Colloque 3M *Matériaux, Mécanique, Microstructures*, sur le thème *Interfaces : de l'atome au polycristal*, édité par O. Hardouin Duparc, CEA Saclay, INSTN, 14–15 juin 2006, pp. 141–153.

Representative Volume Element sizes for copper bulk polycrystals and thin layers

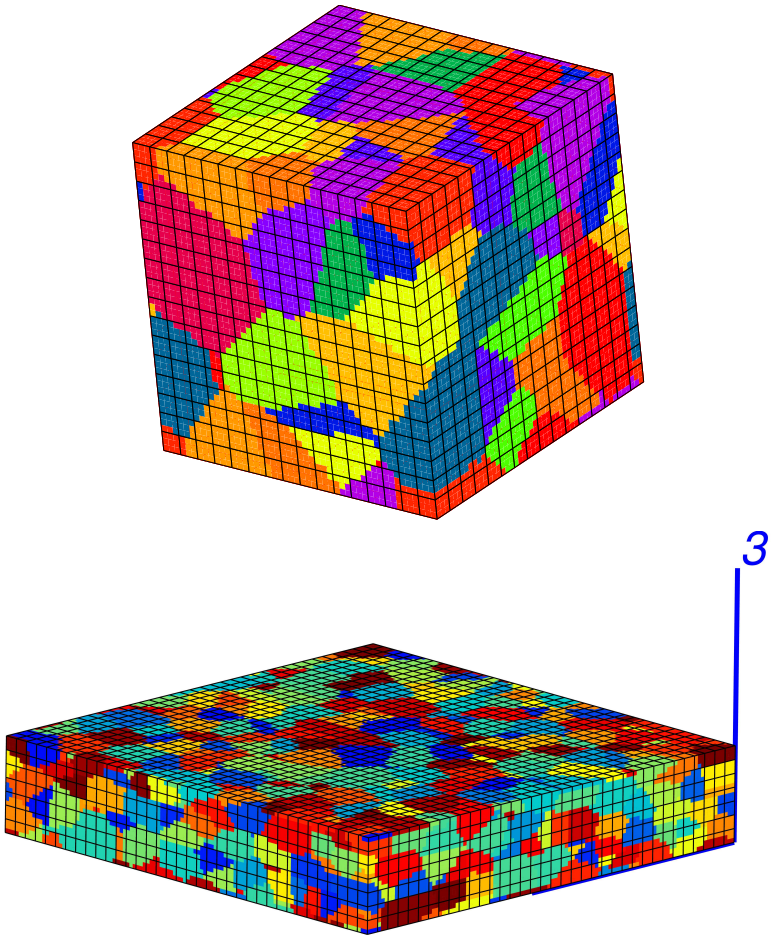
F. El Houdaigui, S. Forest, A.–F. Gourgues, D. Jeulin
Centre des Matériaux, Mines Paris, Paristech, CNRS UMR 7633, BP 87, 91003 Evry Cedex, France
Tél.: +33-(0)1-60-76-30-51, fax: +33-(0)1-60-76-31-60
samuel.forest@ensmp.fr

Abstract: This work addresses the question of the existence and determination of a Representative Volume Element (RVE) for the mechanical properties of thin polycrystalline metal structures. A computational homogenization strategy is developed to determine the number of grains necessary to estimate the effective elastic properties of isotropic polycrystalline copper with a given precision. Finite element simulations of polycrystalline aggregates are presented, using homogeneous and periodic boundary conditions. For each volume size, several realizations are considered. The mean apparent shear modulus and the associated scatter are estimated as a function of the number of grains. Two types of microstructures are considered: thin polycrystalline sheets with a fixed number of grains within the thickness and bulk polycrystals as a reference. For one–grain–thick polycrystalline plates, it is shown that effective properties cannot be defined unambiguously. For 3–grain–thick polycrystalline plates, the bulk properties are recovered, provided that a sufficient number of grains are considered along the in–plane directions. The size of the RVE Element is related to the concept of integral range.

1 INTRODUCTION

Computational homogenization methods are efficient tools to estimate effective properties of heterogeneous materials. They can take realistic distributions of phases and sophisticated constitutive equations of the constituents into account [1]. A key–point in such models is the determination of the appropriate size of volume elements of heterogeneous materials to be computed in order to get a precise enough estimation of effective properties. This is related to the long–standing problem of the determination of the size of the Representative Volume Element (RVE) in homogenization theory.

The present work raises the question of the existence and determination of the RVE in the case of thin polycrystalline structures (metal sheets, plates, layers, films...). For that purpose, the apparent mechanical properties of volumes of increasing sizes are compared in the case of polycrystalline thin sheets but also bulk polycrystals taken as a reference. The statistical and numerical methodology proposed in [2] is used to estimate the size of a RVE in isotropic linear elastic copper polycrystals. The method follows three main steps: the choice of a random model for polycrystalline microstructures containing a finite number of grains; the resolution of boundary value problems on such polycrystalline aggregates of increasing sizes; the analysis of the convergence of the calculated apparent properties towards an asymptotic value as a function of the number of grains and of the boundary conditions. The asymptotic value is regarded as the effective property [3]. In other words, the objective is to find the minimum number of grains required in a volume element to estimate the effective elastic property with a given accuracy. The size of the RVE for several cubic elastic bulk polycrystals was investigated in [4] using three–dimensional FE simulations and periodic boundary conditions. A relationship between the RVE size and the anisotropy coefficient of each material was identified. The author links the notion of representativity of considered material volumes with the decay of the scatter in the calculated apparent properties for increasing grain numbers, as done in [2]. The present contribution lays the stress on the



(a)

(b)

Figure 1: Regular FE mesh superimposed on Voronoi mosaics using the multiphase element technique: (a) cube containing 50 grains with periodicity constraint at the boundaries, (b) layer with 3 grains within the thickness containing 657 grains, with in–plane periodicity constraint. Arbitrary colors are attributed to the grains.

dependence of the result on the choice of boundary conditions and the determination of a statistical parameter quantifying the decrease in scatter with increasing grain number. Such a parameter makes it possible to compare RVE sizes for other microstructures and properties.

Thin polycrystalline materials have only a limited number of grains through the thickness, in contrast to bulk polycrystals. Such microstructures are increasingly encountered in micro–electromechanical systems (MEMS) but also in coatings and layered microstructures [5]. The existence of effective properties and their determination can be useful for subsequent structural computations, since one usually cannot afford considering all grains in the simulations. The proposed methodology for the determination of effective properties is applied to copper thin plates with a fixed number of grains through the thickness. The lateral dimensions of the plate are then increased in order to determine the wanted RVE size, if it exists. The possible bias induced by the boundary conditions in the computation must be carefully investigated. The determination of effective properties in textured copper thin films on a substrate was tackled in [6] but the question of the existence of a RVE and the effect of boundary conditions were not addressed.

