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Grain boundary sliding in thin substrate-bonded Al films*

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Abstract

A systematic investigation of the anelastic relaxation of thin Al films on Si substrates has been carried out. It was found that the relaxation in both bulk and thin film materials can be explained by a model involving the glide of grain boundaries (GBs). The mass transport necessary for the glide occurs via GB diffusion in thin films and via lattice diffusion in bulk material, the different behaviour being due to the smaller (by more than two orders of magnitude) grains in the films. Internal friction thus provides a technique to measure diffusional parameters of GB diffusion in thin films.

1. Introduction

In his pioneering work on the internal friction (IF) of polycrystalline Al, Kê observed a strong relaxation peak which is not present in Al single crystals [1]. This peak was attributed to the viscous sliding of grain boundaries where the restoring force driving the relaxation is built up by elastic back stresses at triple-grain junctions. There remained, however, the puzzling question of why the activation energy derived from IF measurements is as high as 1.4 eV, thus being close to that of lattice self-diffusion in Al [2], whereas one might expect that the GB viscosity should be controlled by GB diffusion where the activation energy is about 0.6 eV [3]. Interestingly enough, from IF experiments on thin Al films adhering to silica substrates an activation energy of 0.55 eV has been derived [4]. Thus a systematic investigation of the mechanical relaxation of substrate-bonded thin Al films may contribute to resolve the long-standing controversy about the origin of the IF peak in polycrystalline Al.

On the other hand, IF has already been employed to characterize various properties of thin Al and Al alloy films. In particular, it was used to determine activation energies in several Al alloys [5], to study the influence of impurities on the GB diffusion at concentration levels as low as 300 ppm and to demonstrate the potential to be a non-destructive test on the adhesion strength between film and substrate [6]. Most of these results were based on qualitative arguments. A better understanding clearly requires a microscopic understanding of the underlying relaxation mechanism.

In this paper we report on a study of the mechanical relaxation in thin Al films on Si substrates, where the film thickness, grain size and film–substrate interface were varied. Also, the mechanical deformation state was varied by employing both bending and torsional vibrations.

2. Experimental details

2.1. Sample preparation

The Al films were evaporated from 5N and 6N Al respectively on to oxidized Si wafers. The thickness of the thermal oxide was varied between 0.3 and 1 μm. Some substrates were additionally coated with a 0.34 μm $MoSi_{2.2}$ layer in order to further modify the film–substrate interface. The base pressure of the evaporating system was below 10^{-7} Pa. After deposition the grain structure of the films was stabilized by annealing for 30 min at 450 °C in a vacuum below 10^{-4} Pa. This procedure resulted in columnar grains with an average size essentially equal to the film thickness and a high degree of ⟨111⟩ fibre texture. Exceptionally large grain sizes (*e.g.* 30 μm for a film 5 μm thick) could be obtained by deposition on substrates heated to 550 °C.

2.2. Internal friction measurement

The IF was measured by means of a highly sensitive apparatus specially designed to study the damping of thin films adhering to thick substrates [7, 8]. T-shaped samples were employed which allowed measurements to be made in both bending and torsional modes of vibration [9].

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The measured total damping Q_c^{-1} of such a composite system is given by [4]

$$Q_c^{-1}=Q_s^{-1}+3\,\frac{t_f M_f}{t_s M_s}\,Q_f^{-1} \tag{1}$$

where Q_s^{-1}denotes the contribution from the substrate, t_f and t_s are the thicknesses of the film and substrate respectively and M_f and M_s are the corresponding elastic moduli. There will be either the Young moduli for a pure bending vibration or the shear moduli for a pure torsional vibration. $Q_f^{-1}$is the damping of the film, which measures the elastic energy dissipation in the film. This has been shown to consist of a superposition of a temperature-dependent background and a relaxation peak [5, 7]. In the IF spectra shown in the following this background and the contribution from the substrate have been subtracted, so only the relaxation peak part of Q_f^{-1}will be plotted.

