peaks were at 148, 167 and 206 K, independent of the orientation of the crystal. As expected, each of the peaks moved to higher temperatures as the frequency of measurement was increased and from these data the activation enthalpies and attempt frequencies were 0.18 eV and 6.6×10^{11} Hz, respectively, for α_1, 0.22 eV and 1.84×10^{13} Hz, respectively, for α_2, and 0.20 eV and 4.95×10^{11} Hz, respectively, for α_3. Similar measurements at 10 MHz on polycrystals of the same purity showed the effects of increasing the applied stress from 0.2 to 29 MPa and of irradiation with doses D_γ of γ-rays up to 8.6×10^8 γvt and doses D_n of neutrons up to 10.8×10^9 nvt. It was found that the peaks' strength Q_{max}^{-1} varied as D^{-m}, where m is a parameter for each irradiated peak.

1. Introduction

Chambers [2] studied the internal function of BCC metals and showed that deformation produced a composite peak α below room temperature, a well defined β complex at around room temperature and a γ peak at higher temperatures. It is generally accepted that the α and γ peaks are intrinsic and associated with kink pair formation and diffusion of geometric kinks on dislocations. The β complex is due to interaction between point defects and dislocations. Fantozzi *et al* [3] reviewed the role of the large difference in the mobilities of screw and edge dislocations in BCC metals on further proliferation of the peaks. Theoretical calculations have shown that the highest activation enthalpy is associated with the γ peak which is due to the generation of kink pairs on screw dislocations. Kink motion on screw dislocations would occur at very low temperatures and would be unlikely to be seen experimentally. More than one type of kink can occur on a dislocation, which may account for the number of peaks observed in the α complex. There is no detailed study of the α complex at megahertz frequencies except the work of Mongy [4] and Polotskii *et al* [5] where the former observed only two peaks and the latter a single peak. In our previous paper (hereafter referred to as I) [1], we studied the mechanical spectroscopy, i.e. the mechanical losses, at low temperatures of molybdenum samples. In the present work a more detailed investigation of the attenuations of longitudinal waves at frequencies of 5, 10 and 28 MHz in polycrystalline samples and single crystals with different crystallographic orientations was studied in successively deformed and also successively γ-ray- and neutron-irradiated molybdenum specimens.

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2. Experimental procedures

Three single crystals of orientations ⟨111⟩ (specimen 1), ⟨110⟩ (specimen 2) and ⟨100⟩ (specimen 3) of purity 99.995% (supplied by the Good-fellow Company) were used. The specimens were in the cylindrical form and were 100 mm long and 15 mm in diameter. They were cut from the original specimens with a low-speed diamond saw. The specimens were annealed at 1000 °C for 1 h in an argon atmosphere. Three specimens were deformed at room temperature by giving them a compressional stress of 20 MPa using a Monsanto tensometer.

The other three specimens were polycrystalline and also of purity 99.995%. The first polycrystal specimen (specimen 4) was given successive deformation at room temperature to a maximum stress of nearly 30 MPa. The other polycrystalline specimens (specimens 5 and 6) were deformed at room temperature using a force of 20 kN which resulted in a deformation of 1%. Then one specimen (specimen 5) was given successive γ-ray doses using ^{60}Co of energy 1.3 MeV at room temperature and after an interval of 3 days the ultrasonic attenuation was measured. Specimen 6 was given successive neutron doses using a neutron source (radium) which is capable of providing fast neutrons with a continuous energy spectrum extending to 10 MeV and emission of 10^5 neutrons s^{-1}. The specimen was also left for 3 days at room temperature after each successive neutron irradiation.

A conventional ultrasonic pulse technique (commercial) was used to study the effect of different applied frequencies of measurements on the α-complex peaks. Further details about the equipment and ultrasonic measurement have been given in an earlier paper by Zein and Alnaser [6].

