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# Grain boundary relaxation in yellow gold bi-crystals

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ABSTRACT

The mechanical loss spectrum of a yellow gold bi-crystal is presented and analyzed in detail. The relaxation strength is monitored as a function of several geometrical parameters such as sample width, length and thickness. It is found that the relaxation strength is proportional to the GB density (the inverse width), whereas it depends linearly on the sample thickness. The experimental findings are compared to finite elements (FE) simulations, where the material can glide frictionless along the grain boundary. The simulations show the same dependencies as the mechanical loss measurements. The relaxation peak in the loss spectrum can be interpreted as due to GB sliding accommodated by the elastic deformation of the grains.



## 1. Introduction

In metals, plastic deformation is achieved by dislocation motion. However, in polycrystalline specimens, a deformation between adjacent grains is often observed at high temperature. A number of studies testify the existence of grain boundary (GB) sliding, for example in ultrafine-grained Cu [1], in Au thin films [2], in AuPd [3] as well as in Pd [4] nanocrystalline materials. Direct observations of GB sliding have been made on single boundaries in Au nanopillars [5] and in Zn bi-crystals [6].

Mechanical spectroscopy is a non-destructive experimental technique that probes the mechanical response of a specimen to a periodic excitation. The technique is sensitive in particular to of GB sliding. A first model for GB sliding was developed by Zener in 1941 [7], which was verified experimentally by Kê [8] in polycrystalline Al. A GB peak in the mechanical loss spectrum has been observed in different polycrystalline materials [9–13]. This peak is absent in single crystalline specimens, which suggests that the relaxation peak is directly related to the presence of GBs.

A recent study on Au polycrystals [14] together with molecular dynamics simulations has shown that the microscopic stress relaxation mechanism acting at the GBs is probably due to GB sliding connected to a change in the GB structure. At elevated temperatures, the GB layer becomes disordered and a shearing parallel to the boundary plane is observed in the simulations.

In polycrystalline specimens, the experimental results reflect the mixed contributions from different types of GBs, which are characterized by distinct structures and properties [15]. In order to characterize the relaxation mechanism responsible for the mechanical loss peak, it is advantageous to study bi-crystals containing a single GB. Few studies on bi-crystals reporting the measurement of mechanical loss spectra are found in the literature, for example those related to pure Cu [16] and Al [17].

The investigations by Shi et al. [18,17] on Al bi-crystals have been performed as a function of the misorientation angle showing that the activation parameters are distinct for high and low angle GBs. Furthermore, it was shown by Jiang et al. [19] that the relaxation strength of the Al bi-crystal's peak is proportional to the GB density, defined as the GB area per unit volume.

Up to now, mechanical spectroscopy measurements on bi-crystals were made on pure metals. The present work shows that the mechanical loss peak of bi-crystals can also be observed in metallic alloys.

In this paper, we investigate the mechanical loss spectrum of a gold alloy bi-crystal as a function of different geometrical parameters such as the sample width, the thickness and the length, which account for different volume fractions with respect to the GB surface and for different stress distributions in the GB plane. The changes of the relaxation strength are compared with finite elements (FE) simulations, where we used the same sample dimensions as in the experiments. From the simulations, an empirical formula of the relaxation strength for rectangular sample geometries is derived, which can then be compared to the results of previous model calculations [14] assuming GB sliding at the origin of the stress relaxation mechanism.

## 2. Experimental methods

Bi-crystals of a commercial 18-carat (75 weight%) gold alloy, which contains 30.5 atomic% Ag and 9.9 atomic% Cu were

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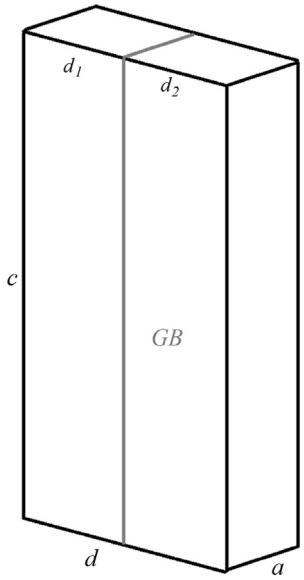


**Fig. 1.** Schematic drawing of the bi-crystal with vertical GB showing the width $d$, the thickness $a$ and the length $c$. The deformation axis is the vertical axis along $c$.

produced by the Bridgman technique. The sample was cut so that the GB is vertical in the torsion pendulum and the torsion axis lies in the GB plane. During the mechanical loss measurements, the lower sample end is fixed while the upper end is twisted periodically. The initial dimensions of the bi-crystal were $(12.3 \times 4.3 \times 0.78)$ mm. Fig. 1 shows the sample geometry. The length $c$, the width $d$ and the thickness $a$ were reduced step by step to observe in which way the loss spectrum of the bi-crystal depends on these parameters. The grain structure and the misorientation angle were controlled before and after the measurements. The misorientation angle remained stable at $\delta = (51.7 \pm 0.2)^\circ$. Tilt and twist components were present, characterizing this specific GB as a random boundary.