All simulations are carried out for linear elastic copper polycrystals with a uniform distribution of crystal orientations, leading to an isotropic texture in both bulk and thin polycrystals. The cubic elasticity constants of pure copper at 300K are taken from [7]: $C_{11} = 168400$ MPa, $C_{12} = 121400$ MPa, $C_{44} = 75390$ MPa. The corresponding value of the anisotropy coefficient $a = 2C_{44}/(C_{11} - C_{12})$ is 3.2.

In the following, vectors are underlined and boldface quantities are second–rank or fourth–rank tensors. The symbol := defines the quantity on the left of the symbol.

2 COMPUTATIONAL HOMOGENIZATION METHOD

2.1 Generation of microstructures

Voronoi mosaics are used here as a random model to represent the polycrystalline morphology, as explained in [2]. For each realization, one given volume V (a cube or a plate with fixed thickness) that contains a given number N_g of Voronoi cells is simulated. In the following, n realizations of volume V are considered. The number of cells for each realization of the microstructure obeys a Poisson distribution with given mean value $\bar{N}_g = N$. The average volume of one Voronoi cell is equal to 1. No unit length is introduced because the models involved in this work cannot account for absolute size effects. As a result, one has $N = V$. A crystal orientation is attributed to each Voronoi cell which is then regarded as an individual grain of the polycrystal. The crystallographic texture is assumed to be uniformly random. It is possible to impose a geometrical periodicity constraint at the boundary of the polycrystalline cube or thin structures, as shown in figures 1 (see also [2]). This condition is enforced in the subsequent FE simulations involving periodicity conditions, if not otherwise stated. It results in a slight decrease of the dispersion of the apparent properties when compared to simulations relying on the initial Voronoi model.

2.2 FE meshing of microstructures and parallel computing

The so–called multi–phase element technique is used in order to superimpose a regular 3D FE mesh on the Voronoi tessellation. The crystal orientation of the closest voxel is attributed to every integration point of each element of the mesh [2]. The elements are 20–node quadratic bricks with 27 Gauss points. Figures 1(a) and (b) show such meshes made of 16x16x16 and 7x36x36 elements respectively. The effect of mesh density, i.e. of the number of elements per grain, on apparent shear modulus μ^{app} was investigated for an elastic polycrystalline volume, containing 50 grains, when the number of finite elements is increased. For each simulation, the geometry of the microstructure is unchanged but the number of degrees of freedom, namely, the unknown displacement components, was changed from 5568 to 56355. From these results, a resolution of 16 elements per grain was chosen for the following calculations.

The largest volume computed in this work is a cube with $42^3 = 74088$ elements, i.e. 937443 degrees of freedom. Such computations are made possible in a reasonable time by using parallel computing. The FE program used in this work implements the subdomain decomposition method FETI [8]. The mesh is split into 32 subdomains and the tasks are distributed on a platform of 32 processors (768 MB RAM, 800 MHz). Compatibility and equilibrium at interfaces between subdomains are restored by an iterative procedure. The whole resolution requires 21 GB of memory.

2.3 Boundary conditions and definition of apparent moduli

Three types of boundary conditions to be prescribed on an individual volume element V are considered:

- *Kinematic uniform boundary conditions* (KUBC): The displacement vector $\underline{\boldsymbol{u}}$ is imposed at all points $\underline{\boldsymbol{x}}$ belonging to the boundary ∂V according to:

$$\underline{\boldsymbol{u}} = \boldsymbol{E}.\underline{\boldsymbol{x}} \quad \forall \underline{\boldsymbol{x}} \in \partial V \quad \Longrightarrow \quad <\boldsymbol{\varepsilon}> := \frac{1}{V}\int_V \boldsymbol{\varepsilon}\, dV = \boldsymbol{E} \tag{1}$$

  where $\boldsymbol{E}$ is a given constant symmetrical second–rank tensor. The macroscopic stress tensor $\boldsymbol{\Sigma}$ is then defined as the spatial average of the local stress tensor $\boldsymbol{\sigma}$.

- *Static uniform boundary conditions* (SUBC): The traction vector is prescribed at the boundary ∂V according to:

$$\boldsymbol{\sigma}.\underline{\boldsymbol{n}} = \boldsymbol{\Sigma}.\underline{\boldsymbol{n}} \quad \forall \underline{\boldsymbol{x}} \in \partial V \quad \Longrightarrow \quad <\boldsymbol{\sigma}> := \frac{1}{V}\int_V \boldsymbol{\sigma}\, dV = \boldsymbol{\Sigma} \tag{2}$$

where $\boldsymbol{\Sigma}$ is a given constant symmetrical second–rank tensor. The outer normal to ∂V at $\underline{\boldsymbol{x}}$ is denoted by $\underline{\boldsymbol{n}}$. The macroscopic strain tensor $\boldsymbol{E}$ is then defined as the spatial average of the local strain $\boldsymbol{\varepsilon}$.

- *Periodicity conditions* (PERIODIC): The displacement field over the entire volume V takes the form

$$\underline{\boldsymbol{u}} = \boldsymbol{E}.\underline{\boldsymbol{x}} + \underline{\boldsymbol{v}} \quad \forall \underline{\boldsymbol{x}} \in V \tag{3}$$

where the fluctuation $\underline{\boldsymbol{v}}$ is periodic. $\underline{\boldsymbol{v}}$ (resp. $\boldsymbol{\sigma}.\underline{\boldsymbol{n}}$) takes the same value (resp. opposite values) at two homologous points on opposite sides of V.

