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INFLUENCE OF PLASTIC DEFORMATION ON THE TEMPERATURE DEPENDENCE OF ULTRASONIC ATTENUATION IN COPPER SINGLE CRYSTALS

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Abstract.- The influence of plastic deformation (0<ε<10%) on the MHz-ultrasonic attenuation a in single crystals of pure Cu, Cu50ppmAu and Cu130ppmAu has been measured over the temperature range 4<T<300K. The major contributions to the total attenuation originate from dislocation resonance, double-kink relaxation, phonon-electron interaction (at T<50K) and non-dislocation effects which are discussed especially with respect to the separation of dislocation resonance and dislocation relaxation contributions. For the activation quantities of Bordini relaxation we obtain Q = 0.105 ± 0.005 eV, τo = (1.2 ± 0.4) x 10⁻¹²s.

1. Introduction.- Ultrasonic attenuation in well annealed pure (typically ≤ 10² ppm impurities) fcc single crystals is governed by overdamped dislocation resonance (DR) as described by the Granato-Lücke (GL) theory /1/. At frequencies f below the resonance frequency fMAX (about 100 MHz for the present samples) this damping is given by

\[ \delta_{GL} = 0.115 \alpha_{GL}[\text{dB/\mu s}]/f[\text{MHz}] = B\Lambda L^4 f \]

(\(\delta_{GL}\) = DR decrement, \(\alpha_{GL}\) = DR attenuation, B = damping force factor, \(\Lambda\) = dislocation density, L = loop length between pinning points). Since B decreases linear with temperature T at T >~ 50K, becoming constant at T < 50K /2/ and f MAX ~ 1/B equ.(1) holds for low temperatures and a linear decrease of \(\alpha_{GL}\) with decreasing T is observed /3/. Since plastic deformation \(\varepsilon\) increases \(\Lambda (\Lambda^{-\varepsilon}/4)\) resonance damping is expected to increase (\(\alpha_{GL}^{-\varepsilon}\)). However, any decrease of loop length L (e.g. due to mutual intersection of dislocations or pinning of dislocation by deformation induced pinning points) will counteract the effect of increasing \(\Lambda\) on \(\alpha_{GL}\). Furthermore deformation results in dislocation Bordini relaxation (BR) attributed to thermally activated double-kink generation /5,6/

\[ \delta_{BO} \sim \Lambda L^N D(\omega \tau) \]

with 1≤N≤2 /7/ and D(\(\omega \tau\)) a Debye type function depending on frequency and relaxation time \(\tau = \tau_o \exp (Q/kT)\). Present knowledge about BR mainly results from Hz- and kHz experiments, i.e. from frequencies well below the typical DR-range. Accordingly \(\delta_{GL}\) contributions are usually not considered (see however /8/). There is only a relatively small number.
of MHz-investigations of BR in Cu/9,10,11,16/. To our knowledge the contributions of BR and DR respectively to the total dislocation damping have not yet been clarified quantitatively.

2. Experimental.- We measured attenuation \( a \) (10 to 50 MHz) in \( <11\bar{1}> \)-oriented crystals of pure Cu (\( \leq 5 \) ppm impurities), Cu+50ppmAu, and Cu+130ppmAu by the pulse-echo technique between 4.2 and 300K after compressional deformations of \( 0 \leq \varepsilon \leq 10\% \) along \( <11\bar{1}> \). After deformation the samples were annealed 1h at 363K to eliminate the deformation produced point defects. The residual resistivity ratio RRR was measured on each sample by the eddy current technique /12/. The x-cut quartz transducers (0.25"Ø) were bonded to the specimens by Nonaq (and in several cases by 3-methyl-pentan (3MP)).

3. Results and evaluation.- Fig.1 introduces the observed phenomena: Point A shows \( a \) before, point B after 0.99\% deformation and 1 h 90°C. The \( a(T) \) curves are to be separated into three dislocation contributions \( a_D = a_{GL} + a_{BoII} + a_{QSD} \) and the non-dislocation background \( a_B \) (plus the phonon-electron effect \( a_{PE} \) at low \( T \)). The background is measured after complete dislocation pinning by \( \gamma \)-irradiation at RT. We use the \( a_B(\varepsilon) \) values obtained by Drescher et al /13/ together with the linear \( a_B(T) \)-dependence measured by Kaufmann /14/. The \( a_{PE}(T) \) contribution is calculated from the RRR of our samples with help of Pippards theory /15/ (the dashed curve in fig.1 at \( T < 70K \) is obtained after subtraction of \( a_{PE}(T)/17/ \). The \( a(T) \) (solid curves, irreversible for cooling and warming) was measured with the Nonaq bond. The (dashed dotted) \( a(T) \) which is reversible and less steep below 200K was measured with the 3MP bond. The difference \( a_{QSD} \) between these \( a(T) \) curves is due to quartz-sample deformation (QSD) which results from different thermal expansion of transducer and sample /14/: At \( T < T_\eta \) (\( T_\eta = solidification \) temperature of Nonaq \( \approx 200\)K) the bond transmits QSD stresses into the sample which (due to dislocation break-away) cause irreversible L-changes and the \( a_{QSD} \)-effects. QSD is avoided with 3MP (\( T_\eta \geq 120\)K); however, the bonding can only be done with considerable effort at 240K. Also with the 3MP bond a peaked \( a(T) \) is measured (fig.1) showing two maxima \( a_{BoI} \) (at \( \approx 150\)K), \( a_{BoII} \) (at \( \approx 60\)K) which indicate the Bordini (BoI) and Niblett-Wilks (BoII) peaks. With Nonaq BoI is seen erroneously at slightly higher \( T \). The \( a_{BoI}(T) \) contributions are superim-
posed on the dislocation resonance contribution \( a_{GL}(T) \) given by the straight line. How do we derive \( a_{GL}(T) \)?

