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GRAIN BOUNDARY RELAXATION IN Bi-CRYSTALS: MECHANICAL SPECTROSCOPY AND MOLECULAR DYNAMICS SIMULATIONS

RELAKSACJA GRANIC ZIAREN W BIKRYSZTAŁACH: SPEKTROSKOPIA MECHANICZNA I SYMULACJE METODĄ DYNAMIKI MOLEKULARNEJ

Different Au-Ag-Cu samples have been studied by mechanical spectroscopy. Both polycrystals and bi-crystals show a relaxation peak at 800 K, accompanied by an elastic modulus change. Since this peak is absent in single crystals it is related to the presence of grain boundaries. Molecular dynamics simulations reveal two microscopic mechanisms, when a shear stress is applied onto a Σ5 grain boundary: at 700 K, the boundary migrates perpendicularly to the boundary plane under an external stress. At 1000 K, only sliding at the boundary is observed. These two mechanisms acting in different temperature intervals are used to model the mechanic response of a polycrystal under an applied stress. The models yield expressions for the relaxation strength Δ and for the relaxation time τ as a function of the grain size. A comparison with the mechanical spectroscopy measurements of polycrystals and the bi-crystals show that the grain boundary sliding model reproduces correctly the characteristics of the grain boundary peak.

Keywords: Internal friction grain boundaries, gold alloy, molecular dynamics

Różne próbki ze stopów Au-Ag-Cu badano metodą spektroskopii mechanicznej. Zarówno w polikryształach, jak i bikryształach występuje pik relaksacyjny w temperaturze 800 K, któremu towarzyszy zmiana modułu sprężystości. Ponieważ pik ten nie występuje w monokryształach to jego występowanie wiązane jest z obecnością granic ziaren. Symulacje dynamiki molekularnej ujawniają dwa mikroskopowe mechanizmy, gdy naprężenie ścinające jest przyłożone do granicy ziarna Σ5: w temperaturze 700 K, granica przemieszcza się prostopadle do płaszczyzny granicznej pod wpływem zewnętrznego naprężenia. W 1000 K, obserwuje się tylko poślizg po granicy. Te dwa mechanizmy działające w różnych zakresach temperatur są używane do modelowania mechanicznej reakcji polikryształu na przyłożone naprężenie. Modele podają wyrażenie na stopień relaksacji Δ i czas relaksacji τ w funkcji wielkości ziaren. Porównanie z wynikami badań polikryształów i bikryształów uzyskanych metodą spektroskopii mechanicznej pokazuje, że model poślizgu granicy ziarna poprawnie odtwarza charakterystykę piku pochodzącego od granic ziaren.

1. Introduction

In many metallic polycrystals, a relaxation peak is observed at about half the melting temperature [1-3], which has been either attributed to GB sliding [4] in agreement with the Zener model [5] or to dislocation mechanisms [6,7]. Observations on Al bi-crystals containing a single GB showed a relaxation peak closely related to the GB and dependent on the misorientation angle [8].

The presented work aims to give an answer to the question: which microscopic mechanism is at the origin of the relaxation peak observed in many metals at intermediate temperatures? For this purpose, molecular dynamics (MD) simulations have been used to illustrate the possible microscopic relaxation mechanisms that act at the GB in a bi-crystal. Furthermore, two models have been developed to describe the mechanical loss spectrum, which give explicit expressions of the relaxation strength and the relaxation time in terms of parameters such as the grain size d, the GB viscosity η and the misorientation angle.