The local behaviour at every integration point inside each grain in the simulation is described by the fourth–rank linear elasticity tensor $\boldsymbol{c}$:

$$\boldsymbol{\sigma}(\underline{\boldsymbol{x}}) = \boldsymbol{c}(\underline{\boldsymbol{x}}) : \boldsymbol{\varepsilon}(\underline{\boldsymbol{x}}) \tag{4}$$

For a given volume V, and owing to the linearity of the considered boundary value problems, fourth–rank tensors of apparent moduli $\boldsymbol{C}_E^{app}$ and apparent compliances $\boldsymbol{S}_\Sigma^{app}$ can be defined by the following macroscopic relations:

$$\boldsymbol{\Sigma} = <\boldsymbol{\sigma}> = \frac{1}{V}\int_V \boldsymbol{\sigma}\, dV = \boldsymbol{C}_E^{app} : \boldsymbol{E}, \quad \boldsymbol{E} = <\boldsymbol{\varepsilon}> = \frac{1}{V}\int_V \boldsymbol{\varepsilon}\, dV = \boldsymbol{S}_\Sigma^{app} : \boldsymbol{\Sigma} \tag{5}$$

The first relation is used for KUBC and PERIODIC problems, the second one for SUBC problems. Note that in general, the tensor $\boldsymbol{S}_\Sigma^{app}$ cannot be expected to coincide with the inverse of $\boldsymbol{C}_E^{app}$. However, for sufficiently large volumes V (along all three directions of space), the apparent moduli do not depend on the type of boundary conditions any longer and coincide with the effective properties of the medium [3]:

$$\boldsymbol{S}_\Sigma^{app-1} = \boldsymbol{S}^{eff-1} = \boldsymbol{C}^{eff} = \boldsymbol{C}_E^{app} \tag{6}$$

For intermediate volumes V, the following inequalities, written in the sense of quadratic forms, hold [9]:

$$\boldsymbol{S}_\Sigma^{app-1} \leq \boldsymbol{C}^{eff} \leq \boldsymbol{C}_E^{app} \tag{7}$$

In the next sections, both $\boldsymbol{C}^{eff}$ and the periodic estimations are checked to remain between the bounds defined by (7).

The following two shear loading conditions $\boldsymbol{E}_\mu$ and $\boldsymbol{\Sigma}_\mu$ are used in this work:

$$\boldsymbol{E}_\mu = \begin{bmatrix} 0 & \frac{1}{2} & 0 \\ \frac{1}{2} & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad \boldsymbol{\Sigma}_\mu = \begin{bmatrix} 0 & a & 0 \\ a & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \quad \text{with} \quad a = 1\,\text{MPa} \tag{8}$$

in the particular Cartesian coordinate frame attached to the cubic volume element. In the case of KUBC and PERIODIC conditions prescribed to a given volume V, one defines the apparent modulus μ_E^{app} by the work of internal forces in the volume V subjected to the loading $\boldsymbol{E}_\mu$:

$$\mu_E^{app}(V) := <\boldsymbol{\sigma} : \boldsymbol{\varepsilon}> = <\boldsymbol{\sigma}> : \boldsymbol{E}_\mu = \frac{1}{V}\int_V \sigma_{12}\, dV \tag{9}$$

In the case of SUBC boundary conditions, an apparent shear modulus μ_Σ^{app} is defined as the work of internal forces generated in V by the application of the loading $\boldsymbol{\Sigma}_\mu$:

$$\frac{a^2}{\mu_\Sigma^{app}(V)} := <\boldsymbol{\sigma} : \boldsymbol{\varepsilon}> = \boldsymbol{\Sigma}_\mu : <\boldsymbol{\varepsilon}> = \frac{2a}{V}\int_V \varepsilon_{12}\, dV \tag{10}$$

These definitions remain formal insofar as the apparent elasticity properties of a given material volume element V are not necessarily isotropic.

	V	n	$\bar{\mu}^{app}$(MPa)	$D_\mu(V)$(MPa)	ϵ_{rel}
KUBC	25	100	52543	3186	1.2%
KUBC	400	50	50088	836	0.4%
KUBC	1000	25	49787	533	0.4%
KUBC	5000	10	49336	222	0.2%
PERIODIC	25	100	49669	3162	1.2%
PERIODIC	123	50	48886	1400	0.8%
PERIODIC	400	50	48784	811	0.4%
PERIODIC	500	50	48764	778	0.4%
SUBC	25	100	43397	3185	1.4%
SUBC	400	50	47308	823	0.4%
SUBC	1000	25	47566	538	0.4%
SUBC	5000	10	48390	178	0.2%

Table 1: Mean apparent shear modulus, associated scatter and relative error on the mean as a function of the domain size and of the number of realizations for three different boundary conditions (bulk copper polycrystals).

2.4 A definition of the RVE size

It is known that the RVE size is property and morphology dependent, but a well–suited parameter is necessary for quantitative comparisons. The choice of the boundary conditions applied to volumes of heterogeneous materials introduce a bias in the estimation of the apparent mechanical properties. A deterministic definition of the RVE is related to the volume size at which the estimated properties do not depend, within a given statistical precision, on the choice of boundary conditions any longer [3]. This leads however to large volume sizes that are sometimes hardly tractable numerically. In contrast, a series of simulations for various microstructures have shown that the use of periodicity conditions provide, with relatively small volumes, estimations that are close to the wanted effective properties [2, 4]. Such estimations based on rather small volumes require however a sufficient number of realizations of the microstructures to get accurate enough estimates. This suggests a pragmatic and statistical definition of the RVE size, as the minimum size for which the effective properties are estimated with a wanted precision [2] for a well–chosen set of boundary conditions. This is the definition of RVE size adopted in the present work. A quantitative definition of this RVE size is given in section 3.2.

3 RESULTS FOR BULK COPPER POLYCRYSTALS

3.1 Apparent shear moduli for bulk polycrystalline copper

Due to the uniform distribution of crystal orientations, the effective medium exhibits an isotropic linear elastic behaviour, described by effective bulk and shear moduli k^{eff} and μ^{eff}. For cubic symmetry, the apparent bulk modulus is not a random variable [7]. It is uniquely determined from the single crystal elasticity constants according to the formula $k^{app} = k^{eff} = (C_{11} + 2C_{12})/3 =$ 137067 MPa. As a result, the homogenization problem reduces to the estimation of apparent shear properties μ^{app} and *in fine* of the effective shear modulus μ^{eff}. It is shown in [2] that the fourth–rank tensor of apparent moduli $\boldsymbol{C}_E^{app}(V)$ obtained for a finite domain V containing N_g grains is generally not isotropic. However, its ensemble average $\bar{\boldsymbol{C}}_E^{app}(V)$, i.e. its mean value over a sufficiently large number of realizations turns out to be isotropic. This has been checked here for polycrystalline copper aggregates. The shear modulus associated with the isotropic elasticity tensor $\bar{\boldsymbol{C}}_E^{app}(V)$ coincides with $\bar{\mu}_E^{app}(V)$, the ensemble average of the apparent shear moduli $\mu_E^{app}(V)$ defined by equation (9) and computed for a domain V of given size (or equivalently containing $N = V$ grains in average). Accordingly, the estimation of $\bar{\mu}_E^{app}(V)$ only requires the determination of $\mu_E^{app}(V)$ for each realization. This is the

