

Self-consistent Scale-bridging Approach to Compute the Elasticity of Multi-phase Polycrystalline Materials

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ABSTRACT

A necessary prerequisite for a successful theory-guided up-scale design of materials with application-driven elastic properties is the availability of reliable homogenization techniques. We report on a new software tool that enables us to probe and analyze scale-bridging structure-property relations in the elasticity of materials. The newly developed application, referred to as SC-EMA (Self-consistent Calculations of Elasticity of Multi-phase Aggregates) computes integral elastic response of randomly textured polycrystals. The application employs a Python modular library that uses single-crystalline elastic constants C_{ij} as input parameters and calculates macroscopic elastic moduli (bulk, shear, and Young's) and Poisson ratio of both single-phase and multi-phase aggregates. Crystallites forming the aggregate can be of cubic, tetragonal, hexagonal, orthorhombic, or trigonal symmetry. For cubic polycrystals the method matches the Hershey homogenization scheme. In case of multi-phase polycrystalline composites, the shear moduli are computed as a function of volumetric fractions of phases present in aggregates. Elastic moduli calculated using the analytical self-consistent method are computed together with their bounds as determined by Reuss, Voigt and Hashin-Shtrikman homogenization schemes. The library can be used as (i) a toolkit for a *forward* prediction of macroscopic elastic properties based on known single-crystalline elastic characteristics, (ii) a sensitivity analysis of macro-scale output parameters as function of input parameters, and, in principle, also for (iii) an *inverse* materials-design search for unknown phases and/or their volumetric ratios.

INTRODUCTION

Elastic properties are among the most important physical parameters characterizing materials. They originate from sub-nanometer inter-atomic bonds (Figure 1a) that are governed by the fundamental laws of quantum-mechanics and propagate through multiple length-scales (see e.g. Figure 1b,c) up to the macroscale (Figure 1d). Atomic-scale elasticity is, for most crystalline materials, anisotropic. This means that the energy necessary to deform a crystal by a certain constant amount of strain is different when applying the strain along symmetrically inequivalent crystallographic directions. The anisotropy may be conveniently visualized by the directional dependence of Young's modulus that connects the energy and the applied strain (see Figure 1d). When going up-scale, most metallic materials used in industrial applications are polycrystalline aggregates consisting in individual single-crystalline grains with different orientations (see Figure 1c). Combining state-of-the-art computational approaches, such as quantum-mechanical calculations (Figure 1e,f) and linear-elasticity homogenization theories (Figure 1g) within a suitable inter-disciplinary scheme, provides a platform for predicting a macroscopic elastic response of these aggregates (Figure 1h) without any empirical input.