3. Experimental results

3.1. Origin of the IF peak in thin Al films

From peak shift experiments, *i.e.* measuring IF spectra at various frequencies, it was found that the activation energy of the underlying relaxation mechanism is 0.6±0.05 eV [4, 10]. This value is close to that of GB diffusion in Al [3]. For $AlSi_{0.01}Cu_{0.002}$ and $AlSi_{0.01}V_{0.005}$ values of 0.9 eV were reported, which are equal to those derived from electromigration lifetime tests on the same alloys [7, 11]. It was also found that controlled addition of Cu to $AlSi_{0.01}$ suppresses the IF peak, when most of the GBs are decorated by Al_2Cu θ phase precipitates [11]. From these findings it was concluded that the microscopic mechanism driving the anelastic relaxation must be related to GB processes.

Further evidence for a GB effect is obtained from the dependence of the relaxation spectra on the thickness of the Al films. Figure 1 shows Q_f^{-1}(see eqn. (1)) for an Al film on an $Si/MoSi_{2.2}$ substrate. After deposition and annealing the film thickness was 4.6 μm and the grain size 4.1 μm. The thickness of the film was then reduced by chemical etching while keeping the grain size fixed. The peak height is slightly reduced for thicknesses between 4.6 and 1.4 μm, while the peak almost completely vanishes at 0.6 μm. The temperature position of the peak is unchanged. Inspection of the film after the etching steps by means of an atomic force microscope showed that the etching process primarily attacks the GBs, where after the last step almost all GBs were removed, leaving Al islands on the substrate. All these findings conclusively show that the relaxation peak in thin Al films on Si substrates is a GB effect.

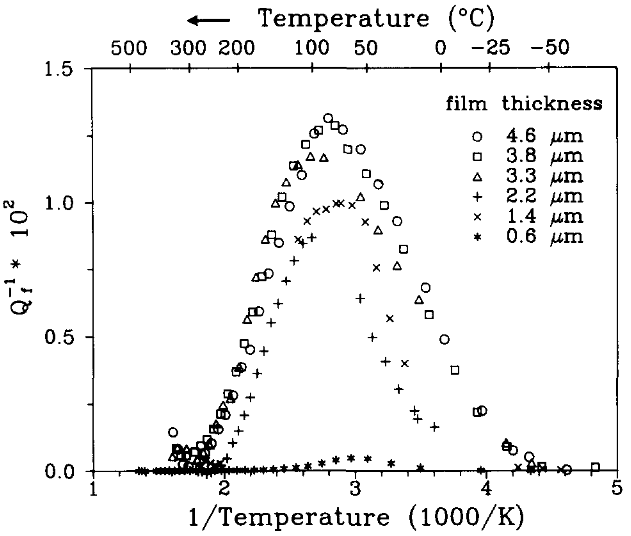

Fig. 1. IF spectra $Q_f^{-1}$of Al on $Si/MoSi_{2.2}$ substrates for various thicknesses of the Al films. The grain size was 4.1 μm and the spectra were measured in torsion at a frequency of 806 Hz.

