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Thermodynamics and Kinetics of Room-temperature Grain Growth in Cu Films

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Thermodynamics and kinetics of room-temperature grain growth in Cu films.

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We studied the energetics and kinetics of grain growth in Cu films, by estimating the magnitude of various possible driving forces for microstructural change that can be present in the as-deposited film. A driving force of at least 100 J/cm^3 is required to account for the speed at which the grain boundaries move at room temperature. This value cannot be accounted for by only considering the minimization of grain boundary energy and possible effects related to surface and strain energy as the main driving mechanisms. It is suggested that the additional driving force which is needed to explain the speed at which the grains recrystallize, is originating from a high density of defects within the as-deposited grains.

I. INTRODUCTION

During the last decade, room temperature grain growth or ‘self-annealing’ has been observed in electroplated Cu [1–7]. Although the grain growth behavior of electroplated Cu is intriguing, the kinetics are hard to study, since the as-deposited grain structure and impurity content of the film strongly depend on the electroplating process.

Although there is early work by Patten et al. [8] describing grain growth during the storage of sputter deposited Cu films, room temperature grain growth in PVD Cu has not been widely observed. Indeed, reports describing room temperature grain growth in electroplated films explicitly state that sputter deposited films prepared during the same study are stable at room temperature [9]. Recently, Rosnagel et al. [10] and Barnat et al. [11] showed that the resistivity of sputter deposited films ($< 100 \text{ nm}$) decreases during storage after deposition. Detavernier et al. [12] observed self-annealing in sputter deposited films up to 1500 nm in thickness, and showed that the phenomenon is very dependent on the deposition parameters (substrate temperature and sputter gas pressure) and hence on the microstructure of the as-deposited film.

In this paper, we study the energetics and kinetics of room temperature grain growth in both electroplated and sputter deposited Cu films. It will be shown that the speed at which the grains are observed to grow can only be explained quantitatively by taking into account the defect density within the as-deposited grains.

II. RESULTS

A. Phenomenology

Figure 1 demonstrates the changes that occur in the microstructure of electroplated films during storage at room temperature. The changes in the microstructure of the film cause a reduction of about 20% in sheet resistance. To explain the incubation time that is observed for electroplated films, Harper et al. [1] suggested a ripen-

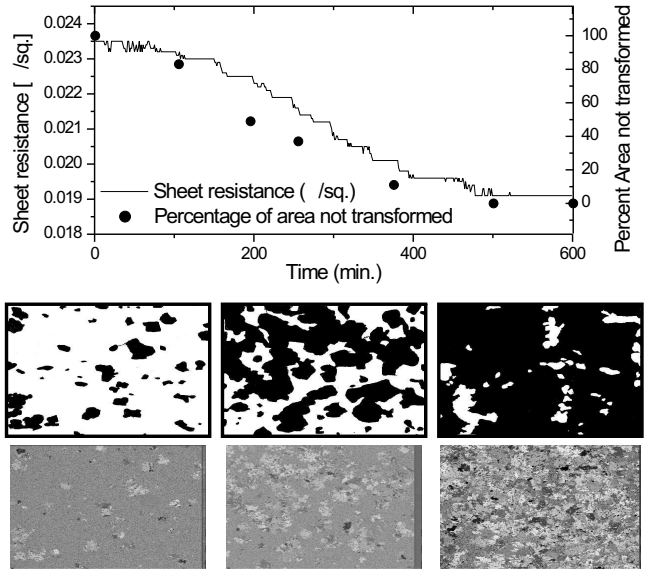


FIG. 1: In situ resistance analysis and percent area recrystallized as a function of time for a 970 nm thick electroplated film stored at 21°C , with corresponding focused ion beam images and maps showing the area of crystallization.

ing mechanism, whereby impurities that are incorporated into the film during deposition are initially pinning the grain boundaries. During storage, these impurities are assumed to coalesce, thus enabling some of the grain boundaries to move. It is known from SIMS measurements that electroplated films contain carbon and sulfur as contaminants, and thermal desorption experiments have shown that carbon-containing molecules are desorbing from the electroplated films during low temperature annealing [6]. However, the fact that self-annealing is also observed for sputter deposited Cu [10–12] suggests that although the Zener pinning mechanism may strongly influence the details of the grain growth kinetics in electroplated Cu (i.e. the incubation time), impurity pinning does not appear to be crucial to the phenomenon of room-temperature grain growth itself.