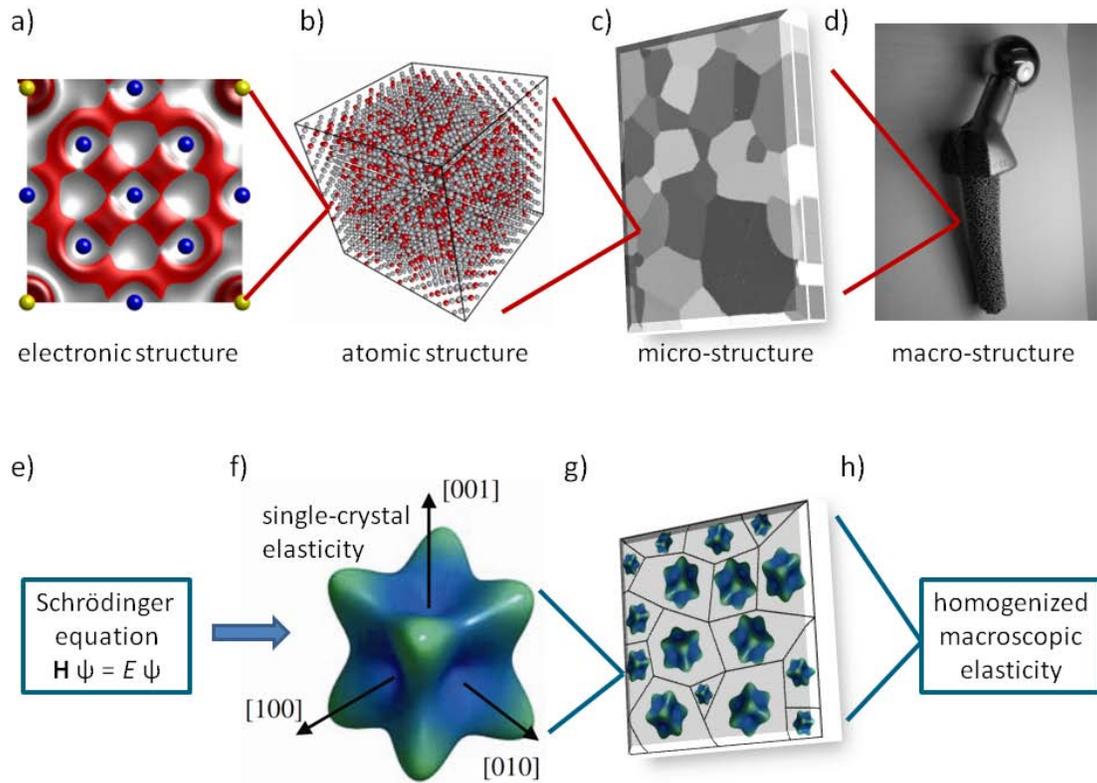


Figure 1. Schematic picture connecting the multi-scale structure of materials (a)-(d), represented here by a stainless-steel hip-replacing medical implant, with corresponding computational approaches to elastic properties (e)-(h).

Innovative industrial products require new materials with macroscopic properties dictated by specific applications and the overall elastic response and its anisotropy are among them. Design specifications related to macroscopic shear or Young's moduli are related, for example, to prevent resonant vibrational frequencies for safety reasons. Another field of applications are interfaces between dissimilar materials where a too large elastic mismatch may result in a failure during the operational lifetime. The latter case applies e.g. to implant alloys, as used for hip replacements, that are in direct contact with elastically much softer bones. The elastic mismatch is known to be one of the reasons causing implant failures and softer implant materials with bone-matching elasticity are being designed (see e.g. our recent work [1] and references therein).

Our work has been motivated by a clear need for a thorough understanding of structure-property relations connecting elastic properties of materials over multiple length-scales. We have developed an open-access web-based application that uses a Python modular library calculating the integral macroscopic elastic response of aggregates employing self-consistent mean-field homogenization methods. In order to avoid an arbitrary complexity stemming from textured aggregates with specific size, shape and orientation distributions of grains, we limit ourselves in this project to so-called texture-free aggregates containing equally-sized grains that are shaped as polygons. Such aggregates elastically respond like a macroscopically homogeneous effective medium and are characterized by only two elastic moduli, such as bulk modulus B , Young's modulus Y or shear modulus μ , that are important materials-design criteria (see e.g. Refs. [1,5]).

THEORY

The polycrystalline elastic response of multi-phase aggregates can be determined within a self-consistent solution for the effective medium [2, 3] from (i) the elastic single-crystalline constants and (ii) the volumetric fractions of individual components. The approach is based on the multiple scattering theory and was originally applied to predict elastic properties of single-phase polycrystals with cubic symmetry by Zeller and Dederichs [4]. The concept was later generalized by Middya and Basu [2] to single-phase aggregates with non-cubic symmetries and further extended by Middaya *et al.* [3] to multi-phase composites. Following the original approach by Zeller and Dederichs [4], the basic steps of the effective medium approach may be found below. A macroscopic effective medium that is elastically homogeneous and contains microscopic fluctuations may be characterized by an effective elastic constant C_{ijkl}^* defined by

$$\langle \sigma_{ij}(\mathbf{r}) \rangle = C_{ijkl}^* \langle \epsilon_{kl}(\mathbf{r}) \rangle \quad (1)$$

where $\sigma_{ij}(\mathbf{r})$ and $\epsilon_{kl}(\mathbf{r})$ are the local stress and strain fields at point \mathbf{r} , respectively, and the angular

brackets denote ensemble averages. Assuming the aggregate is in equilibrium, the local field of elastic stiffnesses $C(\mathbf{r})$ can be decomposed into an arbitrary constant part C_0 and a fluctuating

part $\delta C(\mathbf{r})$. The resulting local strain field, ϵ , can then be written (in a short-hand notation) as

$$\epsilon = \epsilon^0 + GT\epsilon^0, \quad (2)$$

where ϵ_0 and G are the strain and modified Green's function of the medium defined by C_0 . The