2. Experiments and simulations

The mechanical spectroscopy measurements were performed in a forced torsion pendulum at the driving frequency of $f = 0.5$ Hz. The polycrystalline specimen of a commercial 18-carat yellow gold containing 59.6 atomic-% Au, 30.5% Ag and 9.9% Cu. Parts of the material were melted in a Bridgman oven to produce single crystals and bi-crystals grown from 2 single crystalline seeds. The misorientation of the bi-crystal is $\delta = (51.7 \pm 0.2)^\circ$, measured with a Laue camera. Fig. 1 shows the mechanical loss spectra as a function of temperature of a polycrystal, a bi-crystal and a single crystal, which had been cut from the side of the bi-crystal. The samples containing GBs show the P2 peak at around 800 K, whereas the P2 peak

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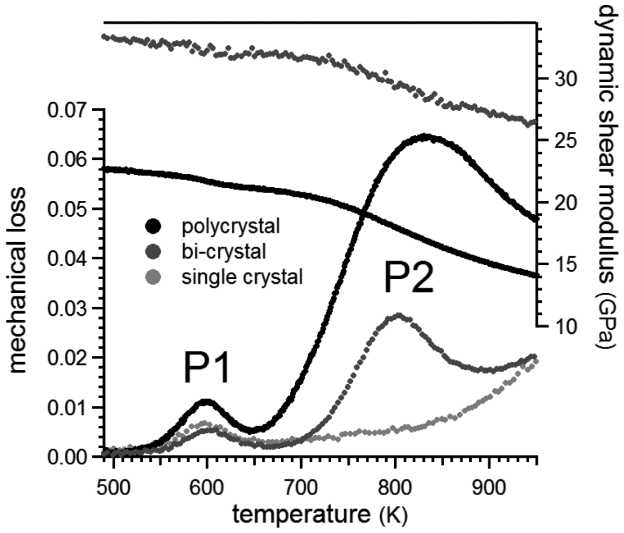

Fig. 1. Mechanical loss spectra as a function of temperature at f = 0.5 Hz. The grain boundary peak P2 is present in the polycrystal and in the bi-crystal, whereas it is absent in the single crystal's spectrum. The P1 peak is a Zener peak due to the presence of Cu atoms in the alloy. Both peaks are accompanied by a drop of the dynamic shear modulus

is absent in the single crystal. The GB peak P2 of the polycrystal is about twice as high as in the bi-crystal and much broader.

Figure 2 a) shows the mechanical loss spectra of the same bi-crystal, where the width d of the rectangular sample was reduced by removing singe-crystalline material parallel to the boundary plane without cutting the GB. The height of the GB peak increases strongly with decreasing d, whereas the peak position and shape remain the same. In fact, it is not only the peak that increases, but the whole high temperature part of the spectrum. The peak height of the bi-crystal's peak as a function of the inverse width $1/d$ is shown in Fig. 2 b). It turns out that the peak height and therefore the relaxation strength is directly proportional to the GB density $s = A/V = 1/d$, where A and V are the GB area and the bi-crystal's volume, respectively.

In order to reveal the stress relaxation mechanism of the P2 peak acting at the microscopic level, molecular dynamics (MD) simulations with the software package Lammps [9] and an inter-atomic potential for Au [10] were performed simulating approximately 20 000 atoms. A Σ5 bi-crystal (Fig. 3 a) is subjected to a shear deformation parallel to the GB by moving an upper slice of atoms with a constant velocity of 10 m/s.

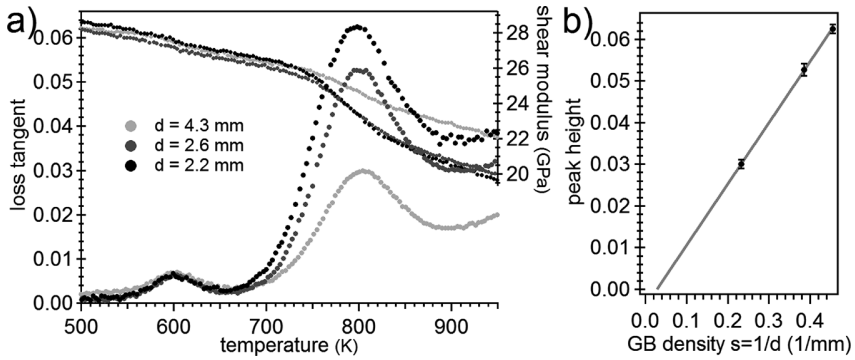

Fig. 2. a) Dependence of the bi-crystal's peak on the width d. b) The peak height as a function of the inverse width $1/d$ shows a linear relationship