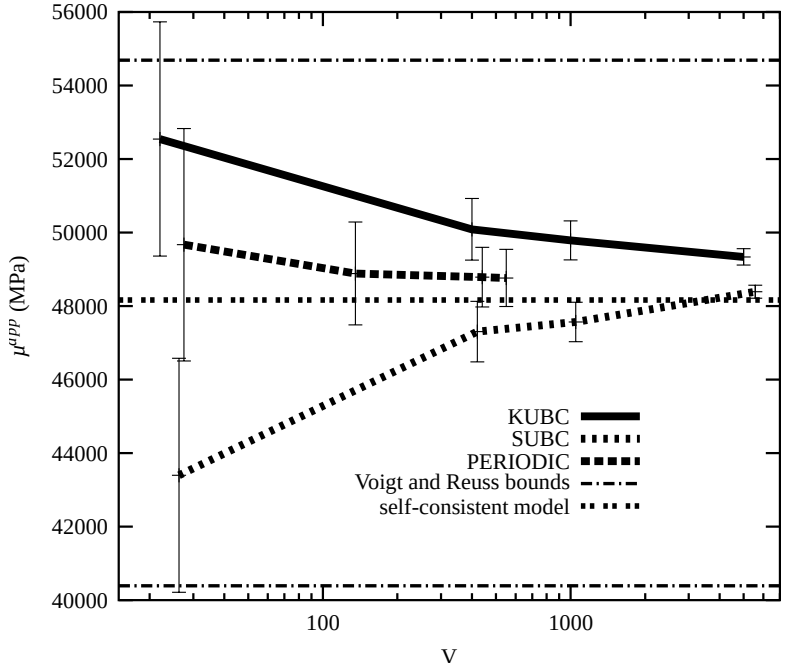


Figure 2: Mean values and intervals of variation for the shear modulus μ^{app} as a function of domain size, for three different boundary conditions (bulk copper polycrystals).

computation strategy adopted in this work. Similarly, using SUBC conditions, it is sufficient to compute μ_{Σ}^{app} for each realization according to equation (10).

The apparent shear moduli $\mu^{app}(V)$ were estimated using cubic volume elements V of increasing size, ranging from $V = 25$ to $V = 5000$ grains, with $n(V)$ realizations for every volume. The value of n is such that the estimation of the mean $\bar{\mu}^{app}(V)$ is obtained with a precision better than 1%. Simulation results for bulk copper polycrystals are shown in table 1.

Mean values and intervals of variation for the apparent shear modulus $[\bar{\mu}^{app}(V) - 2D_{\mu}(V), \bar{\mu}^{app}(V) + 2D_{\mu}(V)]$, are plotted in figure 2, as a function of volume size V. The mean apparent shear moduli strongly depend on the domain size and on the boundary conditions. However, the values converge towards an asymptotic constant μ^{eff} as the volume size increases, as expected. A striking feature of these results is the very fast convergence of the periodic solution and, in contrast, the very slow convergence associated with homogeneous boundary conditions. The periodic estimate is bounded by the KUBC and SUBC estimates:

$$\mu^{Reuss} \leq \bar{\mu}_{\Sigma}^{app} \leq \bar{\mu}_{PERIODIC}^{app} \leq \bar{\mu}_{E}^{app} \leq \mu^{Voigt} \tag{11}$$

where μ^{Reuss} and μ^{Voigt} denote the first order lower and upper bounds for the effective shear modulus of the polycrystal. For decreasing values of V, the apparent moduli $\bar{\mu}_{E}^{app}(V)$ (resp. $\bar{\mu}_{\Sigma}^{app}(V)$) get closer to the upper (resp. lower) limit μ^{Voigt} (resp. μ^{Reuss}). The bias observed on the mean value for all loading conditions for small volumes is clearly due to the specific boundary layer effect induced by each type of boundary condition. Another important result is the rate of decrease in the variance $D_{\mu}^2(V)$ of μ^{app} with increasing V for all three types of boundary conditions.

Finally, the estimated effective shear modulus is compared to the self–consistent estimate according to [7] in figure 2. The self-consistent method predicts a shear modulus of 48167 MPa, which is 1.2% lower than the periodic solution found with 500 grains. This difference lies within the numerical precision associated with the mesh density chosen in section 2.2. Note that this result *a priori* depends on the anisotropy coefficient of the considered cubic material.

3.2 Size of the RVE for bulk polycrystalline copper

The notion of RVE is necessarily related to the choice of a statistical precision in the estimation of the investigated effective property. First, we set a tolerance error α on the bias and find a corresponding volume V_0 such that:

$$|\bar{\mu}^{app}(V_0) - \mu^{eff}| \leq \alpha \tag{12}$$

This condition sets a lower bound for the size of the RVE. Then, the relative precision of the estimation of the mean $\bar{\mu}^{app}(V)$ of apparent shear moduli for a given volume $V \geq V_0$ and a given number of realizations n, can be defined according to the sampling theory by:

$$\epsilon_{rel} = \frac{2D_\mu(V)}{\bar{\mu}^{app}(V)\sqrt{n}} \tag{13}$$

This definition includes explicitly the number of realizations n. In turn, the number of realizations required to correctly estimate $\bar{\mu}^{app}(V)$ is deduced from equation (13) provided that the variance $D^2_\mu(V)$ is known. According to homogenization conditions (8), (9) and (10), the apparent shear modulus is obtained by averaging an additive scalar over the volume V. As a result, for asymptotically large volumes, the variance $D^2_\mu(V)$ of $\mu^{app}(V)$ is given by:

$$D^2_\mu(V) = D^2_\mu \frac{A_3}{V} \tag{14}$$

where A_3 is the integral range, a well–established quantity for additive geometrical properties such as volume fraction. It has the dimension of a volume. D^2_μ is the point variance of $c_{1212}(\underline{\boldsymbol{x}})$, which depends on the crystal orientation at $\underline{\boldsymbol{x}}$. For uniform orientation distributions, it can be expressed in terms of the single crystal cubic elasticity constants as follows:

$$D^2_\mu = < (\boldsymbol{c} : \boldsymbol{c})_{1212} > - < c_{1212} >^2, \text{ with } < c_{1212} >= \frac{1}{5}(C_{11} - C_{12} + 3C_{44}) \tag{15}$$

$$< (\boldsymbol{c} : \boldsymbol{c})_{1212} >= \frac{1}{35}(-6C_{44}C_{12} - 4C_{12}C_{11} + 2C^2_{12} + 2C^2_{11} + 6C_{11}C_{44} + 15C^2_{44}) \tag{16}$$

where $< . >$ denotes here averaging over uniformly distributed crystal orientations. For pure copper, one gets $D_\mu = 13588$ MPa. We choose to identify the integral range A_3 from the results obtained with periodicity conditions because they introduce the smallest bias in the estimated effective shear modulus. We find $A_3 = 1.43$, to be compared with the mean grain size set to 1. It can also be compared to the integral range for the volume fraction of a given orientation $A_3 = 1.17$ given in [2]. The integral range A_3 is a well–suited parameter to compare RVE sizes for different properties and morphologies. It characterizes the rate of decrease in the dispersion of apparent properties for increasing volume sizes, according to equation (14).