The self-annealing phenomenon is dependent on film thickness. Grain growth in thick films (e.g. several μm)

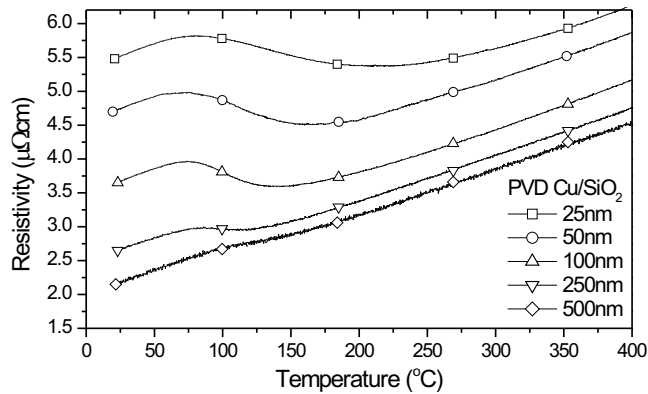


FIG. 2: Resistivity versus annealing temperature for sputter deposited films. Film thickness is indicated in the legend. The temperature was increased at a rate of $1^{\circ}\text{C}/\text{s}$.

is expected to be representative of the intrinsic bulk-like behavior, without complications from surface/interface energy or pinning by grain grooving. However, thick films are difficult to fabricate using physical vapor deposition (PVD) in view of the heating effect of the sputtering process itself and the poor thermal conduction in a vacuum environment. Indeed, if 500 nm films were sputtered at room temperature in a system that does not allow active cooling of the substrate, little room temperature grain growth occurred after deposition (fig. 2). For a PVD system that allowed active cooling of the substrate, self-annealing could still be observed for films thicker than $1\mu\text{m}$ [12].

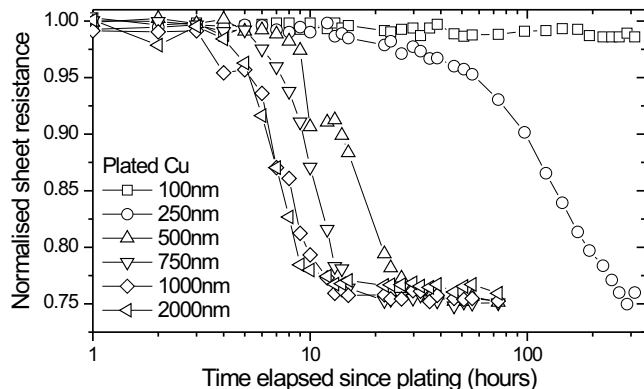


FIG. 3: Normalized sheet resistance versus storage time for electroplated films, stored at room temperature.

For electroplated films, the opposite trend is observed : the thinner the electroplated film, the slower the kinetics (fig. 3). In view of the aqueous environment, cooling is not an issue and the as-deposited film will remain defect-rich and fine-grained even for films with a thickness of several micrometers. For films thinner than 250nm, no self-annealing behavior is observed. Since the incubation

time is believed to be related to the coalescence of impurities in the electroplated films, the grains in thin electroplated films seem to be completely pinned by impurities. It may be speculated that pinning in a 2D environment (thin film) is more effective and easier to maintain than in a 3D grain structure. First, the larger number of grain boundaries per unit area for thicker films will allow easier redistribution of the pinning particles along the grain boundaries. Second, although pinning may persist at some locations along the boundaries, grains are able to grow around these pinning positions in a 3D environment.

B. Kinetics

Kissinger analysis was performed to study the grain growth kinetics, by measuring the sheet resistance in-situ while ramping the temperature at a constant rate. If one performs this experiment for a wide range of different ramp rates, it is possible to determine an activation energy [15]. Both sputter deposited films (thickness 100nm, 99.9999% Cu target, base pressure 10^{-8} Torr, sputtered onto Si(100) substrates covered with a native oxide, capped in-situ with a 5nm Ta layer to prevent surface oxidation) and electroplated films (970 nm) were studied during the experiments. Annealing was done in purified He, at rates between 0.01 and $10^{\circ}\text{C}/\text{s}$. The resistance was measured in-situ during annealing (figure 4). The sudden decrease in resistance indicates a microstructural change in the Cu film which increases the mean free path of the electrons. For higher ramp rates, the decrease in resistance shifts to higher temperatures, indicating that the microstructural change is thermally activated. Using the Kissinger analysis, we obtain an activation energy of 0.9-1 eV for both the sputter deposited Cu and the electroplated Cu. Assuming a 5°C error in the measured temperature, the error on this value is 0.2 eV.