T-matrix is given by

$$T = \delta C(I - G\delta C)^{-1}, \quad (3)$$

where I is the unit tensor. Employing the local stress-strain relation and equations (1) and (2) we obtain

$$C^* = C^0 + \frac{\langle T \rangle}{I + \langle GT \rangle}. \quad (4)$$

The exact evaluation of $\langle T \rangle$ and $\langle G\tau \rangle$ is unfortunately impossible for any realistic case.

However, by neglecting inter-granular correlations, the T-matrix can be rearranged in terms of single-grain t matrices (t_α) for each grain α

$$T = \sum_{\alpha} t_{\alpha} = \tau. \quad (5)$$

Inserting equation (5) into equation (4) gives

$$C^* = C^0 + \langle \tau \rangle (I + \langle G\tau \rangle)^{-1}. \quad (6)$$

For a single phase polycrystal, the self-consistent solution of eq. 6 can be obtained by choosing a

C_0 that satisfies the condition $\langle \tau \rangle = 0$. For a multi-phase polycrystal, a solution to equation (6)

can be found by accounting for the volume fraction v^i and τ^i of each phase i [2] via

$$\left\langle \sum_i v^i \tau^i \right\rangle = 0. \quad (7)$$

The actual equations for specific crystallographic classes may be found for example in Refs. [2,3,5]. Specifically for the single-phase cubic crystallites the methods results in the same equation as the Hershey's homogenization scheme [6].