For frequencies used in the present work the damping was calculated using the following relation:

$$Q^{-1} = 0.036\alpha c/f$$

where α is the ultrasonic attenuation in decibels per centimetre, c is the speed of sound in centimetres per second and f is the frequency of the measurement in hertz.

3. Single-crystal measurements

3.1. Effect of crystal orientation

Figures 1(a), 1(b) and 1(c) show the variations in the relaxation peak temperature and the heights for the three molybdenum specimens having orientations ⟨111⟩, ⟨110⟩ and ⟨100⟩, i.e. specimens 1, 2 and 3, respectively, and measured at three different frequencies (5, 10 and 28 MHz). The peak temperatures of α_1, α_2 and α_3 at 10 MHz occur at 157, 178 and 214 K, respectively, for nearly all the crystallographic directions. The three relaxation peaks observed which are believed to be the α complex seem to be orientation independent, similar to FCC metals. This observation agrees with other work [7] but does not agree with that of Mongy [4] who reported an appreciable anisotropy. This may be due to the type of sample used by Mongy which contains all three orientations. The relaxation process observed by Bordoni [8] at low temperatures in FCC metals is known to be attributed to the oscillatory motion of the edge component of dislocations on their slip planes under the influence of the periodic stress field. Obviously, the relaxation mechanisms does not change when it occurs on the close-packed plane in any one direction.

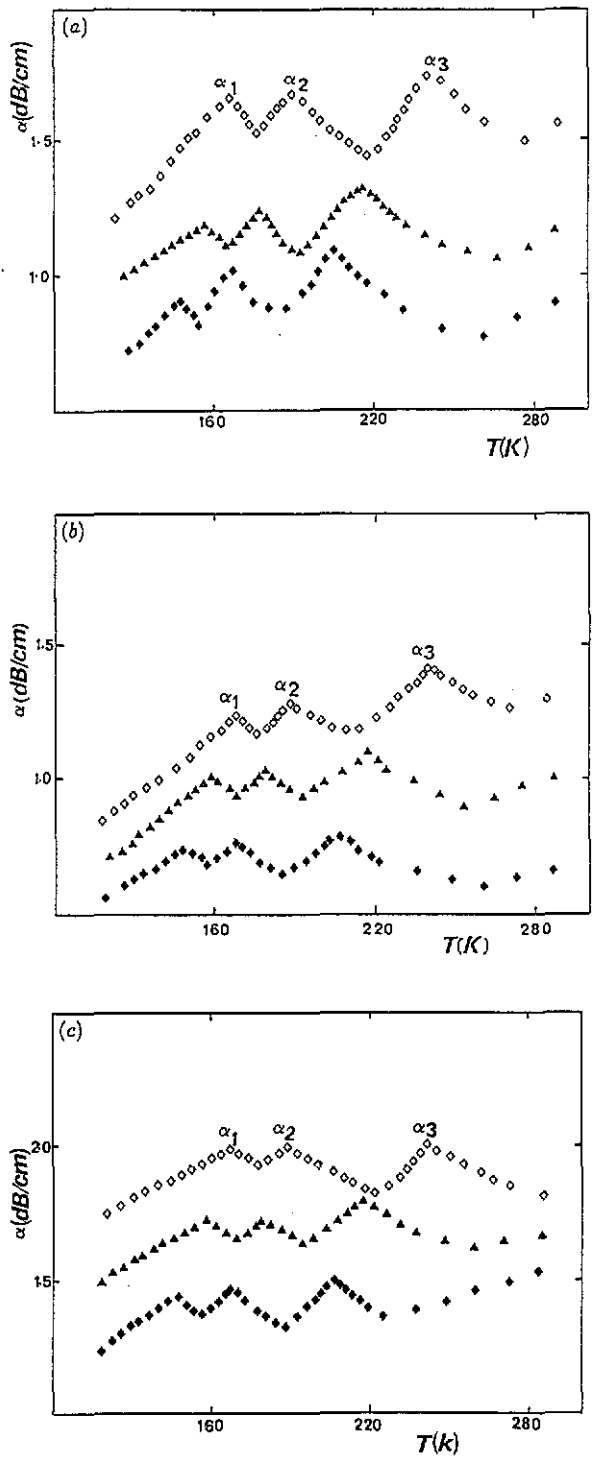

Figure 1. Ultrasonic attenuation at frequencies of 5 MHz (♦), 10 MHz (▲) and 28 MHz (◊) for three Mo single crystals as a function of temperature following deformation at a compressive stress of 20 MPa at room temperature along (*a*) (111), (*b*) (110) and (*c*) (100).