III. DISCUSSION

A. Terminology

In several papers discussing the microstructural evolution of electroplated films during storage at room temperature, different authors have used different terminology : abnormal grain growth, grain growth and recrystallization. The use of these terms may be quite confusing, especially compared to the well-defined terminology used in metallurgical literature. After cold work (e.g. rolling or peening), the microstructure of a bulk material contains a lot of defects. During annealing, several processes are known to take place within the deformed microstructure [13] :

(a) Recovery consists of the annihilation of point defects and dislocations and the spontaneous re-

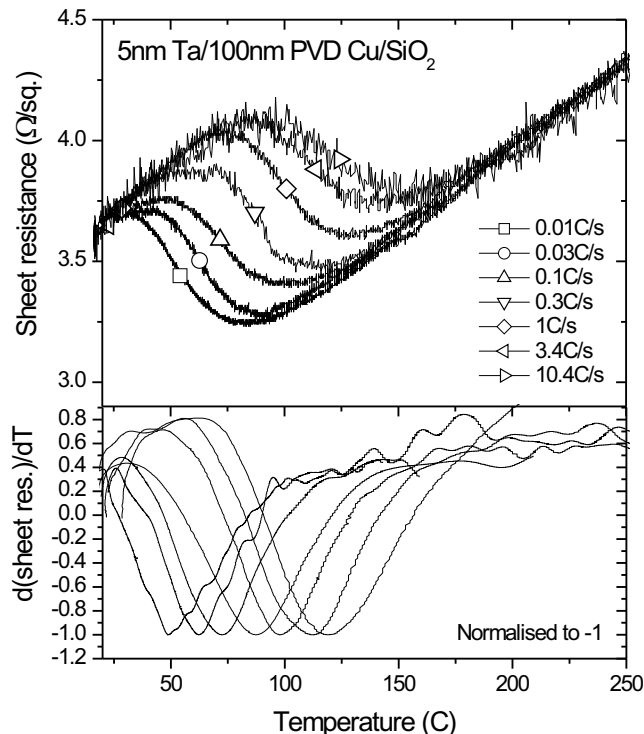


FIG. 4: In-situ sheet resistance versus annealing temperature for 100nm films sputtered onto SiO_2 and capped with a 5nm Ta layer. The rate at which the temperature of the sample was increased is indicated in the legend.

arrangement of dislocations into low-angle sub-grain boundaries (polygonization).

(b) Recrystallization is the formation of large, defect-free grains that are separated from the defected regions by a high-angle grain boundary. The driving force for recrystallization is the stored energy in the form of point (vacancies, interstitials), line (dislocations) and planar (stacking faults) defects within the deformed grains, and is typically of the order of 100 J/cm^3 .

(c) Grain growth consists of an increase of the mean radius of the recrystallized (i.e. defect-free) grains. The driving force in this case is the minimization of total grain boundary energy in the system (typical values are 10 J/cm^3).

(d) For some materials and sample geometries, secondary recrystallization (or abnormal grain growth) has been observed. This process is characterized by a bimodal grain size distribution. Certain grains are believed to be able to grow much faster than the average grain because they either have a boundary with a very large mobility or because they experience an additional driving force for grain growth (e.g. minimization of surface and/or strain energy in addition to the minimization of grain boundary area).

In recent years, these well-known mechanisms for microstructural evolution in bulk samples have been used to explain microstructural evolution in thin films [16–18].

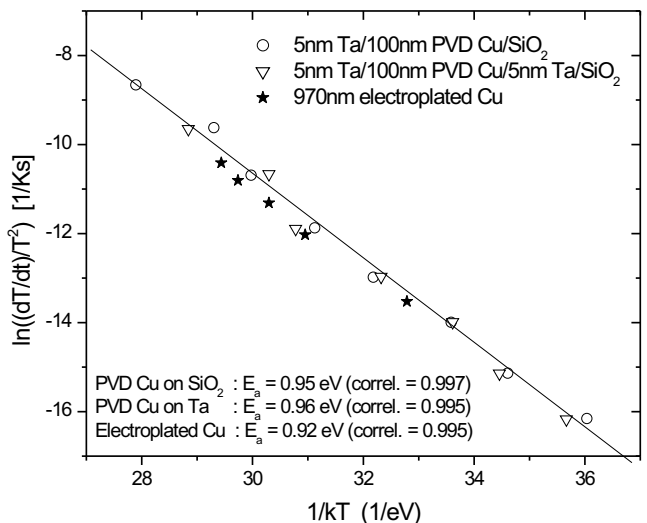


FIG. 5: Kissinger-type analysis of the sheet-resistance data measured during annealing treatments at different ramp rates. The samples are 100nm sputter deposited films on SiO_2 substrates (with and without 5nm Ta interlayer) and 970nm electroplated films. The sheet resistance data for the 5nm Ta/100nm Cu/ SiO_2 sample is given in figure 4.