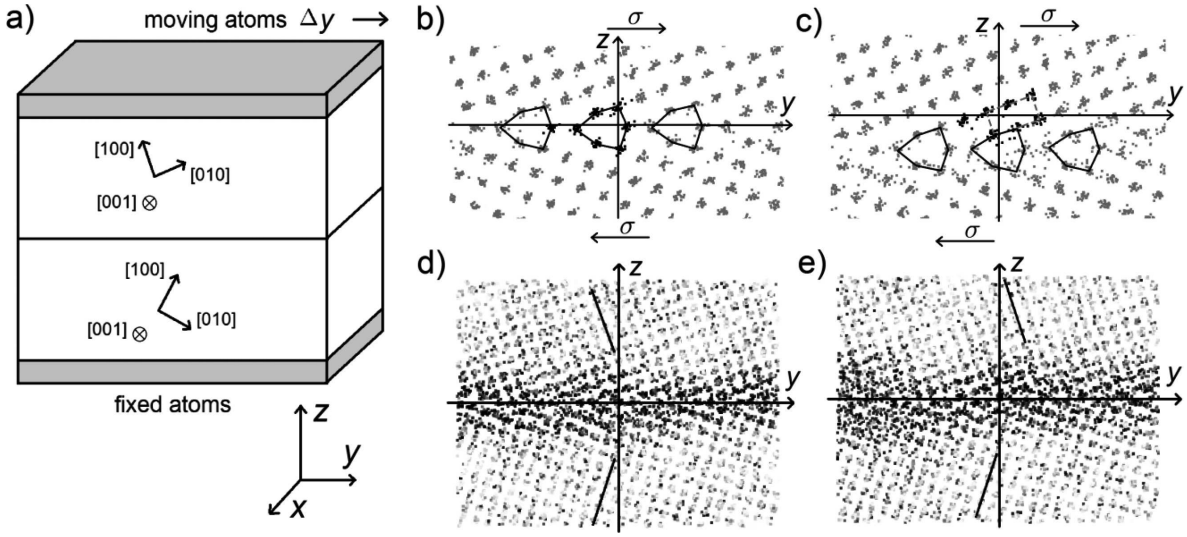

Fig. 3. a) Geometry of the molecular dynamics simulation. The sample is composed of 2 blocks with different crystal orientations. The upper slice of 5 Å moves with a velocity of 10 m s^{-1}. b) and c) show the atomic positions of the molecular dynamics simulations before and after shear deformation at 700 K. The grain boundary is represented by the kite shaped units in black that move downwards. d) and e) Simulations at T = 1000 K: The grain boundary is visible as a black band. Under shear stress, the upper crystal moves to the right with respect to the lower crystal. The black diagonal lines mark the same atom planes before and after shearing

In a low temperature range up to $T = 700$ K, the GB performs a migration perpendicular to the boundary plane coupled to shear. Fig. 3 b) shows the atom positions projected onto the yz-plane before shearing. The GB at $z = 0$ is marked by kite shaped units, which can also be understood as the cores of perfect 1/2[110] edge dislocations. The application of a shear stress (Fig. 3c) provokes a GB migration. In the dislocation picture, the migration corresponds to a glide of the dislocation array on (110)-type planes. At high temperatures above $T = 1000$ K, the GB structure changes radically (Fig. 3d). Under shear, the upper crystal part moves as a block with respect to the lower part. The z position of the boundary plane remains the same. Therefore, GB sliding is observed as a high temperature relaxation mechanism.

3. "Migration model" and "sliding model" for interpreting the mechanical loss spectrum