Equations (12), (13) and (14) can now be used quantitatively to determine a minimal size of RVE for a given precision ϵ_{rel} and a given number of realizations n:

$$V = \frac{4}{n} D^2_\mu \frac{A_3}{\epsilon^2_{rel}(\mu^{eff})^2} \tag{17}$$

In the case of periodic boundary conditions, the choice $(\epsilon_{rel}, n) = (1\%, 10)$ gives a minimal volume corresponding to $V = 445$. For $n = 100$ computations, this volume reduces to 45. This low number of grains still remains in the domain range for which periodic boundary conditions introduce only a slight bias in the estimation of the effective property. The obtained results compare quite well with the prediction of the formula identified in [4] that relates the size of the RVE for a given precision to the anisotropy coefficient a. Taking a relative error $\epsilon_{rel} = 1\%$ and 20 realizations, Nygårds' formula predicts $V = 265$, vs 220 according to our formula (17). Interestingly, the simulations of [10] for 2D elastic copper polycrystals lead to a value $V = 484$, but in the case of homogeneous boundary conditions and without any information about the variance of the results.

	V	n	$\bar{C}_{66}^{app}$ or $\bar{C}_{44}^{app}$(MPa)	$D_\mu(V)$(MPa)	ϵ_{rel}
KUBC C_{66}	50	50	52498	2039	1.1%
KUBC C_{44}	50	50	53234	2771	1.5%
KUBC C_{66}	120	50	52143	1372	0.7%
KUBC C_{44}	120	50	52842	1310	0.7%
KUBC C_{66}	260	50	52350	1101	0.6%
KUBC C_{44}	260	50	52608	945	0.5%
KUBC C_{66}	400	50	52211	797	0.4%
KUBC C_{44}	400	50	52428	675	0.4%
PERIODIC C_{66}	50	50	48794	2066	1.2%
PERIODIC C_{44}	50	50	49884	2985	1.7%
PERIODIC C_{66}	120	50	48467	1437	0.8%
PERIODIC C_{44}	120	50	49609	1375	0.8%
PERIODIC C_{66}	260	50	48788	1149	0.7%
PERIODIC C_{44}	260	50	49405	1024	0.6%
PERIODIC C_{66}	400	50	48604	785	0.5%
PERIODIC C_{44}	400	50	49431	905	0.5%
mixed PERIODIC–SUBC C_{66}	400	50	46350	791	0.5%

Table 2: Mean apparent shear moduli, associated scatter and relative error on the mean as a function of the domain size and of the number of realizations for three different boundary conditions (1–grain–thick polycrystalline copper sheets).

4 RESULTS FOR THIN POLYCRYSTALLINE COPPER SHEETS

In this section, the question of the existence of homogeneous equivalent properties and of the size of the corresponding representative volume element is investigated in the case of linear elastic polycrystalline copper sheets exhibiting a small and fixed number of grains through the thickness. The crystallographic orientations are distributed randomly among the grains in each simulation, which results in an isotropic crystallographic texture. The grain morphology and distribution in space still correspond to the Voronoi mosaic random model.

4.1 Apparent shear moduli for thin copper polycrystalline structures

The polycrystalline copper layer of figure 1(b) is obtained by superimposing a regular mesh of a plate with a given thickness on a bulk Voronoi mosaic. As in the previous section, a crystallographic orientation is attributed to each integration point in each element according to the color of the underlying voxel. The same mesh density as in section 3 is used for the plate, namely 16 elements per grain. Two thicknesses are considered corresponding respectively to an average of 1 grain and 3 grains through the thickness. Square sheets are considered, the normal to the sheet being the direction 3. Two orthogonal in–plane directions parallel to edges of the square plate are labelled 1 and 2. In all simulations presented in this section, the thickness of the plate and grain size are kept constant whereas the width is increased gradually leading to larger volumes V and therefore larger numbers of grains.

The boundary conditions KUBC and PERIODIC defined in section 2.3 are applied to the entire outer surface of the plates, namely both faces normal to direction 3, and the four lateral faces. In the case of PERIODIC boundary conditions, a periodicity constraint is imposed in the morphology of the grains, as depicted in section 2.1 but only with respect to lateral faces. This accelerates slightly the convergence of the apparent moduli. No periodicity constraint is prescribed to the morphology of the grains on the faces normal to direction 3. Special boundary conditions called *mixed PERIODIC–SUBC* are introduced that are especially relevant for free–standing films. According to these conditions, periodicity conditions for all displacement components are imposed to the lateral faces of the plate,