It is assumed that the driving force consists of a summation of a term related to the curvature driven (normal) grain growth, a term related to the difference in elastic [19] or plastic [17] strain energy between two neighboring grains, and a term related to the difference between surface energies of neighboring grains [20]. It is usually assumed that a ‘pinning force’ caused by particles or impurities that are present on the grain boundaries (Zener pinning) may oppose all of the above mentioned driving forces.

B. Room temperature grain growth in Cu

Microstructural evolution (such as grain growth, texture changes, ...) typically involves the movement of grain boundaries within the material. The velocity of a grain boundary is given by $v = M \cdot \Delta G$ with M the mobility of the grain boundary, and ΔG the driving force [13]. In spite of its widespread use in literature, it is worthwhile to explicitly state the assumptions underlying this equation. Suppose that a chemical potential difference ΔG exists between the atoms in two neighboring grains A and B, and that there is an energy barrier E_a for an atom to make the transition across the grain boundary. The probability for an atom to move from A to B will be proportional to $\exp(-E_a/kT)$, while the probability to move from B to A will be proportional to $\exp(-(E_a + \Delta G)/kT)$. The net transport (and therefore the velocity v of the boundary) will therefore be proportional to $\exp(-E_a/kT) \cdot (1 - \exp(-\Delta G/kT))$. If one

assumes that $\Delta G \ll kT$, one can approximate this to

$$v = (M_0 \exp(-E_a/kT)/kT) \cdot \Delta G = M \cdot \Delta G. \quad (1)$$

A more elaborate version of the above derivation was published by Gleiter et al. [21].

In order to assess the mechanism underlying the room temperature grain growth in Cu films, we start from the experimental kinetics data, which indicates an increase in grain size of about 1 μm in typically 5-10 hours. This means that grain boundaries in the film are moving at a speed v of the order of 100-200nm/hour. The driving force ΔG for microstructural evolution is given by the amount of stored energy (in the form of defects, grain boundaries, strain, etc.) that is present in the metastable as-deposited film. We now make estimates for the stored energy density of each of these types of defects (see table I).

(1) A first contribution results from the energy that can be gained by eliminating grain boundary area in the film. We calculate an upper limit for this driving force, assuming that all grain boundaries in the as-deposited film are high-angle grain boundaries with a grain boundary energy $\gamma_{\text{HA}} = 0.625 \text{ J/m}^2$ [24]. If the mean grain radius is R , the energy stored in grain boundaries is given by

$$E_{\text{GB}} = \frac{3\gamma}{2R}. \quad (2)$$

(2) A second type of planar defects is stacking faults within the as-deposited grains. If we assume that a stacking fault occurs on the average every n lattice planes, the stored energy is

$$E_{\text{SF}} = \frac{\gamma_{\text{SF}}}{nd_{111}}, \quad (3)$$

with $\gamma_{\text{SF}} = 0.078 \text{ J/m}^2$ the stacking fault energy [13] and $d_{111} = 0.208 \text{ nm}$ the distance between two (111) planes. Introducing a large number of stacking faults will eventually (for $n = 3$) result in the formation of hexagonal Cu.

(3) Line defects (dislocations) within the as-deposited grains are a third possible source of stored energy. The strain energy associated with one dislocation may be estimated by $\frac{1}{2}Gb^2$, with $G = 4.21 \cdot 10^{10} \text{ N/m}^2$ and $b = 0.256 \text{ nm}$ the Burger's vector. For an areal density ρ of dislocations, the stored strain energy is thus given by

$$E_{\text{disl}} = \frac{1}{2}\rho Gb^2. \quad (4)$$

(4) One may also estimate the driving forces for grain growth caused by anisotropic surface and strain energies. The (111) plane has the lowest surface energy for Cu ($\gamma_{111}/\gamma_{100} = 0.994$, $\gamma_{110}/\gamma_{100} = 1.011$ and $\gamma_{\text{max}}/\gamma_{100} = 1.015$ [19]). The additional driving force for a (111) grain to grow at the expense of a grain with the maximal surface energy is given by $E_{\text{surf}} = -2\Delta\gamma/h$, with h the layer thickness and $\Delta\gamma = \gamma_{111}(1 - 1.015/0.994)$.