3.2. Frequency dependence

Figure 1 also shows that the peaks occur at 148, 167 and 206 K when the measurement

is carried out at a frequency of 5 MHz. These peaks shifted to 157, 178 and 214 K when measured at a frequency of 10 MHz and increased to 168, 189 and 240 K when the measurement is carried out at a frequency of 28 MHz. Using these data we were able to calculate the activation enthalpies and the attempt frequencies for α_1, α_2 and α_3 which were found to be equal to 0.18 eV, 0.22 eV and 0.20 eV and to 9.2×10^{12} Hz, 1.8×10^{13} Hz and 5×10^{11} Hz, respectively, with a correlation coefficient r of -0.992 for α_1, -0.995 for α_2 and -0.973 for α_3. Using the activation parameters given by Grau and Schultz [9] (for comparison) the temperature at which their peaks would have occurred at 10 MHz is in agreement with the present work for α_1, α_2 and α_3. It should be noted that the order of the α peaks was changed in I and the nomenclature of I is used here throughout so that the lowest-temperature peak was α_1 while in the work of Grau and Schultz it is α_3. Peak α_4 in their work [9] would be at too low a temperature (48 K) to be seen in this work. Table 1 shows the results (the results of Polotskii *et al* [5] are also shown in the table).

Table 1. Comparison of peak temperatures at 10 MHz in molybdenum specimens.

Reference	Strain (%)	Peak temperature (K) α_1	α_2	α_3
Polotskii *et al* [5]	3–4	100 (very weak)	185	270 (?)
Grau and Schultz [9]	1	156	169	210
Present work	1	143	167	205

The activation enthalpy was found to vary from 0.14 to 0.19 eV for α_2 [10] and the corresponding values of the attempt frequency varied from 1.6×10^9 to 4×10^{11} Hz. Polotskii *et al* [5] reported the activation parameters for the α_2 peak as 0.19 eV and 10^{12} Hz, respectively. These values are nearly in agreement with our findings, bearing in mind the slight variations in the amount of plastic deformation given to the sample, the thermomechanical treatment and the purity of the specimen used in their work.

The height of the relaxation peaks α_1, α_2 and α_3 were found in general to increase as the frequency increased in all the three crystallographic directions. This is similar to what has been found for FCC metals [11] and to what was theoretically reported [12, 13].

4. Polycrystalline measurements

4.1. Stress dependence

Figure 2 shows the ultrasonic attenuation in polycrystalline molybdenum (specimen 4) given different amounts of strain ranging from 0.024 to 0.99% (i.e. stresses from 0.2 to 29 MPa). Three peaks were found, namely α_1, α_2 and α_3, which occur in the temperature ranges 164–143 K, 190–167 K and 228–205 K, respectively, when the measurements were carried out at 10 MHz. There is a marked change in the temperature and the height of the peaks for very low strains (below the yield stress in the vicinity of 10 MPa) followed by a very low change up to the maximum strain of 1%. It is likely that the initial change is associated with dislocations freeing themselves from impurities. A similar effect was seen by Garcia *et al* [14]. An increase in the dislocation density is expected at higher deformations which will result in a slight change in the peak temperature.