The "migration model" is inspired by the GB migration mechanism, which has been observed in a low temperature range in the simulations. The smallest unit to consider in a polycrystal or a bi-crystal is one GB surrounded by crystalline material. Fig. 4 a) and b) show the set-up for the first model, where the GB is visualized as membrane having a surface tension γ, which bends under shear. It has been shown in Ref. [11] that this system is governed by the differential equation of the standard anelastic solid with a relaxation time

$$\tau_{migration} = \frac{\eta}{4\pi\gamma} = \frac{k_B T}{4\pi\gamma D_0} \exp\left(\frac{H_{act}}{k_B T}\right), \qquad (1)$$

where η is the viscosity of the membrane that can be thermally activated with the diffusion coefficient D_0 and the activation energy H_{act}. The relaxation strength of a Debye peak is

$$\Delta_{migration} = \frac{\vartheta^2 a^2}{16\gamma d J_N}, \qquad (2)$$

where ϑ is the misorientation angle, J_N is the elastic compliance and a is defined in Fig. 4 a). Analogous expressions can be found for the "sliding model", where sliding is possible at the GB plane, but triple lines are fixed. We find [11]:

$$\tau_{sliding} = \frac{\eta a}{\delta k} = \frac{k_B T a}{\delta k D_0} \exp\left(\frac{H_{act}}{k_B T}\right) \text{ and } \Delta_{sliding} = \frac{a}{2dkJ_N}, \qquad (3)$$

where δ is the thickness of the viscous GB layer and k is a restoring force constant due to the fixed triple lines. k is independent of the geometrical length scales a, c, d of Fig. 4 c). In a polycrystal without preferred grain orientation, one can assume that the grains have a more or less cubic shape. In the expressions for the migration model (Eqs. (1) and (2)) one can set $2a = d$ and in the sliding model (3) $a = d$. The relaxation strength of a polycrystal with grain size d is therefore

$$\Delta_{migration,poly} = \frac{\vartheta^2 d}{64\gamma J_N} \text{ and } \Delta_{sliding,poly} = \frac{1}{2kJ_N}. \qquad (4)$$

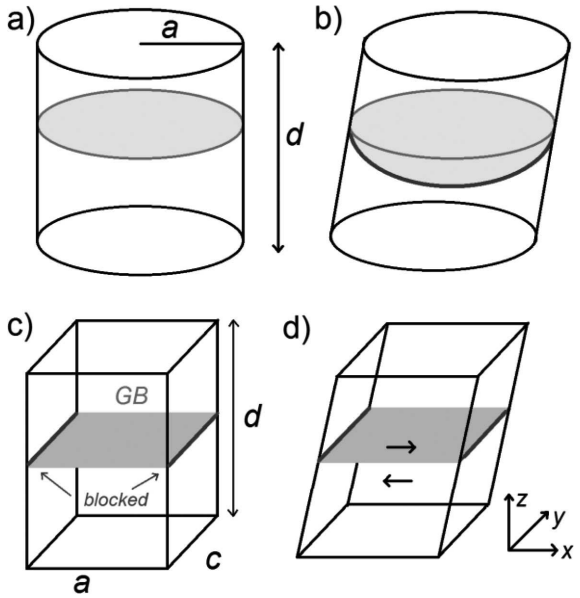

Fig. 4. Geometry for the migration model (a and b) and the sliding model (c and d)

If GB migration coupled to shear were the microscopic relaxation mechanism, the relaxation strength of a polycrystals should scale with the grain size d. This is not observed in the experiments [12]. We can therefore rule out GB migration being responsible for the GB peak. In fact, the GB peak has the relaxation strength independent of d and the relaxation time $\tau \propto d$, which is in agreement with the GB sliding. Furthermore, the measurements of the bi-crystal prove the explicit dependence $\Delta \propto d$, when the parameter a is constant in Eq. (3).

4. Summary

A mechanical loss peak closely related to GBs has been observed in polycrystalline samples as well as in bi-crystals of the same material. The relaxation peak height increases when the GB density in the bi-crystal is increased. Molecular dynamics simulations on a bi-crystal under shear show two different microscopic mechanisms, which could give rise to the relaxation peak: GB migration coupled to shear or GB sliding. Two simple models taking into account the two relaxation mechanisms have been developed, which lead to expressions of the relaxation strength Δ as well as the relaxation time τ. A comparison with the mechanical spectroscopy measurements shows that the sliding model is in agreement with the peak in the polycrystal as well as with the measurements in the bi-crystal, whereas the migration model cannot reproduce correctly the dependency on the grain size. The mechanical loss peak is therefore due to GB sliding.

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