For comparison, a polycrystalline specimen was cut from a cylindrical wire of 2 mm in diameter. This sample had a homogeneous and small grained structure with an average grain diameter of 30 μm. A single crystal was grown in the same production step as the bi-crystal in the Bridgman oven. The bi-crystal's width was reduced by spark cutting, removing 1 mm of single crystalline material from the side. The left-over single crystal could be measured in the torsion pendulum.

The mechanical loss and the dynamic shear modulus have been measured in a forced torsion pendulum as a function of temperature at an imposed frequency of $f = 0.5$ Hz and at a constant strain amplitude of $1 \times 10^{-5}$. The heating and cooling rate was 2 K/min. In the case of an anelastic relaxation with a characteristic time $\tau$, the mechanical loss as a function of $\omega = 2\pi f$ takes the form of a Debye peak [20]:

$$\tan \phi = \Delta \cdot \frac{\omega\tau}{1+\omega^2\tau^2} \tag{1}$$

where $\Delta$ is the relaxation strength. The height of the Debye peak $\Delta/2$ yields the relaxation strength, which is also defined as

$$\Delta = \frac{\gamma_{an}^{\infty}}{\gamma_{el}} = \frac{\epsilon_{an}}{\epsilon_{el}} \tag{2}$$

that relates the unrelaxed shear strain angle $\gamma_{el}$ with the relaxed (anelastic) strain angle $\gamma_{an}^{\infty}$. The maximum of the peak in Eq. (1) is obtained for $\omega\tau = 1$. If the relaxation process is thermally activated, the relaxation time $\tau$ follows an Arrhenius equation: $\tau = \tau_0 \exp(H_{act}/(k_B T))$, where $\tau_0$ is the limit relaxation time and $H_{act}$ denotes the activation enthalpy.

If a peak in a mechanical loss spectrum appears broadened, one can assume that the relaxation time varies continuously around a mean value $\tau_m$ according to a log-normal distribution [21]:

$$\Psi(z) = \frac{1}{\sqrt{\pi}\beta} \exp\left(-\frac{z^2}{\beta^2}\right) \quad \text{with } z = \ln\left(\frac{\tau}{\tau_m}\right) \tag{3}$$

The broadening is characterized by the distribution parameter $\beta$. Assuming that the relaxation strength $\Delta$ is identical for all values of $\tau$, one can calculate the analytic expression of the broadened Debye peak as a convolution of Eq. (1) with (3):

$$\tan \phi = \frac{\Delta}{2\sqrt{\pi}\beta} \int_{-\infty}^{\infty} \frac{\exp\left(-\frac{z^2}{\beta^2}\right)}{\cosh((\ln \omega\tau) + z)} dz. \tag{4}$$

A thermally activated and broadened Debye peak can be fully described by four parameters: the relaxation strength $\Delta$, the activation energy $H_{act}$, the limit relaxation time $\tau_0$ and the broadening factor $\beta$.

## 3. Results

Fig. 2 shows the mechanical loss spectrum and the dynamic shear modulus as a function of temperature of the polycrystal, the bi-crystal and the single crystal, which had been a part of the bi-crystal before. The mechanical loss shows peak P1 at around 600 K that may be attributed to a Zener relaxation due to stress induced diffusion of Cu atom pairs [22]. The P1 peak is present in all samples. Heating and cooling of the bi-crystal superimpose perfectly indicating that the microstructure is stable between room temperature and 950 K. The spectra of the single crystal and the bi-crystal superimpose very nicely except for the peak P2. The polycrystal's spectrum is equally shown and its P2 peak is about twice as high and much broader.

Since the P2 peak is exclusively present in samples containing GBs, it can be concluded that the peak P2 is closely related to the presence of GBs. It can be ruled out that P2 is due to dislocation relaxation in the bulk or to a relaxation related to twin boundaries. The contribution of the bulk material is given by the spectrum of the single crystal and thus, the difference between the single crystal's spectrum and the bi-crystal's spectrum must be due to the grain boundary, which is the only structural defect that distinguishes single and bi-crystal.

From Fig. 2, it can be seen that the mechanical loss increases at high temperatures. However, in the fit of the complete spectrum, the high temperature background should be taken into

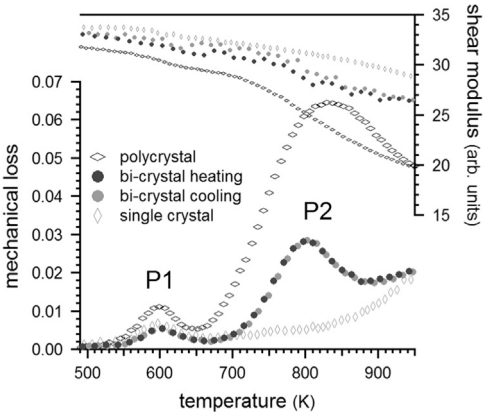


**Fig. 2.** Mechanical loss spectrum of the bi-crystal with a vertical GB. A relaxation peak accompanied by a modulus drop is observed at 800 K. The spectrum of a polycrystal and a single crystal is shown for comparison. The single crystal has been cut from one side of the bi-crystal.