(5) The stress σ at a temperature T induced by a thermal expansion mismatch $\Delta\alpha$ between film and substrate is given by $\sigma = M\Delta\alpha(T - T_0)$, with the biaxial modulus M dependent on the components of the tensor of elastic stiffness. Anisotropy in either the thermal expansion and/or the elastic properties of the film will result in a dependence of the elastic strain energy density $\sigma\epsilon/2$ on the orientation of the grain on the substrate. The elastic strain energy density within a grain with its (hkl) direction parallel to the surface can be calculated using

$$F(h, k, l) = c_{11} + c_{12} + X(h, k, l) - \frac{2(c_{12} - X(h, k, l))^2}{c_{11} + 2X(h, k, l)}$$

with $X(h, k, l) = (c_{12} + 2c_{44} - c_{11}) \cdot (h^2k^2 + k^2l^2 + h^2l^2)$ and $c_{11} = 168.4 \text{ MPa}$, $c_{12} = 121.4 \text{ MPa}$, $c_{44} = 75.4 \text{ MPa}$ [19]. The maximum driving force caused by a difference in elastic energy between two grains is then given by $E_{\text{strain}} = \epsilon^2(F(1, 1, 1) - F(1, 0, 0))$, with ϵ the strain [23].

(6) Finally, strain energy in the regime of plastic deformation may also provide a driving force for grain growth in thin films. For instance, although the elastic stress in Al is isotropic, the yield stress depends on grain orientation [17]. However, for the present case of room temperature storage of Cu films, the thermal stress is zero, since all depositions were done at room temperature and there is no post-deposition anneal. Based on wafer curvature measurements, the intrinsic stress within the film is known to be between -50 and 150 MPa. Therefore, one can safely assume that none of the grains within the film are ever subjected to sufficiently high stresses to reach the regime of plastic deformation.

For completeness, we also included the Zener pinning mechanism in table I, because of its importance in electroplated films. The driving force for Zener pinning (actually the 'pinning force') is given by $E_{\text{Zener}} = -\frac{3\gamma\rho_p}{2r}$, with ρ_p the volume fraction of pinning particles and r their radius. In table I, the values for these parameters were chosen to illustrate that the pinning force related to the Zener mechanism may actually balance the driving force that originates from eliminating grain boundary area.

Recalling equation 1, grain growth kinetics is not only determined by the driving force ΔG , but also by mobility M . A detailed study was performed on grain boundary mobility in bulk Cu by Viswanathan et al. [24]. Their data are reproduced in figure 6. They studied the interface mobility in Cu bicrystals that were grown with different misorientations (2, 5, 9, 18, 32°). For a given orientation, they studied both pure tilt-type boundaries and mixed (tilt/twist) boundaries. Since we are only interested in an upper estimate of the grain boundary mobility, we only consider the fastest moving boundaries, which are mixed-type boundaries with the highest degree of misorientation. Viswanathan et al. measured a mobility of $0.02083 \text{ cm}^4/\text{Js}$ at 575°C . One can extrapolate

TABLE I:

Mechanism	Equation	Assumption	ΔG in J/cm ³
Grain boundaries	$E_{GB} = \frac{3\gamma}{2R}$	$R = 50\text{nm}; \gamma = 0.625\text{J/m}^2$	≈ 20
Stacking faults	$E_{SF} = \frac{\gamma_{SF}}{nd_{111}}$	$n = 3 - 20; \gamma_{SF} = 0.078\text{J/m}^2$	$\approx 125 - 20$
Dislocations	$E_{disl} = \frac{1}{2}\rho Gb^2$	$\rho = 10^{12} - 10^{13}/\text{cm}^2$	$\approx 15 - 140$
Surface energy	$E_{surf} = -2\Delta\gamma/h$	$h = 500\text{nm}$	≈ 0.05
Elastic strain	$E_{strain} = \epsilon^2(F(1, 1, 1) - F(1, 0, 0))$	$\epsilon = 0.2\%$	≈ 2
Zener pinning	$E_{Zener} = -\frac{3\gamma\rho_p}{2r}$	$\rho_p = 0.2\%; r = 0.1\text{nm}$	≈ -20

