Low-temperature ultrasonic attenuation measurement in very pure gold polycrystals at megahertz frequencies

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Low-temperature ultrasonic attenuation measurement in very pure gold polycrystals at megahertz frequencies

W E Alnaser

Deanship of Scientific Research, University of Bahrain, PO Box 32038, Bahrain

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Abstract. The ultrasonic attenuation in a polycrystal gold crystal of purity 99.999% has been measured at three different frequencies, 5, 10, and 30 MHz, from 77 to 300 K. Mainly one peak (the dislocation relaxation peak, or Bordoni peak) was observed at 178–174, 186–185, and 237–235 K at 5, 10, and 30 MHz respectively. The highest peak temperature observed was for samples given a low stress (8 MPa) while the lowest was for 130 MPa stress. On further increasing the stress to 259 MPa, the peak temperature increases slightly.

The present work shows that the peak height decreases as the frequency of measurement increases while it increases as the mechanical stress increases. The activation energy and the attempt frequency decrease as the compression deformation to the sample increases.

1. Introduction

Absorption peaks associated with Bordoni [1] are observed by examining ultrasonic attenuation in deformed metals in the temperature range between liquid-nitrogen and room temperature at megahertz frequencies. The absorption mechanism is attributed to the formation of pairs of dislocation kinks under the combined influence of thermal fluctuations and applied stress. The absorption peaks occur when the applied oscillating shear stress is equal [2, 3] to the frequency of formation of kink pairs. These absorption peaks are called either internal friction peaks or Bordoni peaks or mechanical loss peaks. The positions of these peaks depend upon the range of frequency of measurement and purity and deformation of the sample under test. The principal features of these peaks have been reviewed by a number of authors [2–7]. Kink-pair formation theory and its modification [8–14] is the most realistic interpretation for these peaks.

According to Schulz and Lenz [15], in well annealed very pure crystals ($\leqslant 10^2$ ppm impurities) at frequencies f below the resonance frequency f_{max} ($f_{max} \approx 100$ MHz) the Granato–Lucke damping [16] (dislocation resonance—DR) is given by

$$\delta_{GL} = 0.115\alpha_{GL}(\mathrm{d}B\ \mu\mathrm{s}^{-1})/f\ (\mathrm{MHz}) \sim B\Lambda L^4 f. \qquad (1)$$

α_{GL} is the attenuation due to Granato–Lucke damping, B is the damping force factor, Λ is the dislocation density, and L is the loop length between pinning points. At a temperature higher than 50 K, the damping force factor increases linearly. Plastic deformation (ε) affects the dislocation density, therefore resonance damping is expected to increase ($\alpha_{GL} \sim \varepsilon$). Schulz and Lenz added that any decrease of loop length L (due to mutual intersection of dislocations or pinning of dislocation by deformation-induced pinning points) will counteract the effect of increasing Λ on α_{GL}. On the other hand the deformation (i.e. the applied stress introduced by compressing the metal at room temperature) results in dislocation Bordoni

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relaxation (Bo) which is attributed to thermally activated kink-pair generation—according to the following relation [8, 9]:

$$\delta_{Bo} \sim \Lambda L^x D(\omega\tau). \quad (2)$$

Here $1 \leqslant x \leqslant 2$ and $D(\omega\tau)$ is a Debye type function depending on frequency and relaxation time $\tau = \tau_0 \exp^{W/kT}$ (W is the activation energy of the relaxation process, k is the Boltzmann, constant, T is the temperature, and τ_0 is the attack frequency).

In this paper, the effect of the applied stress (8–260 MPa) on the Bordoni peak strength and its temperature (as well as the background damping) will be studied at three different very high frequencies of measurement (5, 10 and 30 MHz).

No work has been previously reported on ultrasonic attenuation in a polycrystalline gold specimen of a very high purity subjected to a wide range of applied compressional mechanical stresses at these high frequencies (5, 10 and 30 MHz). We can only recall here the work of Grandchamp [17], Bordoni *et al* [18] and Okuda [19] who briefly studied gold specimens.

2. Experimental procedure

A gold polycrystalline specimen of purity 5 N was ordered from Good Fellow Company, UK. The specimen was cut to a length of 10 mm and a diameter of 13 mm with parallelism greater than 99.5%. The specimens were first annealed for 10 h in argon atmosphere at a temperature of 500 °C. Successive compressional stresses were then applied using a Monsanto tensometer at room temperature. After each deformation, the sample were left for 3 d to eliminate the Koster effect [20]. The temperature was measured using two platinum sensors placed close to the specimen and connected to a digital thermometer. The measurements were made with a linear warm-up rate of 0.7 K $\min^{-1}$.