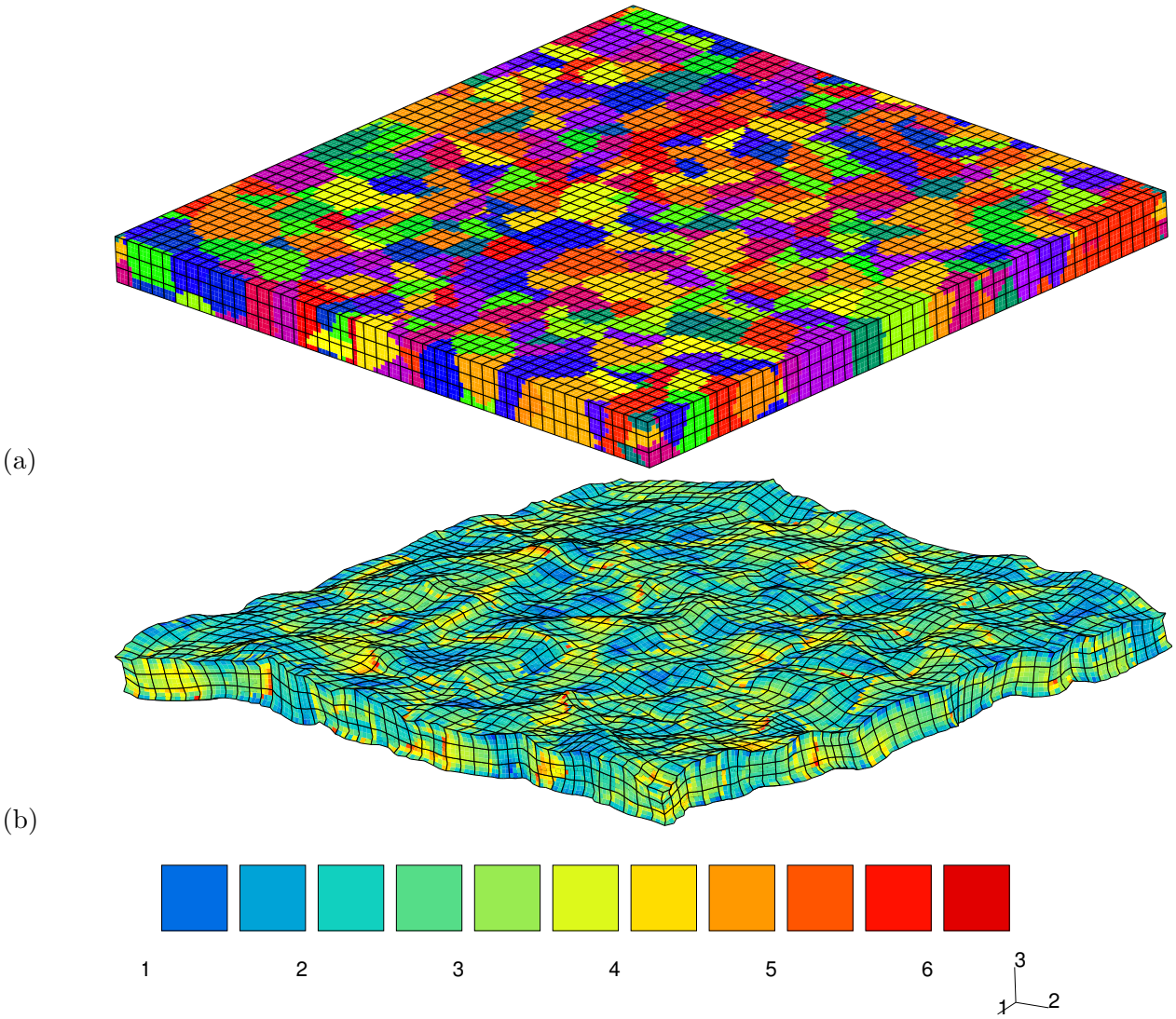


Figure 3: Finite element computations of a plate containing 500 grains with one grain within the thickness: (a) finite element mesh and morphology of the grains, (b) stress heterogeneities induced by in–plane shear under mixed PERIODIC–SUBC conditions. The deformation state of the grains is magnified for the illustration. The represented variable is the normalized equivalent von Mises stress $\sigma_{eq}/\sigma_{eq}^{min}$.

whereas the stress vector is prescribed to both surfaces normal to direction 3.

The volume sizes, or equivalently the numbers of grains in the sample plates, considered in the simulations of the elastic deformation of copper polycrystalline plates are listed in table 2 in the case of one grain through the thickness and in table 3 in the case of three grains, for all types of boundary conditions. They range from 50 to 400 grains per polycrystalline plate in average. The numerical analysis is restricted to the computation of the apparent shear moduli of a large number of plates corresponding to different realizations of the microstructure. Whatever the type of boundary conditions, the apparent shear moduli are defined as proper ratios of mean shear stress and shear strain components over the whole volume of the plate, according to the formula:

$$\begin{pmatrix} \Sigma_{23} \\ \Sigma_{31} \\ \Sigma_{12} \end{pmatrix} = \begin{pmatrix} C_{44}^{app} & \times & \times \\ \times & C_{55}^{app} & \times \\ \times & \times & C_{66}^{app} \end{pmatrix} \begin{pmatrix} 2E_{23} \\ 2E_{31} \\ 2E_{12} \end{pmatrix} \tag{18}$$

In the case of KUBC and PERIODIC boundary value problems, the components $E_{12} = 1/2$ and $E_{23} = 1/2$ are imposed successively to estimate C_{66}^{app} and C_{44}^{app} respectively, the remaining components

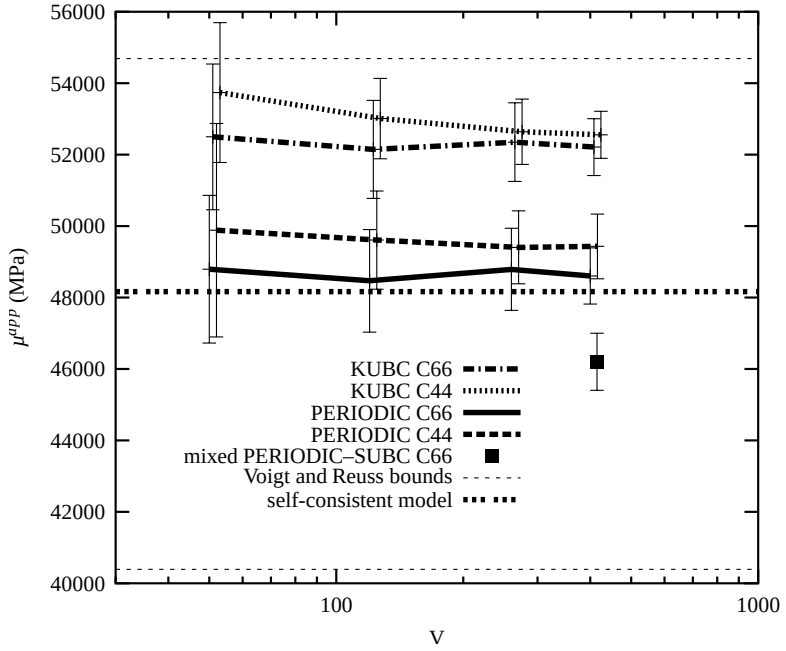


Figure 4: Mean values and intervals of variation for the shear modulus μ^{app} as a function of domain size, for three different boundary conditions (one–grain thick copper polycrystalline sheets).

of the average strain tensor $\boldsymbol{E}$ being set to zero. In the case of mixed PERIODIC–SUBC problems, the components $\Sigma_{12} = a$ and $\Sigma_{23} = a(= 1$ MPa) of the average stress tensor $\boldsymbol{\Sigma}$ are prescribed successively to the volumes, the remaining components being set to zero.