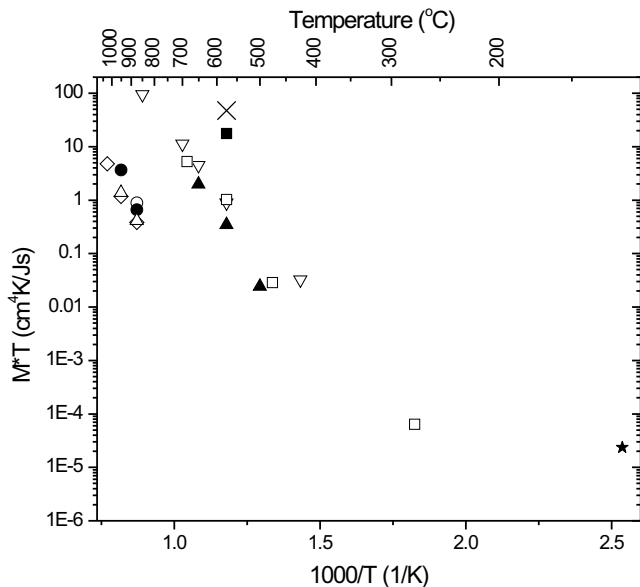


FIG. 6: Grain boundary mobility in bulk Cu as a function of temperature. The data is taken from Viswanathan et al. [24] and Vandermeer et al. [25]. The data from Viswanathan was measured on bicrystals with misorientations of 2° (\circ), 5° (\diamond), 9° (Δ), 18° (∇) and 32° (\square), for pure tilt-type boundaries (open symbols) and mixed tilt/twist boundaries (solid symbols). The data from Vandermeer (\star) was measured for high angle grain boundaries during recrystallization at 121°C. The 'x' indicates the extrapolation of the value measured by Vandermeer to the measurement temperature of 575°C used in the work of Viswanathan, using eqn. 5 with $E_a=0.92\text{eV}$.

this value to other temperatures :

$$M(T) = M(T_0) \cdot \exp\left(-\frac{E_a}{k} \cdot \left(\frac{1}{T} - \frac{1}{T_0}\right)\right) \cdot \frac{T_0}{T}. \quad (5)$$

A recent paper by Vandermeer et al. [25] provides an in depth study of grain boundary mobility during recrystallization of cold-deformed Cu. They measured a mobility of $6.31 \cdot 10^{-8} \text{ cm}^4/\text{Js}$ at 121°C. If we use the known activation energy of 0.92 eV for grain boundary self-diffusion [26] and extrapolate the mobility measured by Vandermeer at 121°C to the measurement temperature of 575°C used in the work of Viswanathan, we obtain $0.0557 \text{ cm}^4/\text{Js}$ (point indicated by 'x' in figure 6),

a value within a factor of 3 from the measured value by Viswanathan. The fact that the known bulk mobilities in Cu (measured by two different groups and different techniques) are self-consistent and that the measured values can be extrapolated over more than 450°C using the activation energy for grain boundary self-diffusion, indicates that we can use the same activation energy to extrapolate the high temperature mobility values reported in literature to room temperature. Extrapolation of the value measured by Vandermeer from 121°C to 20°C results in a room-temperature mobility of $7.1 \cdot 10^{-8} \text{ cm}^4/\text{Js}$. The importance of the mobility M suggests that more experimental work is needed on this topic.

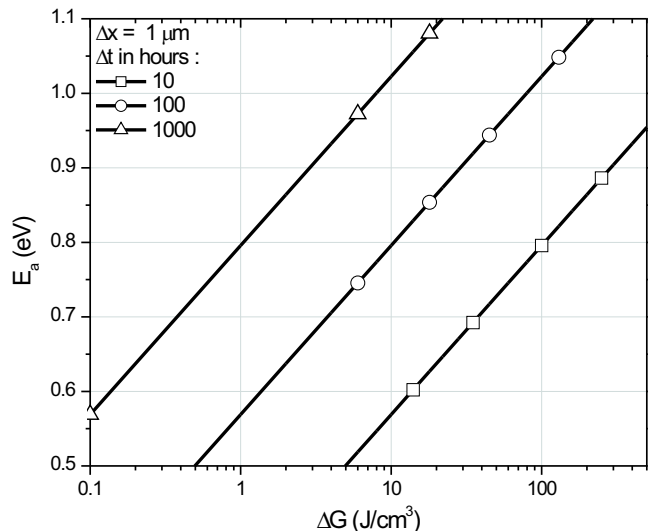


FIG. 7: Sets of solutions $(E_a, \Delta G)$ of $v = M(20^\circ\text{C}) \cdot \Delta G$ for different values of $v = \Delta x/\Delta t$, with $M(20^\circ\text{C}) = M(294\text{K}) = M(121^\circ\text{C}) \cdot \exp\left(-\frac{E_a}{k} \cdot \left(\frac{1}{294\text{K}} - \frac{1}{394\text{K}}\right)\right) \cdot \frac{394\text{K}}{294\text{K}}$ with $M(121^\circ\text{C}) = 7.08 \cdot 10^{-8} \text{ cm}^4/\text{Js}$ the measured value for the mobility of high angle grain boundaries at 121°C.