In order to convert the short ultrasonic electrical pulses (1.5 μs) to longitudinal waves in the sample, an X-cut gold-plated quartz crystal (diameter 6 mm) was used. It was coupled with the gold specimen using a very thin layer of Nonaq stopcock grease. The combination of this sample–transducer coupling is believed to give minimum ultrasonic attenuation due to diffraction, non-parallelism, and bonding losses [18]. A single-ended pulse echo technique was used. The attenuation was measured from the average ratio of the amplitude of echoes which were displayed in the oscilloscope after amplification. The maximum strain amplitude for this technique does not exceed 10^{-7}. The damping Q^{-1} ($= \delta/\pi$), δ is the decrement, was calculated by substituting the values of the ultrasonic attenuation, α (dB cm^{-1}), sound speed in gold, v (cm s^{-1}), and frequency of measurement, f (s^{-1}), in the relation

$$Q^{-1} = 0.036\alpha v/f. \quad (3)$$

To calculate the exact value for the peak height (due to the dislocation relaxation alone), the total attenuation α is assumed to be given by

$$\alpha = \alpha_{Bo}(T) + \alpha_{GL}(T) + \alpha_{QSD}(T) + \alpha_d + \alpha_{np} + \alpha_{nd} + \alpha_{bl}. \quad (4)$$

The total attenuation is a combination of temperature dependent and temperature independent attenuation. In equation (2), α_{Bo} is the attenuation due to Bordoni peak (dislocation relaxation strength), α_{GL} the attenuation due to Granato–Lucke damping [16], and α_{QSD} is the attenuation due to quartz–sample deformation (QSD) [20, 21], which is nearly absent at deformation of more than 1% and mainly occurs close to the temperature of solidification of the acoustic bond [22]. This QSD arises from the difference in the thermal expansion of the quartz crystal and the sample. This difference will act as a stress applied to the sample when the bond between the quartz (transducer) and the specimen becomes solid. Maxima usually occur at 140–170 K. α_d, α_{np} and α_{bl} are attenuation due to diffraction loss, non-parallelism, and bonding losses respectively. α_{nd} is attenuation due to non-dislocation. This was first reported by Schulz and Lenz [15]. At strain $\varepsilon > 1\%$, α_{nd} increases until ε reaches 13% (in copper), where it then levels off. α_{nd} is attributed to the non-dislocation background (which causes the increase of the apparent attenuation) because deformation induces inhomogenates in the lattice such as kink bands which occur in both single crystals and polycrystals.

The model established by Alnaser [23] to analyse the background component of the attenuation is used here to calculate the peak height more accurately.

3. Results and discussion

Table 1 shows the variation of the dislocation relaxation strength (after subtracting all the background losses using Alnaser's model [23]. For comparison, the ultrasonic attenuation as a function of temperature for a gold specimen subjected to successive amounts of deformation (using a Monsanto tensometer) at room temperature, i.e. stress from 8 MPa to nearly 260 MPa and measured at 5, 10 and 30 MHz, is presented in figure 1(a)–(c) respectively. Only one broad peak (Bordoni peak) was found (the subsidiary peak, i.e. the Niblett–Wilks peak, was absent). Similar results were observed when studying the dislocation relaxation peak in silver [24] (since both gold and silver have high stacking fault energy). The peak occurs in the temperature ranges of 178–168 K at 5 MHz frequency, 186–182 K at 10 MHz frequency, and 237–232 K at 30 MHz frequency. This proves that the peak temperature is frequency dependent. Using the Arrhenius method to calculate the activation energy W, we found that $W = 0.154$ eV and the attempt frequency $f_0 = 2 \times 10^9$ Hz, i.e.

$$f = 2 \times 10^9 \exp\left(-\frac{0.154}{kT_m}\right) \tag{5}$$

where f is the frequency of measurement, k is the Boltzmann constant (eV), and T_m is the temperature (K). The value calculated in the present work is in agreement with those quoted in the literature [25], taking into consideration the slight thermomechanical treatment and purity variation of our sample compared with others.

