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Grain boundary formation stages in a deformed yellow gold single crystal studied by mechanical spectroscopy

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ABSTRACT

A monocrystalline 18-carat yellow gold sample is deformed stepwise from 2% to 12% at room temperature. The mechanical loss spectrum is recorded and analysed. At low deformations the exponential high temperature background increases. At a critical deformation of 8% an intermediate temperature peak appears. Such peak typical of polycrystals is related to grain boundary sliding. The comparison of a polycrystal formed of low angle grain boundaries (LAGB) only, and samples deformed between 2% and 8% evidence the existence of an intermediate stage. The high temperature peak P3 characterizes the mobility of LAGB that is controlled by the movement of dislocations.

1. Introduction

Grain boundaries emerge as interfaces between individual crystallites during solidification or recrystallization of a material. A mechanical loss peak has been observed at intermediate temperature ($\sim 0.5T_m$=melting temperature for f=0.5 Hz) in a polycrystalline gold alloy [1] and has been related to grain boundary sliding [2,3] since this peak is absent in single crystals of the same alloy composition.

Grain boundary peaks have been observed in the past, for example in aluminium [4,5]. On the other hand, Woirgard et al. [6,7] reported internal friction measurements in different pure metals (Al, Cu, Ni). They observed similar relaxation peaks as well in single crystals than in polycrystals. They concluded that damping originates mainly from lattice dislocation motion. More recent studies [8,9] report about dislocation motion inside sub-grain boundaries that lead to relaxation peaks in internal friction measurements.

Recrystallization of a heavily deformed solid takes place when the material is heated up and dislocation mobility increases. During the first stage, called recovery, the dislocation density is reduced by mutual annihilation of dislocations with opposite Burgers vectors. Reordering of the remaining dislocations leads to the formation of sub-grains (low dislocation density) and sub-grain boundaries (high dislocation density) [10]. The formation of well-defined grain boundaries followed by grain growth leads to the final polycrystal.

Recrystallization is evidenced by mechanical spectroscopy by a large recrystallization peak that depends on the heating rate, for example in Cu [11], in Ag [12] or in Au [13]. This is the case for a heavily deformed sample with an initial large dislocation density. But what happens for low deformations, when the dislocation density is not large enough leading to recrystallization? Are there intermediate stages? This problem has been investigated by [14] that concludes that some elastic misfit energy must be stored in order to initiate recrystallization.

The present paper investigates the microstructure changes of a single crystal that is deformed stepwise to introduce dislocations up to the critical density for recrystallization. Such microstructure changes are monitored by internal friction. The changes of the internal friction spectra provide specific information about the microstructure and in particular about the appearance of grain boundaries.

2. Mechanical spectroscopy

Mechanical loss is caused by anelastic relaxation in a material, which produces a lag of the specimen's strain response with respect to an applied cyclic stress. In a torsion pendulum, a periodic stress with frequency f is applied on a sample. The phase lag ϕ between stress and strain is directly measured and can be related to the mechanical loss: $\tan\phi$. The dynamic shear modulus G at the frequency f is obtained from the ratio of stress and strain

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amplitude [15]. In the case of an anelastic relaxation with a characteristic time τ, the mechanical loss as a function of $\omega = 2\pi f$ has the shape of a Debye peak [16]:

$$\tan\phi = \frac{\Delta}{\sqrt{1+\Delta}} \cdot \frac{\omega\tau}{1+\omega^2\tau^2} = \frac{\Delta}{2\sqrt{1+\Delta}} \cdot \frac{1}{\cosh(\ln \omega\tau)} \quad (1)$$

and the dynamic modulus is

$$M(\omega) = M_u - \frac{\delta M}{1+\omega^2\tau^2} \quad (2)$$

where M_u is the unrelaxed modulus. The height of the Debye peak $\Delta/(2\sqrt{1+\Delta})$ yields the relaxation strength Δ that is also defined as

$$\Delta = \frac{\gamma_{an}^{\infty}}{\gamma_{el}} \quad (3)$$

that relates the unrelaxed shear strain angle γ_{el} with the relaxed (anelastic) strain angle γ_{an}^{∞}. The maximum of the peak described by Eq. (1) is obtained for $\omega\tau = 1$. Therefore, the measured position of the Debye peak yields immediately the relaxation time $\tau = 1/\omega_{max}$ for a given temperature. If the relaxation is thermally activated, the relaxation time will follow an Arrhenius equation:

$$\tau = \tau_0 \exp\left(\frac{\Delta G}{k_B T}\right) = \tau_0 \exp\left(\frac{\Delta H - T\Delta S}{k_B T}\right) = \tau_0' \exp\left(\frac{H_{act}}{k_B T}\right) \quad (4)$$

where ΔG is the activation free energy and τ_0 denotes the limit relaxation time. The entropy change with temperature can generally be neglected, so that the activation enthalpy H_{act} is the parameter issued from the measurements.