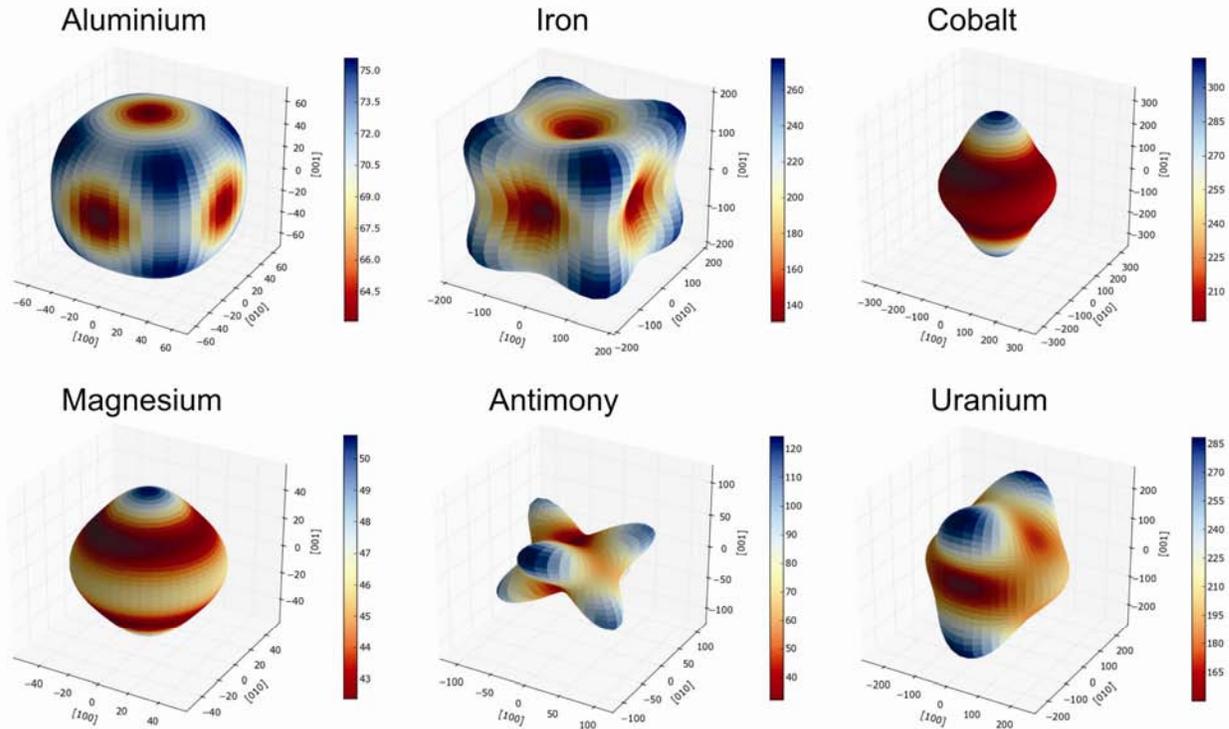


Figure 2 Directional dependencies of single-crystalline Young's modulus (in GPa) visualized by the SC-EMA code based on elastic constants collected from literature (Al [7], Fe [8], Co [9], Mg [10], Sb [11], U [12]). Please mind different color-coded pressure scales for each sub-figure.

DISCUSSION

In order to illustrate the method in case of single-phase polycrystals, we have used published elastic constants for a few elements (Al [7], Fe [8], Co [9], Mg [10], Sb [11], and U [12]) crystallizing in phases with different symmetries. The single-crystalline anisotropy of these elements is visualized as directional dependencies of Young's modulus in Figure 2. Based on the elastic constants, we have employed the method as described above to predict polycrystalline elastic moduli (bulk modulus B , shear modulus μ , Young's modulus Y , and Poisson ratio ν). The computed values are summarized together with available experimental data in Table I. The agreement between our prediction and experimental data is in general very good.

Table I SC-EMA computed homogenized elastic moduli (bulk B , shear μ , and Young's Y in GPa) and Poisson's ratio ν with experimental data in brackets (<http://www.periodictable.com>).

element	bulk modulus B	shear modulus μ	Young modulus Y	Poisson's ratio ν
Aluminium [5]	76 (76)	28 (26)	74 (70)	0.337 (0.350)
Iron [6]	179 (180)	80 (76)	209 (200)	0.286 (0.310)
Tin [7]	61 (50)	19 (18)	51 (50)	0.361 (0.360)
Cobalt [8]	190 (180)	82 (75)	215 (209)	0.312 (0.310)
Magnesium [9]	36 (45)	17 (17)	44 (45)	0.292 (0.290)
Antimony [10]	45 (42)	33 (20)	80 (55)	0.207 (-)
Uranium [11]	113 (100)	85 (111)	203 (208)	0.201 (0.230)