The shift of the peak temperature with frequency is explained by Seeger and Schillen [2, 8, 9] as the dislocation line being forced over the Peierls potential hills by the creation of a pair of kinks in a thermally activated process.

The reason why only one peak is found (and not the subsidiary) may be either lack of dislocations with an angle of 60° between the Burger vector and the dislocation line or the other peak having smaller magnitude compared to the background (other losses).

It is important to examine the dependence of the peak height (Q^{-1}_{max}) on frequency. According to Esnouf and Fantozzi [14], who modified Seeger's model [8], the relaxation strength should decrease as the frequency of measurement increases. We found that this is true. The relation between Q^{-1}_{max} and f is found to be as follows:

$$Q^{-1}_{max} = 1.27 \times 10^{-2}(1 - 0.056\ell n f). \tag{6}$$

This is similar to the result reported by Esnouf and Fantozzi [14], i.e. $\delta = \delta_1(1 - 0.04\ell n f)$, where δ is the decrement ($\delta = Q^{-1}\pi$) and δ_1 is the decrement at 1 Hz.

Furthermore, an interesting observation was that as the mechanical stress (σ) increased from 8 to 259 MPa the peak temperature decreased up to a point (192 MPa) beyond which it nearly levelled off or slightly increased (>192 MPa). This variation was in qualitative

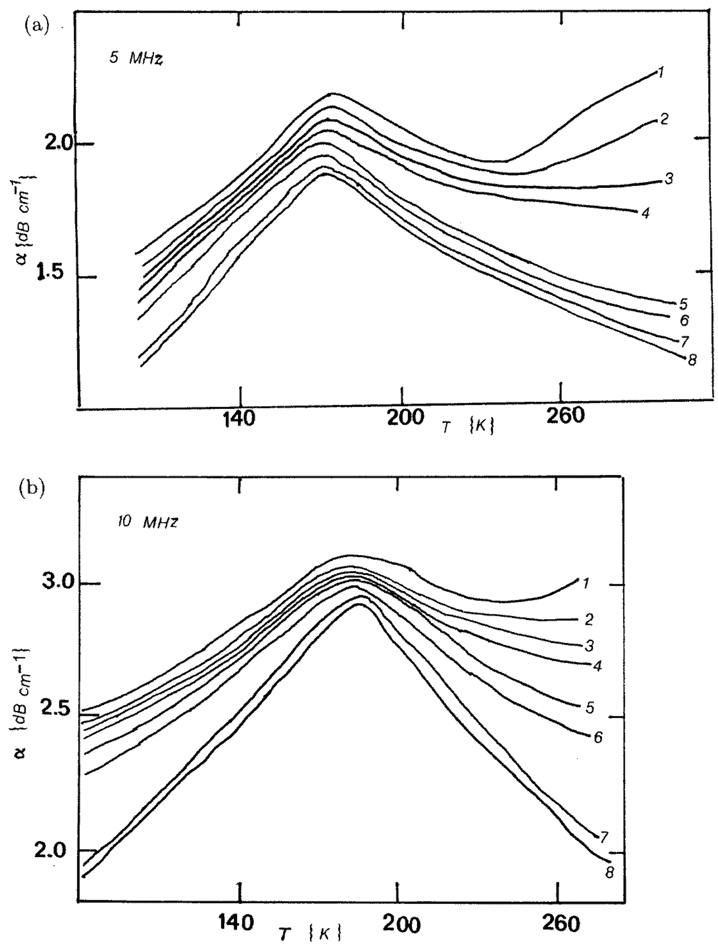

Figure 1. The ultrasonic attenuation as a function of temperature at (a) 5 MHz, (b) 10 MHz, and (c) 30 MHz frequency in very pure polycrystalline gold subjected to successive stresses: 1, 8 MPa; 2, 32 MPa; 3, 65 MPa; 4, 91 MPa; 5, 130 MPa; 6, 192 MPa; 7, 233 MPa; 8, 259 MPa.

agreement with Seeger's theory of pair-kink generation [2, 8] and its modification [10, 12] taking into account the different internal stresses and dislocation loop length present at different deformations. When the sample was lightly deformed, longer dislocations (ℓ) are present and higher Q_{max}^{-1} was expected. According to the theory (refer to equation (2)) Q_{max}^{-1} was proportional to $\Lambda\ell^2$; Λ is the dislocation density. At high stresses, the internal stress decreases because of formation of a cell structure in the specimen in which the majority of dislocations are located in the cell walls.