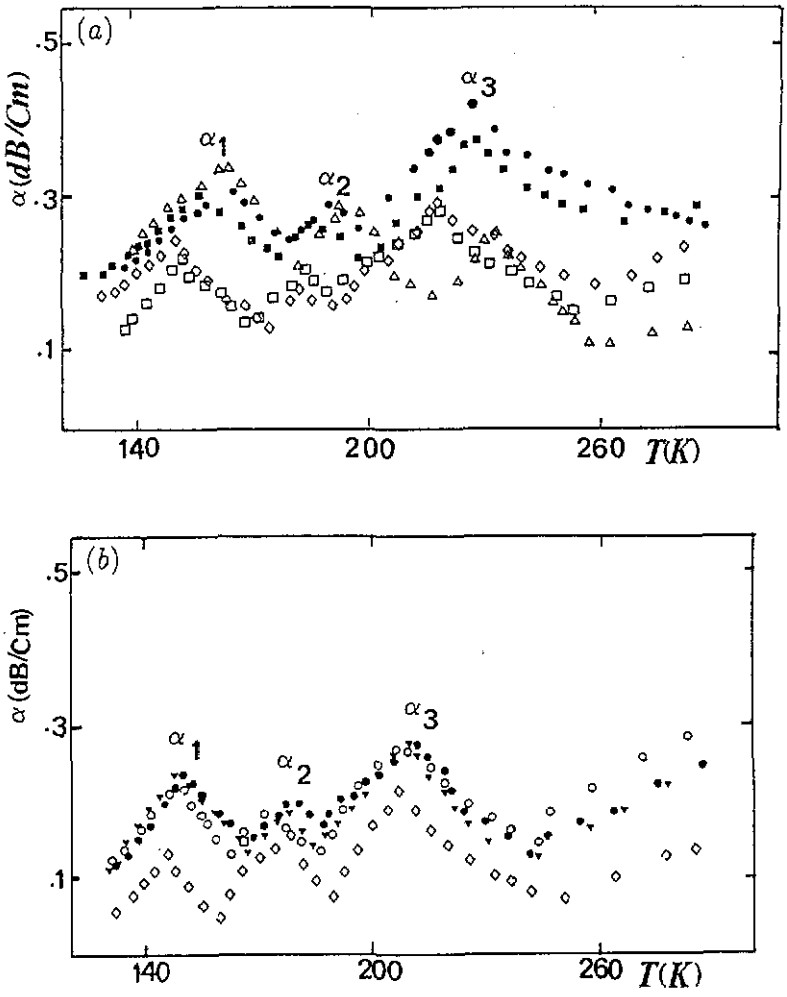

Figure 2. Ultrasonic attenuation at 10 MHz of a polycrystalline Mo sample (sample 4) as a function of temperature following successive deformations with the following strains: (*a*) ●, 0.024%; ■, 0.030%; △, 0.034%; □, 0.040%; ◇, 0.050%; (*b*) ●, 0.18%; ▼, 0.25%; ○, 0.40%; △, 0.99%.

4.2. Effect of irradiation

Figures 3 and 4 show the variation in the peak heights and temperatures of α_1, α_2 and α_3 in polycrystalline molybdenum irradiated by γ-rays (specimen 5) and neutrons (specimen 6), respectively. The peak height decreases very dramatically on irradiation. Also, we found that irradiation noticeably reduced the peak temperature of α_1 and α_3 but the result is not very clear for α_2. This could be attributed to the type of dislocation responsible for each relaxation peak. Seeger and Wuthrich [15] first suggested that more than one type of kink can occur in dislocations and this may account for the number of peaks observed in BCC metals. Garcia *et al* [16] suggested that α_3 and α_2 arise from kink pair formation on non-screw dislocations while α_1 arises from geometric kink motion on screws. Vesely [17] and Loesch and Brotzen [18] reported that at low strains the dislocation configuration in the crystal deformed at low temperatures (156 K) produces dislocation structures which

are predominantly screw dislocations while high-temperature deformation (450 K) produces predominantly edge dislocation structures. Since the deformations of our specimens were carried out at room temperature (300 K) then the existence of the suggested types of dislocation responsible for the α complex is much more feasible. Rieu [19] showed by electron microscopy that, for all orientations, non-screw dislocations are dominant at higher strains in single crystals. This explains why the lowest-temperature peak is so weak in the work of Polotskii *et al* [5] (their samples were strained by over 3%).