If we use $7.1 \cdot 10^{-8} \text{ cm}^4/\text{Js}$ as an estimated value for the mobility M of high-angle grain boundaries at room temperature, we can estimate the driving force ΔG which is necessary to obtain a velocity v of about $1\mu\text{m}$ in 10 hours for self-annealing. One obtains $\Delta G = v/M = 392 \text{ J/cm}^3$ or 2.79 kJ/mole . We checked the sensitivity of the resulting ΔG on the assumptions that are used during

the extrapolation of the known bulk mobility to room-temperature. Figure 7 illustrates that for E_a close to 0.9 eV, the driving force $\Delta G > 100 \text{ J/cm}^3$. This value is clearly bigger than the typical driving force of 10 J/cm^3 expected for curvature driven ‘grain growth’ as the term is defined in metallurgical textbooks.

The speed at which the grain boundaries are moving can only be explained quantitatively by taking into account the defect density in the as-deposited film. Defects within the as-deposited grains may affect the growth kinetics in two ways. Firstly, the presence of stacking faults and dislocations can provide the necessary driving force ΔG (table I). A high defect density within the as-deposited film is energetically similar to the presence of certain alloying elements in Cu. It has been observed that the grain size for Cu-Co and Cu-Ag alloys is considerably larger than that of pure Cu after the same annealing treatment [29]. If the as-deposited film is oversaturated with the alloying element, the element will precipitate at the grain boundaries during grain growth. The precipitation energy that is continuously released while the growing grain sweeps through the Cu alloy constitutes a significant driving force for grain boundary migration. Similarly, the movement of a grain boundary that is sweeping through a defect-rich region in the film will also be driven by the energy that is released by annihilating the defects. Secondly, defects that are built into the film could increase the mobility M . Our calculations for the mobility assume a thermally activated process. However, it is known that diffusion can be significantly enhanced if the density of point defects is higher than the equilibrium value at a given temperature. A non-equilibrium concentration of point defects can be achieved by non-thermal processes such as irradiation or ion bombardment [27, 28]. It is conceivable that defects that get quenched into the growing film during the low-temperature deposition process could play a similar role in Cu films.

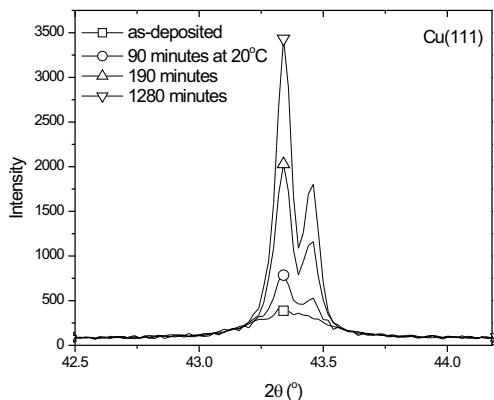


FIG. 8: X-ray diffraction showing the Cu(111) peak for a 970nm thick electroplated film. The time during which the film was stored at room temperature is indicated in the legend.

The fact that the defect density within the film is indeed decreasing during storage is illustrated by the XRD measurements in fig. 8. The Cu(111) peak becomes much narrower and undergoes a splitting into $\text{CuK}\alpha 1$ and $\text{CuK}\alpha 2$ components. The peak broadening in the as-deposited film is related to the small grain size and strain caused by defects within the grains. The very distinct peak splitting for the film after storage for about three hours can only be explained if the grains in the aged Cu film have a very low defect density.

C. Microstructural changes at room temperature in thin films

One may wonder why the room temperature ‘self-annealing’ behavior has only been reported for Cu films. A clue may be provided by the fact that Cu (like Ag and Au) has a low stacking fault energy, resulting in slow recovery kinetics, since dislocations are not able to annihilate through cross-slip. During calorimetry measurements on Cu that had been subjected to torsion at 77K, a first peak occurs below 273K, indicating the disappearance of point defects, while the peak indicating the recovery of dislocations only occurs at 350K [13].

The above suggests that room temperature grain growth will occur in materials that are deposited using a low temperature deposition process (either electroplating or sputter deposition onto a cooled substrate), (1) if the as-deposited microstructure contains a lot of defects (grain boundaries, dislocations, stacking faults, ...) caused by the low temperature deposition process, and (2) if the material has a low stacking fault energy, limiting the effectiveness of intra-grain recovery to cleanse the grains from those defects.