Fantozzi *et al* [26] suggested that the peak temperature T_{max} decreases when pinning (or shortening) the dislocation loop. Therefore, increasing stress causes initially long dislocations but then with increasing dislocation density Λ dislocations become tangled, causing shorter dislocations. For high applied stresses, cell structures are formed producing again long dislocations inside them. This is the case for applied stress of more than 192 MPa where the peak temperature begins to increase again. It is the combination of Λ and ℓ and which one is more significant than the other that determine the variation of T_m and $Q_{\rm max}^{-1}$

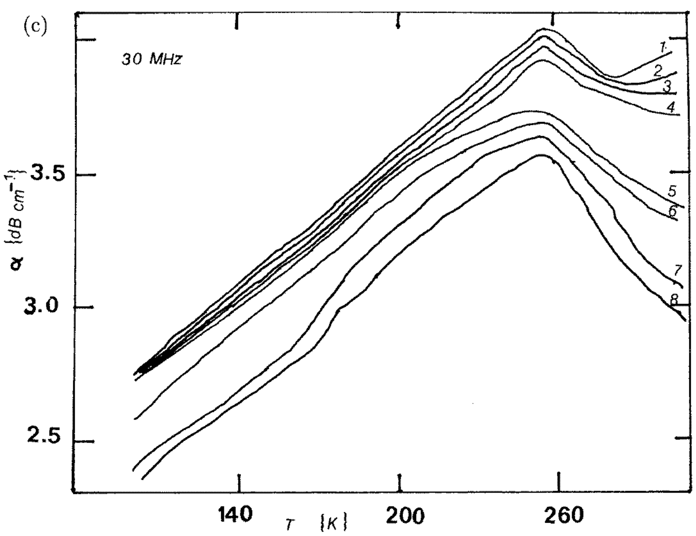

Figure 1. (Continued)

Table 1. The variation of the Bordoni peak height and temperature in very pure polycrystalline gold at different megahertz frequencies.

σ	5 MHz		10 MHz		30 MHz	
(MPa)	T_m (K)	$Q^{-1} \times 10^4$	T_m (K)	$Q^{-1} \times 10^4$	T_m (K)	$Q^{-1} \times 10^4$
8	178	9.98	186	3.50	237	1.46
32	172	10.50	184	5.40	236	1.65
65	170	12.20	183	6.13	234	2.00
91	168	13.20	182	7.05	233	2.16
130	172	14.96	182	7.60	232	2.26
192	173	15.30	183	9.38	233	2.46
233	174	16.63	185	11.52	235	2.77
259	174	20.20	185	12.52	235	2.81

with the applied stress.

Since the peak temperature is related to the activation energy W and W is related to stress (σ), W is expected to decrease as σ increases. This has been found: for $\sigma = 8$ MPa, W was 0.16 eV, which shifted to 0.10 eV for 130 MPa. The attempted frequency also shifted to lower values as σ increased. For lower stresses f_0 was 2×10^9 Hz and for higher values it was equal to 1.2×10^9 Hz. Grandchamp [17] reported $W = 0.20$ eV, $f_0 = 8.4 \times 10^{12}$ Hz while Okuda [19] reported $W = 0.19$ eV and $f_0 = 10^{13}$ Hz and Bordoni *et al* [18] reported $W = 0.16$ eV and $f_0 = 7 \times 10^{10}$ Hz. Last, the background damping increases, which supports (1).

It is very interesting to note that the background attenuation, which is mainly Granato–Lucke damping, decreases substantially as the applied stress increases since larger stress causes the shortening of L, i.e. $\alpha_{GL} \propto L^4$. Also for a particular applied stress the attenuation (and hence the damping) increases as the frequency of measurement increases, i.e. $\alpha_{GL} \propto f$. These two observations are in support of (1).

4. Conclusion

One main, relatively broad, dislocation relaxation peak is found in studying the ultrasonic attenuation at low temperatures in gold polycrystal specimens having high purity subjected to different plastic deformations at room temperature and measured at megahertz frequencies.

The peak temperature shifts to a higher value as the frequency of measurement increases while the peak height decreases. The peak temperature decreases as the applied stress to the sample increases, while the peak height continues to increase. The experimental result is in full agreement with Seeger's model [2, 8] for explaining the dislocation relaxation peak.

The behaviour of the background attenuation with the applied stress and the frequency of measurement are in agreement with the Granato–Lucke theory [16].

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