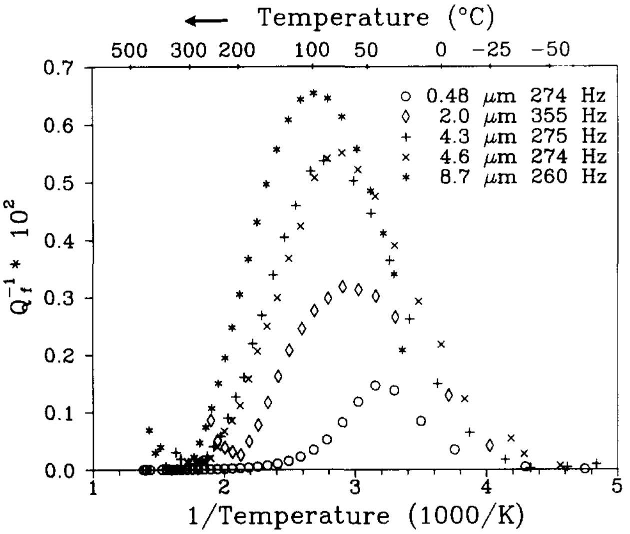

Fig. 2. IF spectra $Q_f^{-1}$of Al on $Si/MoSi_{2.2}$ substrates for various thicknesses of the Al films. The grain size was equal to the film thickness and the spectra were measured in bending at frequencies around 270 Hz, the slight differences being due to different thicknesses of the substrates.

3.2. Relaxation times

Figure 2 shows IF spectra of Al films on $Si/MoSi_{2.2}$ substrates for various thicknesses of the deposited films. The peak position shifts to higher temperature with increasing film thickness and the peak height is considerably reduced for the 2.0 and 0.48 μm films. As mentioned above, for these films the grain size is proportional to the film thickness. Taking into account the results shown in Fig. 1, it must be concluded that the peak shift reflects the grain size dependence of the spectra. The reduction in height can be attributed to the influence of the substrate, which, owing to adhesion, could hinder relative displacements of neigh-

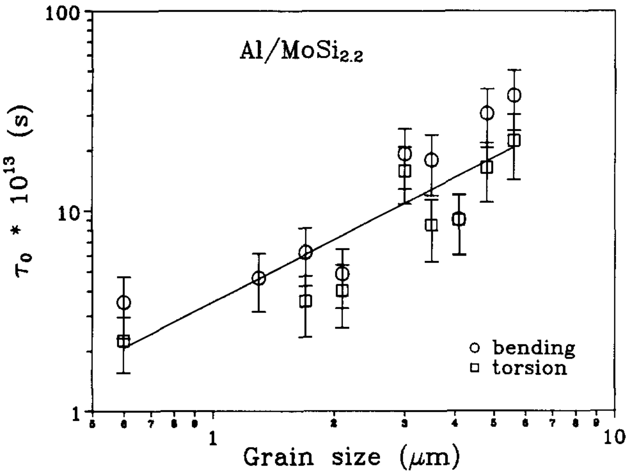

Fig. 3. Grain size dependence of the pre-exponential factor τ_0 for Al films on Si/MoSi$_{2.2}$ substrates. The activation energy was assumed to be 0.62 eV.

bouring grains near the interface. If this occurs for a thin layer whose width is independent of the grain size, the relaxation strength, *i.e.* the peak height, will be considerably reduced for very thin films.

The peak position is given by the condition $2\pi f\tau = 1$, where τ is the characteristic time of the underlying relaxation mechanism and f is the vibration frequency of the sample [12]. If τ is thermally activated, *i.e.* $\tau = \tau_0 \exp(H/kT)$, where H is the activation energy and τ_0 is a pre-exponential factor, the temperature position of the peak, T_p, is obtained from the relation

$$T_p = -\frac{H}{k}\ln(2\pi f\tau_0) \tag{2}$$

It is reasonable to assume that a moderate change in grain size does not influence the activation energy. Thus the peak shift shown in Fig. 2 is expected to result from a change in the pre-exponential factor. Figure 3 shows a plot of τ_0 *vs.* grain size which was obtained from the data of Fig. 2 using eqn. (2) and taking $H = 0.62$ eV. One can see that τ_0 depends linearly on the grain size L.

4. Discussion

Stress relaxation by GB processes occurs in at least three distinct steps generally characterized by significantly different time scales [13, 14]:

(1) GB sliding and build-up of stress concentrations at triple-grain junctions;

(2) diffusional relaxation of these stress concentrations;

(3) long-range diffusional relaxation (Coble creep).

The first and fastest process is associated with a pronounced IF peak [1, 12], the second may result in a weak additional peak under favourable conditions [4, 15], whereas the third can only lead to a peak in a compound system containing a restoring component, *e.g.* in a thin film–substrate system. Here we focus interest on the most important first process.