Fig. 2 shows \( \alpha(T) \) for Cu+50ppmAu measured with Nonaq immediately (45 min) after 0.3% deformation. The strongly increased \( a_D \) is seen again (c.f. fig.1) however, there are no peaks indicating Bordini or QSD contributions. Thus the observed \( a_D(T) \) is completely due to dislocation resonance: \( a-(a_{PE}+a_B) = a_{GL} \).

The nearly linear \( a_{GL}(T) \) behaviour is in agreement with \( B-T \) in equ. (1). For further evaluation we assume the linear \( a_{GL}(T) \) for all experiments. Avoiding the QSD disturbance we observed \( a_{BO} \) contributions only for \( \varepsilon > 0.3\% \) (c.f. figs.1,3,4).

After \( \varepsilon = 9.3\% \) (fig.3) \( a_{BO} \) is very pronounced. Assuming \( a_{BO} \approx 0 \) at 4.2K subtraction of \( a_{PE} \) yields \( a_{GL} \) at 4.2K and the dashed curve at higher T. To find the correct slope for \( a_{GL}(T) \) we consider high temperatures \( (T>T_{BO}) \) where \( \omega T_{BO} << 1 \). For varying slopes we logarithmically fit the resulting normalized BR contribution \( a^*_B \equiv a_B(T)/\hat{a}_B = [a(T)-(a_{GL}(T)+a_B)] / \text{MAX} \{ [a(T)-(a_{GL}(T)+a_B)] \} \) to the slope of the high temperature asymptote of \( D(\omega T_{BO}) \), (c.f. equ.(2)), determined by the activation energy \( Q \) for double kink generation. Using \( Q = 0.11 \text{ eV} /16/ \) the best fit obtained yields \( a^*_B(300\text{K}) = 0.2 \) (for 50 MHz, vertical bar at 300K in fig.3). Since \( a^*_B \sim f \) the BR contributions for 30

FIG 2 \( \alpha(T) \) after 0.3% deformation (unannealed sample)

FIG 3 \( \alpha(T) \) after \( \varepsilon = 9.3\% \)

FIG 4 \( \alpha(T) \) after \( \varepsilon = 2.8\% \)
and 10 MHz in fig.3 are readily obtained. Disregarding small variations found we use this $a_{BO}^*$ (300)-ratio for all deformations. The validity of this $a_{BO}^{*} \leftrightarrow a_{GL}$ separation is demonstrated by fig.5a. The obtained $a_{BO}^* - f$ dependence means that the peak height $\hat{a}_{BO}^*/f$ is independent of $f$, as must be the case for a relaxation process. The separation is further supported by the fact that $a_{GL}$ is nearly $- f^2$ as expected for DR at $f \ll f_{MAX}$ (equ.(1)). Fig.5b shows Arrhenius plots which for the doped samples yield $Q = 0.11 \pm 0.005$ eV, $\tau_0 = (9.4 \pm 0.5) 10^{-13}$ s independent of $\varepsilon$. Slightly different values ($Q = 0.10 \pm 0.005$ eV, $\tau_0 = (1.6 \pm 0.5) 10^{-12}$s) are derived for the pure samples. Comparison with a single Debeye peak shows that the measured BoI peak is broadened by the factor $\approx 2$.