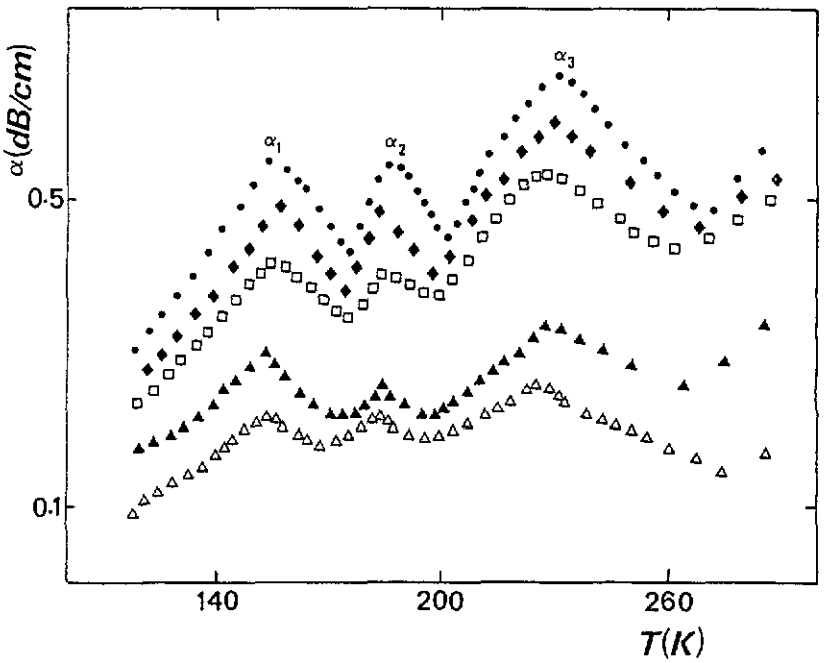

Figure 3. The effect of γ irradiation doses on the ultrasonic attenuation at 10 MHz in polycrystalline Mo (specimen 5): ●, before irradiation; ♦, 2.15×10^{18} γvt; □, 4.28×10^{8} γvt; ▲, 6.45×10^{8} γvt; △, 8.6×10^{8} γvt.

Figures 5 and 6 are plots of $\log Q_{\max}^{-1}$ versus $\log D_\gamma$ and $\log D_n$ (D_γ is the gamma dose and D_n is the neutron dose). The slope for the α_1 peak is 0.42, for α_2 it is 0.70 and for α_3 it is 0.47 for γ irradiation. For neutron irradiation the slope is 0.28 for α_1, 0.3 for α_2 and 0.33 for α_3. This suggests that $Q_{\max}^{-1} \propto \Lambda D_{\gamma,n}^{-n}$ where Λ is the dislocation density and n is the value of the slope for each peak. However, since $D_{\gamma n}$ is proportional to the concentration C of the defects and on the assumption that L (the free loop length) is related to C by $L \propto C^{-n}$, then $Q_{\max}^{-1} \propto \Lambda L^n$. This relation shows that α_2 is the most sensitive peak to irradiation, followed by α_3 and α_1.

Since the interaction energy of a defect with screw dislocations is less than its interaction with an edge dislocation [20] and since the activation enthalpy of α_2 (0.22 eV) is higher than that of α_3 (0.20 eV), this leads us to conclude that α_2 may be attributed to kink pair formation in long dislocations probably with a Burgers vector at 70° with respect to the ⟨111⟩ direction on ⟨110⟩ planes, while α_3 may be due to kink pair formation in short dislocations, or probably due to dislocations at larger angles than 70° with respect to ⟨111⟩.