The microscopic description of stress relaxation by GB sliding is based on the idea that fast glide processes are hindered by GB irregularities [13, 14]. Figure 4(a) shows a schematic sketch of a GB which is not flat on an atomistic scale but contains steps of length l and height h. The average width of the GB is δ. The shear stress σ_s is relaxed by fast glide along the horizontal parts of the GB, leading to normal stresses σ_n at the steps (Fig. 4(b)) which are relaxed by dislocation climb. This is the rate-limiting process; σ_n is the driving force for mass transport from steps under compressional stress to those under tension as indicated in Fig. 4(c).

This transport may occur via bulk or GB diffusion. Assuming here that GB diffusion is dominant, the sliding rate $\dot{u}$ of neighbouring grains is given by

$$\dot{u} = \frac{2\delta\Omega|j_{GB}|}{h} \tag{3}$$

where Ω is the atomic volume and j_{GB} is the particle current density in the GB, which is proportional to the GB diffusion constant D_{GB}, to the difference between the normal stresses, $2|\sigma_n|$, and inversely proportional to the step distance:

$$|j_{GB}| = \frac{2|\sigma_n|D_{GB}}{lkT} \tag{4}$$

Using $l\sigma_s = h\sigma_n$, one obtains for the sliding rate

$$\dot{u} = 4\frac{\sigma_s\Omega\delta}{kTh^2}D_{GB} \tag{5}$$

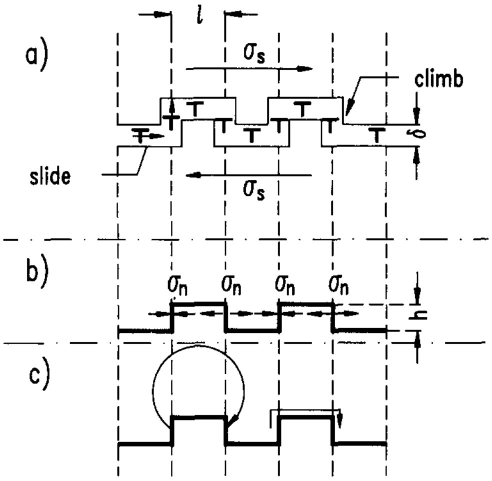

Fig. 4. (a) Schematic view of GB dislocation slide and climb under the action of a shear stress σ_s. (b) Normal stresses σ_n at steps in the GBs. (c) Stress relaxation by diffusion: left, lattice diffusion; right, GB diffusion.

The relaxation time τ can then be estimated from the condition $u=\sigma_s L/2G\tau$ (G denotes the shear modulus), which finally gives

$$\tau=\frac{\sigma_s L}{2G\dot{u}}=\frac{kTh^2L}{8G\Omega D_{GB}\delta} \tag{6}$$

Including bulk diffusion with a diffusion constant D_V, one arrives at an analogous result but where D_{GB} has to be replaced by the effective diffusion constant

$$D_{eff}=\frac{l}{\delta}D_V+D_{GB} \tag{7}$$

A similar result can be found in ref. 13.

This model explains the apparent difference between the IF spectra in bulk and thin film Al. The bulk Al used typically had grain sizes of 200 μm or larger and $l=100$ μm is then a reasonable estimate. At 300 °C, *i.e.* the temperature where the peak in bulk Al was found, the diffusion constant for bulk diffusion in Al is 3×10^{-17} m^2 s^{-1} [2]. The diffusion constant for GB diffusion is not very well known mainly owing to uncertainties in the determination of the pre-exponential factor. If one takes $H=0.62$ eV as the activation energy for GB diffusion, it turns out that D_V is the dominating contribution in eqn. (7) if $D_{GB,0}$ is 3×10^{-6} m^2 s^{-1} or smaller. This value is within the range reported in the literature [3]. In thin films the grain size is two orders of magnitude smaller than in bulk Al and the peak is near 100 °C. Then the GB contribution dominates in eqn. (7) in accordance with the experiments.