It cannot be expected *a priori* that the mean apparent properties $\bar{C}_{66}^{app}$ and $\bar{C}_{44}^{app}$ will coincide, even for a sufficiently high number of realizations or number of grains, due to the special morphology of the plates with a small number of grains through the thickness. That is why both properties are evaluated. For reasons of statistical homogeneity of grain distribution and isotropy of the given crystallographic texture, the mean apparent property $\bar{C}_{55}^{app}$ coincides with $\bar{C}_{44}^{app}$. It is therefore not reported here. For the same reasons, the components labelled with a cross $\times$ in equation (18) vanish in average (but, of course, not in general for an individual plate). A number of 50 realizations for each investigated volume size corresponding to distinct grain and orientation distributions was found to be sufficient to ensure a relative error always smaller than 1.7% in the estimation of the apparent properties. A relative precision of 0.4–0.5% was even reached for larger volumes. An example of such computations is given in figure 3. The finite element mesh of a thin sheet with one grain through the thickness is shown in figure 3(a). The figure 3(b) gives the fields of stress concentration in the case of in–plane shear loading according to mixed PERIODIC–SUBC boundary conditions. The stress concentration is defined here as the ratio of the local von Mises equivalent stress σ_{eq} divided by the minimum value reached in the aggregate for a given mean shear strain E_{12} (resp. shear stress Σ_{12}). The roughness of the deformed plate and the stress gradient that develops along the normal direction are visible.

Mean values and intervals of variation for the apparent shear moduli C_{44}^{app} and C_{66}^{app} are plotted, as a function of volume size V, in figure 4 in the case of one grain through the thickness. Each mean apparent shear modulus for each type of boundary conditions is found to slightly depend on the volume size and to converge towards an asymptotic value. These limit values can be defined unambiguously since the mean apparent properties do not significantly vary for the considered largest volume sizes. They are called $\bar{C}_{44}$ and $\bar{C}_{66}$ respectively. In the meantime, the scatter of the individual apparent properties decreases slowly with increasing the volume size. Two main results can be pointed out. First, the asymptotic mean apparent shear modulus $\bar{C}_{66}$ (resp. $\bar{C}_{44}$) depends on the type of boundary conditions:

$$\bar{C}_{66}^{KUBC} \neq \bar{C}_{66}^{PERIODIC} \neq \bar{C}_{66}^{mixed}, \quad \bar{C}_{44}^{KUBC} \neq \bar{C}_{44}^{PERIODIC} \neq \bar{C}_{44}^{mixed} \tag{19}$$

The same ranking of asymptotic shear moduli according to the type of boundary conditions was found

for bulk polycrystals:

$$\bar{C}_{66}^{mixed} < \bar{C}_{66}^{PERIODIC} < \bar{C}_{66}^{KUBC}, \quad \bar{C}_{44}^{mixed} < \bar{C}_{44}^{PERIODIC} < \bar{C}_{44}^{KUBC} \tag{20}$$

The found difference of 7% between $\bar{C}_{66}^{PERIODIC}$ and $\bar{C}_{66}^{KUBC}$ is significant compared to the relative precision of the estimations. It can be noticed that the asymptotic in–plane shear modulus $\bar{C}_{66}^{PERIODIC} = 48604$ MPa is close to the effective shear modulus $\mu^{eff} = 48764$ MPa found for bulk copper polycrystals. Second, an anisotropy of asymptotic shear properties pertains for large plates:

$$\bar{C}_{66}^{KUBC} \neq C_{44}^{KUBC}, \quad \bar{C}_{66}^{PERIODIC} \neq \bar{C}_{44}^{PERIODIC} \tag{21}$$

unlike what happens to bulk polycrystals. The found differences of about 1.7% between $\bar{C}_{66}^{PERIODIC}$ and $\bar{C}_{44}^{PERIODIC}$ is significant, compared to the relative precision of the estimations. The effective shear properties based on KUBC boundary conditions are close to the values that one gets by considering 2D crystal aggregates under plane strain conditions [10].

4.2 Discussion on the existence of a RVE for thin structures

The new feature of the analysis of thin layers is that the boundary conditions induce a bias in their asymptotic elastic behaviour, even for very large plates. This is in contrast to the bulk behaviour of elastic polycrystals for which the apparent shear modulus $\bar{\mu}^{app}$ was found to converge towards a single asymptotic value μ^{eff} irrespective of the type of boundary conditions. Even though asymptotic shear properties can be associated to the polycrystalline plate for a given type of boundary conditions, we reserve the term of effective properties to the case of asymptotic values that eventually do not depend on the type of boundary conditions. Accordingly, unambiguous effective properties do not exist for copper polycrystalline layers with one grain through the thickness. As a result, a RVE cannot be defined unambiguously in this case. In practice, the asymptotic values found for each type of boundary conditions can be used to model the elastic behaviour of thin copper layers provided that the *in situ* material layers undergo similar boundary or interface conditions. For instance, in the case of thin copper sheets with free surfaces, the asymptotic shear properties obtained from mixed PERIODIC–SUBC conditions (or equivalently SUBC conditions) will be the relevant ones for further structural analyzes. The found slight anisotropy of the asymptotic tensor of elastic moduli should also be taken into account. The authors in [6] also study the evolution of scatter when increasing the number of grains in textured copper one–grain thick coatings on a substrate in the case of mixed KUBC–SUBC–PERIODIC boundary conditions. They also found asymptotic values of the apparent elastic properties, which are specific of the chosen boundary conditions.

When the number of grains within the thickness increases, the bias introduced by the boundary conditions is reduced. The case of thin copper sheets having 3 grains through the thickness was investigated in the present work. The table 3 shows that 50 simulations over 400 grains with PERIODIC boundary conditions provide mean apparent shear properties close to the shear modulus of bulk polycrystalline copper (the difference is less than 0.8%). There is still a difference of 2% between the asymptotic values of the apparent properties $\bar{C}_{66}^{PERIODIC}$ and $\bar{C}_{66}^{KUBC}$. The bias introduced by the boundary conditions does still exist but is significantly smaller than for one–grain–thick sheets. Furthermore, the anisotropy of the shear moduli has almost disappeared since $\bar{C}_{66}^{PERIODIC}$ and $\bar{C}_{44}^{PERIODIC}$ do not differ from more than 0.05% with a relative precision of 0.35% on the mean. Accordingly, the concepts of effective properties and RVE are found to be meaningful in the case of 3–grain thick copper polycrystalline sheets with a tolerance of 2%. The concept of RVE is also characterized by the value of the integral range A_3^{layer} that can be defined as in section 3.2, equation (14). The value $A_3^{layer} = 1.40$ is deduced from the results of table 3, which is almost equal to the integral range found for bulk polycrystals. Finally, for a given precision of $\epsilon_{rel} = 1\%$ in the estimated mean and for $n = 10$ realizations and periodic boundary conditions, the RVE size is found to be equal to 435 grains, which is close to the result of 445 grains found for bulk polycrystals. The main difference is that, in the case of copper polycrystalline layers, the RVE does not have the shape of a cube. It is a plate with 3 grains within the thickness and 12 grains along the edges, instead of a cube of about 8×8×8 grains.