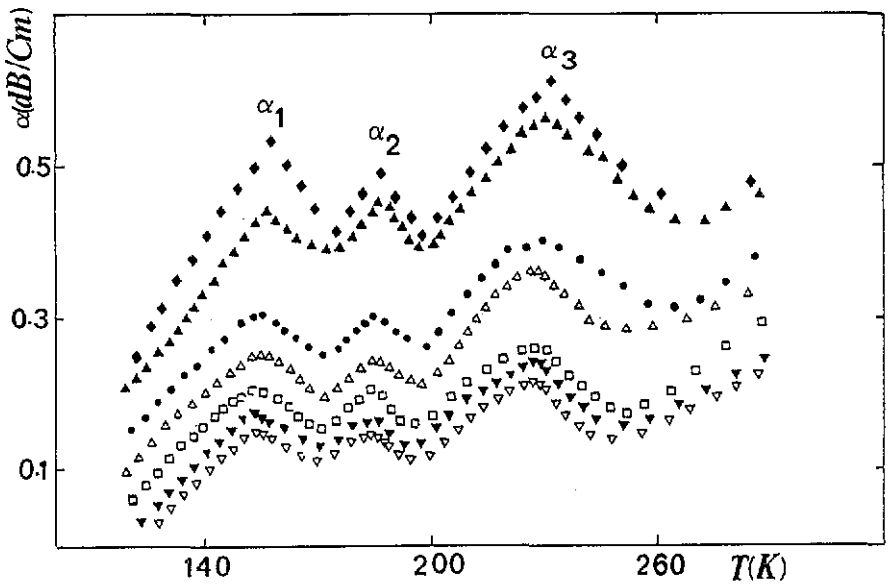

Figure 4. The effect of neutron irradiation doses on the ultrasonic attenuation at 10 MHz in polycrystalline Mo (specimen 6): ♦, before irradiation; ▲, 1.3×10^9 nvt; □, 3.6×10^9 nvt; △, 5.4×10^9 nvt; ●, 7.2×10^9 nvt; ▼, 9.0×10^9 nvt; ▽, 10.8×10^9 nvt.

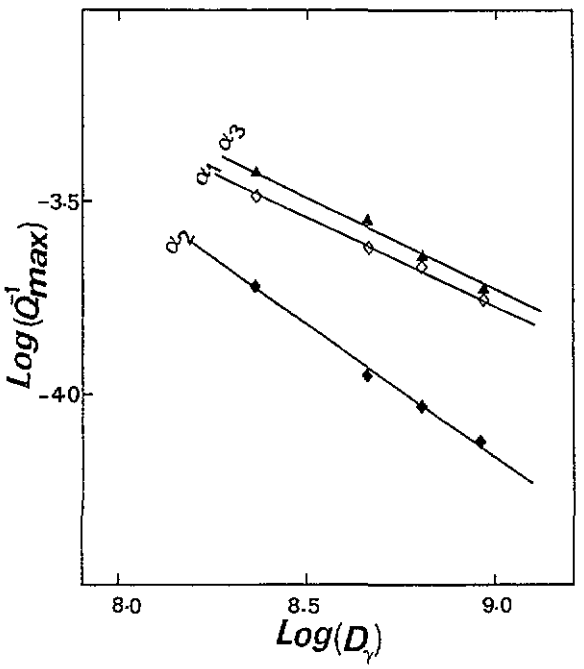

Figure 5. Plot of the logarithm of the relaxation strength versus the logarithm of the γ-ray dose.

It has to be mentioned that γ irradiation was found to pin the dislocations in molybdenum more than the neutrons do.

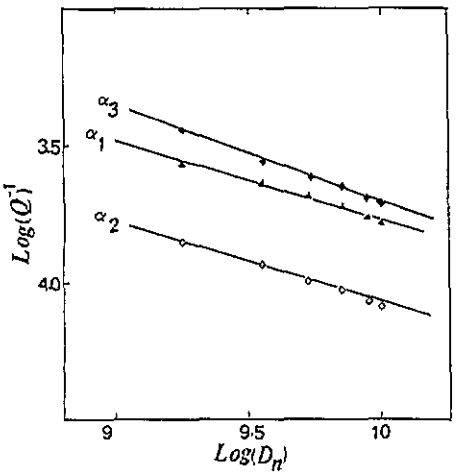

Figure 6. Plot of the logarithm of the relaxation strength versus the logarithm of the neutron dose.