It should also be noted that eqn. (6) reflects the measured linear grain size dependence of the relaxation time. The absolute values $\tau_0=10^{-12}$ s (thin film Al) and $\tau_0=10^{-14}$ s (bulk Al) are in agreement with the experimental results.

Relaxation contribution of diffusional processes which require mass transport from one grain to another can be ruled out since their relaxation time would be proportional to L^3, which is in contrast with the experimental observations. Also, the magnitude of τ_0 would be off by orders of magnitude.

An important question concerns the relaxation strength Δ. Experimentally it is derived from the measured peak height by $\Delta=5.4Q^{-1}_{f,\,max}$, which takes into account that the peaks are broadened owing to a distribution of relaxation times which originates from a distribution of grain boundary structures (see eqn. (6)). The measured value of 0.15 (see *e.g.* Fig. 1) is a factor of 2 smaller than the value of 0.38 calculated in ref. 13. Qualitatively this can be explained by the influence of the substrate, which, for perfect adhesion between film and substrate, would inhibit the sliding of neighbouring grains near the interface. Thus only a fraction of the elastic energy can relax in films as compared with bulk material.

5. Summary

We have shown that the IF spectra observed in both bulk and thin film Al can be explained by a model involving grain boundary sliding and dislocation climb at steps in the grain boundaries. For bulk Al the mass transport necessary for the climb process occurs by diffusion through the volume of the material, while for thin films grain boundary diffusion dominates. The reason for the different behaviour lies in the different grain sizes, which are are least two orders of magnitude smaller for thin films than for bulk Al. The model quantitatively describes the measured relaxation times and sufficiently explains the measured relaxation strength if one takes into account adhesion of the thin films to the substrate. These results show that internal friction is a suitable (possibly the only) method to determine parameters for grain boundary diffusion in thin films.

References

1 T.S. Kê, *Phys. Rev., 71* (1947) 533.
2 H. Mehrer, N. Stolica and N.A. Stolwijk, in *Landolt-Börnstein, New Series,* Vol. III/26, Springer, Berlin, 1990, p. 60.
3 I. Kaur and W. Gust, *Handbook of Grain and Interface Boundary Diffusion Data*, Ziegler, Stuttgart, 1989.
4 B.S. Berry and W.C. Pritchet, *J. Phys. (Paris), Colloq. C5, 42* (1981) 1111.
5 H.G. Bohn, M. Prieler and C.M. Su, in M.A. Nastasi, D.M. Parkin and H. Gleiter (eds.), *Mechanical Properties and Deformation Behavior of Materials Having Ultra-fine Microstructures*, Kluwer, Dordrecht, 1993, p. 229.
6 H.G. Bohn and C.M. Su, *M.R.S. Symp. Proc., 239* (1992) 215.
7 F. Vollkommer, *Ph. D. Thesis*, RWTH Aachen, 1989; *Berichte der Kernforschungsanlage Jülich, Jül-2321*, 1989.
8 F. Vollkommer, H.G. Bohn, K.-H. Robrock and W. Schilling, *28th Ann. Proc. Reliability Physics, 1990*, p. 51.
9 C.M. Su and H.G. Bohn, *Mater. Sci. Forum, 119–121* (1993) 285.
10 C.M. Su, personal communication, 1991.
11 C.M. Su, H.G. Bohn, K.-H. Robrock and W. Schilling, *J. Appl. Phys., 70* (1991) 2086.
12 A.S. Nowick and B.S. Berry, *Anelastic Relaxation in Crystalline Solids*, Academic, New York, 1972.
13 R. Raj and M.F. Ashby, *Metall. Trans., 2* (1971) 1113; *Acta Metall., 23* (1975) 653.
14 H. Trinkaus and M.H. Yoo, *Philos. Mag. A, 57* (1988) 543.
15 H. Trinkaus, in preparation.