5 CONCLUSION AND PROSPECTS

The statistical definition of the Representative Volume Element, first proposed in [2], was applied to aggregates of linear elastic copper uniformly oriented crystals for bulk and thin structures. It is associated with the choice of a targeted relative precision ϵ_{rel} on the estimated effective property and of a number n of realizations. The corresponding RVE size is then given by equation (17), where the integral range A_3 fully characterizes the asymptotic response of volumes made of the considered heterogeneous material. Bulk copper polycrystals were considered as a reference to analyse the behaviour of thin sheets and layers. The following conclusions were drawn:

- Effective properties do not exist for all types of polycrystalline aggregates. In particular, it was shown that it is not possible to determine shear properties that are independent of the type of boundary conditions for copper polycrystalline thin sheets with only one grain within the thickness. In such cases, however, asymptotic apparent properties can be associated to each type of boundary conditions. They may be anisotropic even though the crystallographic and morphological textures of the aggregate are isotropic. Such asymptotic values can be used in structural computations provided that the boundary conditions are indeed relevant for the considered situation. This is the case of SUBC or mixed PERIODIC–SUBC conditions for one–grain thick copper polycrystalline sheets with free surfaces or for free–standing films.

- For polycrystalline copper sheets having three grains through the thickness, the asymptotic apparent shear modulus determined using periodic boundary conditions coincides with the effective shear modulus for bulk polycrystals with a precision better than 1%.

- The value of the integral range $A_3 = 1.4 \pm 0.05$ was found for bulk copper polycrystals but also for 3–grain thick copper sheets.

- As a result of the proposed definition, a rather small RVE size can be determined by using periodic boundary conditions rather than homogeneous boundary conditions. This confirms the results found in [2] and [4]. For $\epsilon_{rel} = 1\%$ and $n = 10$, the required number of grains is a little higher than 400 grains for bulk and 3–grain thick polycrystals.

- The shape of a RVE is not necessarily a cube. It can have a rather small (but not too small!) number of grains along one direction, provided that the number of grains is sufficiently high along the two other directions. The adequate ratio was determined in the case of 3–grain thick copper plates with a tolerance of 2%.

The question of RVE size addressed in this work can be raised also in at least two other situations: the case of textured bulk and thin films (especially {111} and {001} textures relevant in the latter situation [6]), and the case of the elastoplastic behaviour of polycrystals.

Another important application is the modeling of the mechanical behaviour of polycrystalline coatings which often have only one grain within the thickness. In this case, mixed periodic–KUBC–SUBC conditions should be developed to include the substrate constraint effect (first attempts can be found in [6] for copper films on a substrate and in [11] for zinc coatings).

Acknowledgements

The authors are indebted to P. Featonby and J.–C. Leroy (SAGEM) for stimulating discussions during this work. The authors thank SAGEM and the DGA for their financial support.

References

1. G. Cailletaud, S. Forest, D. Jeulin, F. Feyel, I. Galliet, V. Mounoury, and S. Quilici. Some elements of microstructural mechanics. *Computational Materials Science*, 27:351–374, 2003.

	V	n	$\bar{C}_{66}^{app}$ or $\bar{C}_{44}^{app}$ (MPa)	$D_\mu(V)$(MPa)	ϵ_{rel}
KUBC C_{66}	120	50	51212	1483	0.8%
KUBC C_{44}	120	50	50814	1516	0.8%
PERIODIC C_{66}	123	50	48805	1400	0.8%
PERIODIC C_{44}	123	50	48914	1400	0.8%
PERIODIC C_{66}	400	50	49123	821	0.5%
PERIODIC C_{44}	400	50	49148	614	0.35%
KUBC C_{66}	1000	50	50418	530	0.3%
KUBC C_{44}	1000	50	50993	446	0.25%
KUBC C_{66}	2000	10	50464	243	0.3%
KUBC C_{44}	2000	10	50891	274	0.3%
KUBC C_{66}	5000	1	50200		
KUBC C_{44}	5000	1	51000		

Table 3: Mean apparent shear modulus, associated scatter and relative error on the mean as a function of the domain size and of the number of realizations for PERIODIC conditions (3–grain–thick polycrystalline copper sheets).

2. T. Kanit, S. Forest, I. Galliet, V. Mounoury, and D. Jeulin. Determination of the size of the representative volume element for random composites : statistical and numerical approach. *International Journal of Solids and Structures*, 40:3647–3679, 2003.

3. K. Sab. On the homogenization and the simulation of random materials. *Eur. J. Mech. Solids*, 11:585–607, 1992.

4. M. Nygårds. Number of grains necessary to homogenize elastic materials with cubic symmetry. *Mechanics of Materials*, 35:1049–1057, 2003.

5. M. Hommel and O. Kraft. Deformation behavior of thin copper films on deformable substrates. *Acta materialia*, 49:3935–3947, 2001.

6. A. Wilkström and M. Nygårds. Anisotropy and texture in thin copper films–an elastoplastic analysis. *Acta materialia*, 50:857–870, 2002.

7. B.K.D. Gairola and E. Kröner. A simple formula for calculating the bounds and the self–consistent value of the shear modulus of polycrystalline aggregates of cubic crystals. *Int. Engng Sci.*, 19:865–869, 1981.

8. Z–set package. www.nwnumerics.com, www.mat.ensmp.fr. 2001.

9. C. Huet. Application of variational concepts to size effects in elastic heterogeneous bodies. *J. Mech. Phys. Solids*, 38:813–841, 1990.

10. Z.-Y. Ren and Q.-S. Zheng. A quantitative study of minimum sizes of representative volume elements of cubic polycrystals–numerical experiments. *Journal of the Mechanics and Physics of Solids*, 50:881–893, 2002.

11. R. Parisot, S. Forest, A. Pineau, F. Grillon, X. Démonet, and J.-M. Mataigne. Deformation and Damage Mechanisms of Zinc Coatings on Galvanized Steel Sheets, Part I : Deformation Modes. *Metallurgical and Materials Transactions*, 35A:797–811, 2004.