5. Background damping

It was reported that the dislocations that are not parallel to the close-packed direction do not contribute to dislocation relaxation but contribute to the background damping [15]. We have found that the highest background damping was in samples of ⟨100⟩ orientation followed by ⟨111⟩ and ⟨110⟩. This was found at all frequencies of measurement, i.e. 5, 10 and 28 MHz.

When the polycrystalline data are compared with the single-crystal data, the damping of the polycrystal is found to be considerably lower than the damping of the single crystals. This may be attributed to the role of the dislocations introduced in these two types of sample. In fact, the plastic deformation may be expected to have two main effects:

(i) an increase in the dislocation density through the operation of dislocation sources;
(ii) the production of point defects through dislocation interaction.

When the amount of deformation is small (see figure 2), the first effect will predominate and one would expect an increase in the attenuation. However, as the amount of deformation increases, a point will be reached where the shortening of the dislocation loop lengths because of pinning by point defects will become an important factor, and accordingly the attenuation will decrease. According to the theory of Granato and Lücke [21] the amplitude-independent decrement δ_I for frequencies well below that for which the decrement is a maximum, and hence the background damping, has the following proportionality:

$$\delta_I \propto l^4 \Lambda f B$$

where l is the average loop length, Λ is the dislocation density, f is the frequency of measurement and B is the damping force. We therefore may conclude that interaction between dislocations is the main reason why the background damping in polycrystalline samples is less than that in the single crystals, i.e. the polycrystals have a shorter loop length than single crystals have and may also have larger dislocation densities. According to Seeger *et al* [22], the background damping is associated with the energy required to form a pair of kinks. The activation enthalpy for the background damping is estimated to be equal to half that required for forming a pair of kinks; hence the damping is proportional

to the number of kinks present in thermal equilibrium. Since the relaxation peaks occur at the same temperature in the polycrystalline samples and the single crystals, i.e. at similar values of activation enthalpy, then the assumption of Seeger *et al* may be valid.

One further possible explanation for this observation, i.e. the fact that the background attenuation in a polycrystal is less than that in a single crystal, is as follows. Schulz and Lenz [23] introduced the concept of attenuation α_{ND} due to non-dislocations and it was studied further by Schmidt *et al* [24]. They noticed that, at a strain ϵ of more than 1%, α_{ND} increases until ϵ reaches 13% where α_{ND} levels off. They attributed this non-dislocation background (which causes the increase in the apparent attenuation) to deformation-induced inhomogeneities in the lattice, such as kink bands (as was proved by metallographic analysis). Honeycombe [25], Cahn [26] and Calnan [27] found that this effect occurs in either single-crystal or polycrystal aggregates and stated that these kink bands cause a curvature in the lattice. As a result of this effect the sound wave will be scattered as a result of deformation since it induces lattice inhomogeneities and therefore an increase in the apparent attenuation will be recorded. These may lead us to suggest that kink bands are more significant in deformed single crystals than in polycrystals.

It is worth mentioning that quartz-sample deformation (QSD) may have an important effect in calculating the peak height and the background damping. The QSD arises from the difference between the thermal expansions of the quartz crystal and the sample. This difference will act as a stress applied to the sample when the bond between the quartz crystal (transducer) and the sample becomes solid and a maximum is sometimes observed at 140–170 K [23], i.e. in the vicinity of peak α_1. This defect is absent after straining a copper sample by 0.3%. Kaufmann *et al* [28] found that the true temperature dependence of B (dislocation drag constant) is masked by QSD, which is difficult to avoid. We therefore intended to give a deformation of 1% to reduce the QSD effect and to minimize the errors in estimating the peak heights.

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