4. Discussion.- Recently Nibblett reported on BR measured as function of $\varepsilon$ and crystal orientation /11/. Nibblett mentions a large background damping (not to be confused with the present $a_B$ and not further discussed in /11/) and observed (in contrast to the present results) effects already after $\varepsilon \approx 0.1\%$ deformation. However, his deformed samples were stored at RT for 3 days and then measured with Nonaq bonding. Under such conditions we also observe peaked $\alpha(T)$ curves which we, however, attribute to QSD effects: i.e. during RT "annealing" the fresh dislocations are slightly pinned by deformation produced defects which anneal out at the dislocations. Cooling below 200K results in QSD induced breakaway of the dislocations from these pinning points. This leads to a $\alpha(T)$ maximum at temperatures in the range of the BoI peak /14/ (c.f. also fig.1). Such QSD effects are observed after annealing (or longer RT storage) reducing L compared with the loop length immediately after deformation. Therefore $a_{QSD}$ is absent in freshly deformed samples (fig.2). Under the present annealing condition (1h,90°C) QSD effects are comparatively unimportant at higher deformations where $a_{BO}$ is dominating (fig.3,4) especially in doped crystals. We have carefully considered QSD after small $\varepsilon$ ($<1\%$) in pure Cu and repeated doubtful experiments with 3MP bond. We conclude that MHz-Bordoni relaxation becomes observable only for $\varepsilon \approx 1\%$. 

![FIG 5a](image1.png)

*Frequency dependence of Bordoni peak height (a) and Arrhenius plots which for the doped samples yield $Q = 0.11 \pm 0.005$ eV, $\tau_0 = (9.4 \pm 0.5) 10^{-13}$ s independent of $\varepsilon$. Slightly different values ($Q = 0.10 \pm 0.005$ eV, $\tau_0 = (1.6 \pm 0.5) 10^{-12}$s) are derived for the pure samples. Comparison with a single Debeye peak shows that the measured BoI peak is broadened by the factor $\approx 2$. 

![FIG 5b](image2.png)

*FIG 5b*
Fig. 6 collects our results showing the influence of \( \varepsilon \) and impurity content on the main MHz attenuation contributions \( \alpha_{\text{GL}}, \alpha_{\text{BO}} \) and \( \alpha_B \) (at \( f=10\text{MHz}, T=T_{\text{BoI}}=131\text{K} \)). The non-dislocation \( \alpha_B \)-surface is unaffected by doping and small deformations (\( \varepsilon < 2\% \)). For \( \varepsilon > 2\% \) the \( \alpha_B \) values increase due to sound wave scattering caused by deformation induced lattice inhomogeneities /13/. The shape of the dislocation resonance \( \alpha_{\text{GL}} \)-surface is the same as measured by Drescher et al /13/ on pure Cu at RT. The \( \alpha_{\text{GL}} \) increase up to \( \varepsilon \simeq 0.2\% \) (pure Cu) and \( \varepsilon \simeq 0.4\% \) (130ppmAu) is due to increasing dislocation density \( (\Lambda \varepsilon) /4 \). The \( \alpha_{\text{GL}} \)-levelling-off and -decrease at higher \( \varepsilon \) is caused by loop-length shortening /13/. We point out that the background damping mentioned by Niblett /11/ also exhibits a maximum as function of deformation. The Bordoni relaxation \( \alpha_{\text{BO}} \)-surface emerges at \( \varepsilon = 1 \) to 2\% from the background due to \( \alpha_{\text{GL}} + \alpha_B \). Since Bordoni relaxation is known to be brought out by internal stresses /6/ we attribute the observed \( \alpha_{\text{GL}} \) reduction in this \( \varepsilon \)-range (at least partly) to dislocation/dislocation strainfield interaction in e.g. tangles or dipoles.

Compressional deformation and the \( \alpha_B \) increase at high \( \varepsilon \) prohibited the investigation of \( \alpha_{\text{BO}}(\varepsilon) \) beyond \( \varepsilon \simeq 10\% \). Nevertheless, the results clearly show that \( \alpha_{\text{BO}} \) increases less than proportional to \( \varepsilon \) indicating L-shortening effects (as observed for \( \alpha_{\text{GL}} \) at \( \varepsilon > 0.2 \) to 0.4\%). Since it can be assumed that \( L^{-1/\sqrt{A}} \) and \( A-\varepsilon \) the value \( x=2 \) in equ.(2) would lead to saturation of \( \alpha_{\text{BO}} \) at high \( \varepsilon \); but \( x=1 \) would result in a \( \alpha_{\text{BO}}-\varepsilon^{-1/2} \)-dependence. Experiments with \( \varepsilon > 10\% \) are needed for final conclusions. In comparison with \( \varepsilon \) the impurity influence on the different \( \alpha \)-contributions in fig.6 is minor. (We attribute the pronounced BR at \( \varepsilon = 10\% \) in the 130ppmAu doped sample to enhanced easy glide which is known to occur in solution hardened crystals). Both \( \alpha_{\text{GL}} \) and \( \alpha_{\text{BO}} \) decrease with increasing impurity level in accordance with theory (equ.(1) and (2)): The effect is stronger on \( \alpha_{\text{GL}} \simeq L^4 \) than on \( \alpha_{\text{BO}} \simeq L^X \) (with \( 1 < x < 2 \)). These results suggest additional MHz
Bordoni experiments on still higher concentrated alloy crystals as well as on strongly irradiation pinned pure material.

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