In most cases, more than a single relaxation time contributes to the same peak and the measured peak is broadened compared to the Debye peak in Eq. (1). The broadening is quantified by a parameter β assuming a log-normal distribution of relaxation times around a mean value τ_m [17]:

$$\Psi(z) = \frac{1}{\sqrt{\pi}\beta} \exp\left(-\frac{z^2}{\beta^2}\right) \quad \text{with } z = \ln\left(\frac{\tau}{\tau_m}\right) \quad (5)$$

The analytic expression of the broadened Debye peak is given by the convolution of Eq. (1) with (5):

$$\tan\phi = \frac{\Delta}{2\sqrt{1+\Delta}\sqrt{\pi}\beta} \int_{-\infty}^{\infty} \frac{\exp\left(-\frac{z^2}{\beta^2}\right)}{\cosh((\ln \omega\tau) + z)}\, dz. \quad (6)$$

An exponential background can be described by three parameters K, n and H_{act} [18]:

$$Q^{-1} = \frac{K}{(2\pi f)^n} \exp\left(-\frac{nH_{act}}{k_B T}\right) \quad (7)$$

where n is the broadening factor and K is the amplitude of the background.

Mechanical spectroscopy measurements were performed by means of a forced inverted torsion pendulum on a commercial 18-carat (75 wt%) gold alloy, which contains 30.5 at% silver and 9.9 at% copper. The polycrystalline specimen was cut from a wire, 2 mm in diameter, supplied by Varinor SA, Delémont, Switzerland. Some parts of the wires were re-melted in order to obtain single crystalline material by the Bridgman technique. X-ray measurements with a Laue camera confirmed the single crystalline nature over the whole length of the specimen. Another remelted sample showed low angle grain boundaries (LAGBs) with misorientations between adjacent grains lower than 10°.

The mechanical loss measurements were performed either in isothermal conditions as a function of frequency (between 0.004 Hz and 1 Hz) or at fixed frequency as a function of temperature. Temperature scans were performed at a frequency of 0.5 Hz, at a stress amplitude of 0.5 MPa and a heating/cooling rate of 2 K/min if not indicated differently.

The deformation of the single crystal was performed stepwise from small to large deformations on the same sample. The single crystalline wire was clamped on both ends and it was twisted four times into each direction by an angle corresponding to 2%, 4%, 6%, 8% and 10% shear deformation at room temperature around the sample axis. After each deformation, the isothermal and isochronal measurements were performed on the sample in the torsion pendulum. After 10% deformation the sample was polished and etched in order to observe the microstructure.

3. Results

3.1. Undeformed single crystal

Fig. 1(a) shows the mechanical loss spectrum of a polycrystalline sample in comparison to the single crystalline sample made from the same material. The mechanical loss shows a peak P1 at around 600 K that may be attributed to the Zener relaxation due to stress induced diffusion of the copper atoms [19]. This peak is also present in the spectrum of the single crystal. The large peak P2 at around 750 K is due to grain boundaries as it is absent in

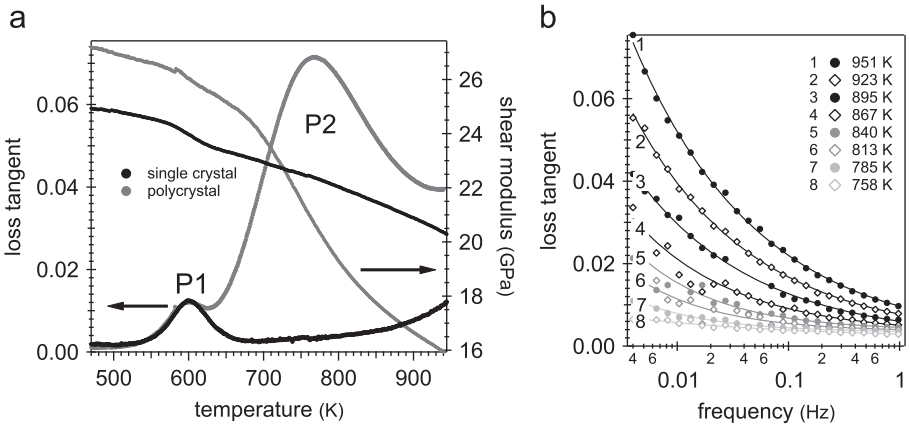

Fig. 1. (a) Typical mechanical loss spectrum of a polycrystal and a single crystal with the corresponding mechanical modulus. The mechanical loss of the polycrystal contains two peaks, the Zener peak P1 at 600 K and a large and brought peak P2 at 760 K that is not present in the single crystal. The peaks are superimposed on a high temperature exponential background. (b) Isothermal measurements of the monocrystalline background. For lower temperature the background shifts to lower frequencies.