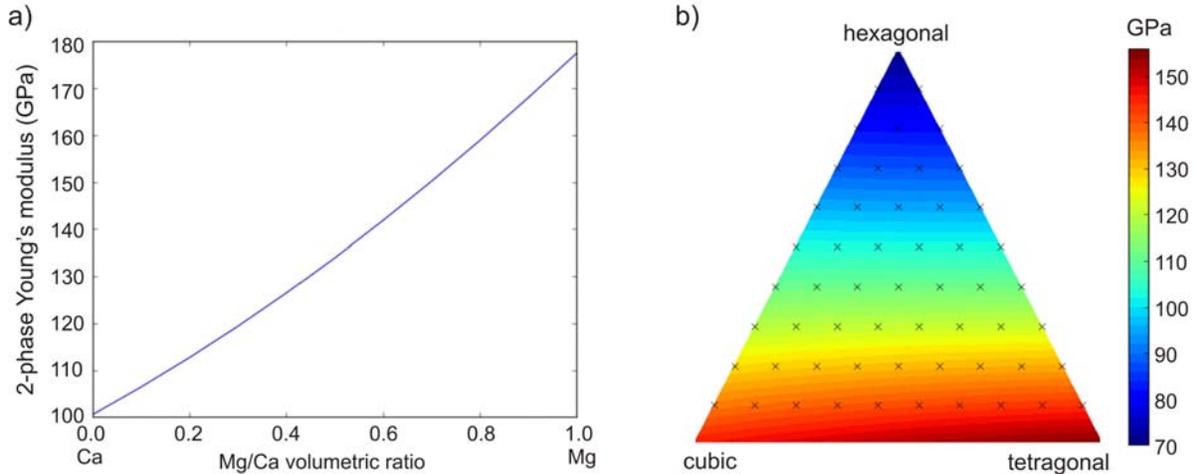


Figure 3 Computed dependencies of homogenized shear modulus in 2-phase Mg-Ca composites (a) and generic 3-phase (b) composites as functions of the volumetric ratio of the phases as visualized by the SC-EMA software package. The 2-phase composites contain phases of the same symmetry (elemental Ca and Mg that both crystallize in the hexagonal close-packed phase), while the generic three-phase composites in part (b) possess cubic, tetragonal, and hexagonal, respectively (see the text for details). Crosses in (b) indicate computed data-points.

Finally, in order to illustrate further features of our software, exemplarily the elasticity of 2-phase Mg-Ca composites and generic 3-phase ones are shown in Figure 3. Figure 3a visualizes the polycrystalline Young's modulus of 2-phase Ca-Mg composites as a function of the Mg/Ca volumetric fraction. Figure 3b shows the shear modulus of hypothetical 3-phase aggregates with (i) a cubic phase with elastic constants $C_{11} = 107$ GPa, $C_{12} = 60$ GPa, and $C_{44} = 28$ GPa, (ii) a tetragonal phase with constants $C_{11} = 275$ GPa, $C_{12} = 179$ GPa, $C_{13} = 152$ GPa, $C_{33} = 165$ GPa, $C_{44} = 54$ GPa, and $C_{66} = 113$ GPa, and (iii) a hexagonal phase with $C_{11} = 167$ GPa, $C_{12} = 13$ GPa, $C_{13} = 66$ GPa, $C_{33} = 66$ GPa, $C_{55} = 140$ GPa. As seen, the predicted trends of both Young's modulus for 2-phase composites in Figure 3a and the shear modulus of 3-phase ones in Figure 3b are rather monotonous and nearly linear. This is essential as far as a future inversion of this homogenization technique is concerned. The same close-to-linear trends were also predicted in case of two-phase Ti-Nb alloys in Ref. [5]. It is thus likely that after a thorough analysis of the underlying mathematical backbone of this mean-field homogenization method, it will be possible to find its inversion in order to further accelerate a theory-guided materials design of alloys with application-driven macroscopic elastic properties.

CONCLUSIONS

To conclude, we have developed a software tool for scale-bridging modeling of elastic properties of materials. The main purpose of the code is the prediction of the homogenized elastic response of texture-free aggregates containing either one or more phases that can be of cubic, tetragonal, hexagonal, orthorhombic, or trigonal symmetries. In case of multi-phase polycrystalline composites, the shear moduli are computed as a function of volumetric fractions of phases present in the aggregate. The input single-crystalline elastic constants are also checked with respect to conditions of mechanical stability.

For single-phase polycrystals the software allows studying the elastic anisotropy at atomistic scale by visualizing the value of single-crystalline Young's modulus along the main three crystallographic axes. Specifically in case of cubic crystals, the code also evaluates its planar counterpart, the area modulus (see details in Refs. [13, 14]) for (001), (110) and (111) atomic planes. The input single-crystalline elastic constants are then used to compute polycrystalline elastic moduli (bulk, shear, and Young) employing self-consistent homogenization scheme together with corresponding Voigt [15], Reuss [16] and Hashin-Shtrikman [17] bounds.

Our software library [18] allows to analyze scale-bridging structure-property relations and can be used as (i) a toolkit for a *forward* prediction of macroscopic elastic properties (ii) a sensitivity analysis of macro-scale output parameters as function of input parameters, and (iii) in principle also for an *inverse* theory-guided materials-design search for unknown phases and/or their volumetric ratios that would result in alloys with specific (e.g. application-dictated) macroscopic elasticity. Further development is planned regarding (i) higher-order elastic properties, (ii) application of *inverse* modeling tools, and (iii) description of samples with textures.

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