In spite of the technological importance of metal films, relatively little quantitative work has been reported regarding the defect density of as-deposited films. For sputter deposition, the ‘zone-model’ [30] describes the effect of the deposition conditions on the microstructure of the as-deposited film. The substrate temperature determines the surface mobility on the growing film, while the sputter gas pressure determines the mean free path in the plasma, thus affecting the exposure of the growing film to bombardment by energetic particles. At low temperature and high sputter gas pressure (no ion bombardment), a porous zone 1 structure is formed because of the low surface mobility and self-shadowing during deposition. Ion bombardment causes the formation of a densified zone-T film. At higher substrate temperature (zone 2, typically for $T > 30\%$ of the melting point), the atoms arriving at the surface have sufficient mobility to avoid the formation of a porous structure, even in the absence of ion bombardment. Electroplating is a more complex process, and a multitude of parameters are needed to describe the plating conditions. To the best of our knowledge, no general microstructural model has been developed for electroplated films. However, plating from aqueous solu-

tions is an intrinsically low-temperature process. Without the appropriate additives, plating involves dendritic growth, which results in a porous microstructure. Additives are used to modify the reaction kinetics at the surface of the growing film, in order to densify the film. In summary, at low substrate temperature, both sputter deposition and electroplating result in a porous (zone 1) type microstructure. By taking the necessary precautions (i.e. ion bombardment during sputter deposition or additives for electroplating), a zone-T type microstructure is formed. Experiments using positron annihilation spectroscopy and differential scanning calorimetry could provide valuable insight in the nature and density of the defects that are present in the as-deposited film. Based on the results reported above, the microstructure of such films seems to resemble the microstructure of cold-worked bulk metals [31].

D. Abnormal grain growth in thin films

We conclude this paper with a brief remark regarding abnormal grain growth during high-temperature annealing of thin films. Abnormal grain growth is generally assumed to arise from a combination of the inhibition of normal grain growth (either by grain boundary grooving and/or impurity pinning) and the occurrence of a small number of special grains in the film that have a lower free energy, providing a net driving force for their growth. The resulting texture is claimed to be dependent on the driving force which selects the ‘special’ grains. For the case of Cu, it has been argued that a (111) fiber texture should develop for surface energy driven growth, (100) fiber texture for strain energy driven growth in the elastic regime and (110) fiber texture for strain energy driven growth in the plastic regime [18].

Abnormal growth implies that one type of grains has exceptionally fast-moving boundaries. Since $v = M \cdot \Delta G$, there are two possible causes: (1) a special driving force ΔG for selected grains (thermodynamics-based selection) and/or (2) a difference in M for different grain boundaries (kinetics-based selection). For Cu, the data by Viswanathan et al. [24] in figure 6 show that the difference in mobility between a low angle grain boundary and a random boundary is several orders of magnitude. Suppose that a Cu film has a strong (111) fiber texture, resulting in a film consisting of mostly low-angle grain boundaries. Even if the mechanism providing the driving force for grain growth is identical for all grains, the high mobility of the high angle grain boundaries surrounding a (100) grain that is embedded within a matrix of (111) grains will cause a sufficiently large (100) grain to grow rapidly and consume the surrounding matrix of (111) grains. It is certainly not our intention to claim that

all abnormal grain growth occurs by a kinetics-based selection mechanism, but only to point out that differences in mobility may be significant, especially in textured materials [32–35].

IV. CONCLUSION

- Since the microstructural evolution during annealing of thin films is not fundamentally different from bulk samples, one should use the existing terminology from the field of bulk metallurgy (i.e. recovery - recrystallization - grain growth - abnormal grain growth).
- Contrary to popular belief, the self-annealing phenomenon for Cu is not a unique consequence of the electroplating process. Although there are differences in the details of the kinetics, the overall behavior is similar for both electroplated and sputter deposited films.
- Although impurity pinning may strongly influence the kinetics (e.g. the incubation time), Zener pinning by impurities from the plating bath is not crucial to the phenomenon of self-annealing.
- If M is extrapolated from bulk experiments, the driving force ΔG needed to explain the velocity $v = M \cdot \Delta G$ at which the grain boundaries move is $> 100 \text{ J/cm}^3$, suggesting that a high density of dislocations and/or stacking faults is the primary driving force for recrystallization.
- Since $v = M \cdot \Delta G$, abnormally fast growth of a certain type of grains during high temperature annealing of a material may have two origins: either the mobility M (kinetic selection rule) or the driving force ΔG (thermodynamic selection rule) is larger than for the average grain. Papers discussing abnormal grain growth in thin films usually focus on differences in ΔG , while tacitly assuming that M is identical for all grain boundaries. Especially for textured films, this assumption may be invalid.

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