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Universal Elastic Anisotropy Index

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Practically all elastic single crystals are anisotropic, which calls for an appropriate universal measure to quantify the extent of anisotropy. A review of the existing anisotropy measures in the literature leads to a conclusion that they lack universality in the sense that they are nonunique and ignore contributions from the bulk part of the elastic stiffness (or compliance) tensor. Proceeding from extremal principles of elasticity, we introduce a new universal anisotropy index that overcomes the above limitations. Furthermore, we establish special relationships between the proposed anisotropy index and the existing anisotropy measures for special cases. A new elastic anisotropy diagram is constructed for over 100 different crystals (from cubic through triclinic), demonstrating that the proposed anisotropy measure is applicable to all types of elastic single crystals, and thus fills an important void in the existing literature.

Introduction.—The elastic response of a single crystal is seldom isotropic so that an appropriate measure to quantify the extent of anisotropy is needed. Ledbetter and Migliori [1] rightly point out the importance of elastic anisotropy in diverse applications such as phase transformations, dislocation dynamics, and other geophysical applications, to name a few. More recently, it has been demonstrated that the elastic anisotropy influences the nanoscale precursor textures in shape memory alloys [2] and others [3] have reasoned out the high anisotropy exhibited by polonium simple cubic crystal. The quantification of the elastic anisotropy in terms of a single parameter goes back to Zener who first introduced an anisotropy index for cubic crystals [4]. Subsequently, Chung and Buessem [5] empirically modified the Zener anisotropy index to make it single valued. Both the above measures are applicable to cubic crystals and become meaningless in other crystal classes. Recently, Ledbetter and Migliori [1] defined a general anisotropy measure based on the solution of Christoffel's equations that could be potentially used for all crystal classes. Their definition of elastic anisotropy is based on the shear component of the modulus and neglects the spherical component. Thus, all the currently available measures for anisotropy in the literature lack universality. The above definitions also have limited use in understanding the finite-size scaling behavior (i.e., passage from the level of one crystal to that of an effective or homogenized continuum [6,7]) for a variety of polycrystals [8]. Such a role is played by the universal anisotropy index. In the subsequent sections, we briefly review the existing anisotropy measures and highlight their limitations. We then propose a new universal anisotropy index and establish its relationship to the existing anisotropy measures for special cases.

Zener anisotropy index.—Zener [4] introduced this measure to quantify the anisotropy of cubic crystals

$$A = \frac{2C_{44}}{C_{11} - C_{12}}, \tag{1}$$

where C_{11}, C_{12}, and C_{44} are the three independent components that completely represent the elastic modulus tensor. Some authors [9] prefer to quantify the anisotropy as $\varepsilon = 1 - A$ instead of A. Physically, C_{44} represents the resistance to deformation with respect to a shearing stress applied across the (100) plane in the [010] direction. Similarly, $\frac{(C_{11} - C_{12})}{2}$ represents the resistance to shear deformation by a shear stress applied across the (110) plane in the [1$\bar{1}$0] direction. For a locally isotropic crystal, the above shear resistances would turn out to be identical. Also, for cubic crystals, the bulk resistance ($\frac{C_{11} + 2C_{12}}{3}$) is isotropic. Based on these observations, Zener proposed Eq. (1) to quantify the extent of anisotropy in cubic single crystals. Notice that Zener could have indeed used A^{-1} instead of A to quantify the anisotropy. Thus, A is restricted to cubic crystals and lacks universality.

Chung-Buessem anisotropy index.—In order to obtain a single-valued anisotropy measure, Chung and Buessem [5] proposed the following empirical measure

$$A^C = \frac{G^V - G^R}{G^V + G^R} = \frac{3(A - 1)^2}{3(A - 1)^2 + 25A}, \tag{2}$$

where G^V and G^R are the shear modulus' Voigt and the Reuss estimates, respectively. Note that A^C is generally different if A^{-1} is used instead of A. Thus, A^C is empirically motivated and applicable only to cubic crystals.

Ledbetter-Migliori anisotropy index.—In an attempt to come up with a generic anisotropy measure, Ledbetter and Migliori [1] introduced

$$A^* = v_2^2/v_1^2, \tag{3}$$

where v_1 and v_2 denote the minimum and maximum shear sound-wave velocities among all propagation directions **n**

and polarization directions **p**. Although Ledbetter and Migliori [1] claim that Eq. (3) applies to all seven crystal symmetries, we argue otherwise. First, note that A^* is not a unique anisotropy measure: one could have defined $A^* = v_1^2 / v_2^2$ instead of Eq. (3) to describe the crystal anisotropy. Furthermore, Ledbetter and Migliori [1] conclude that $A^* = A$ for cubic crystals. Now, by definition, $A^* \geq 1$. However, a variety of cubic crystals such as alkali halides (CsF, KBr, KCl, NaF, etc.) have Zener anisotropies less than unity, so that Eq. (3) fails to quantify the anisotropy of such crystals. Thus, A^* lacks universality like all other existing measures.

Perhaps the key reason for the nonuniversal nature of Eqs. (1)–(3) is that such definitions do not account for the full tensorial nature of the elastic stiffness of a given crystal, so that any contribution of the bulk part of that tensor is disregarded. The definitions Eqs. (1)–(3) are indeed helpful as long as one considers cubic crystals that exhibit an isotropic bulk resistance. For all other crystal types, the bulk response is in general anisotropic and one must account for such contributions in order to quantify the extent of anisotropy accurately.

Universal anisotropy index.—We now proceed to derive a new measure to quantify the single crystal anisotropy, overcoming the dilemma of nonuniqueness in its definition and also accounting for the bulk contributions. Consider a particular realization $\omega(\in \Omega)$ of a single crystal exhibiting an anisotropic response subjected to either of these boundary conditions [6]: (i) uniform displacement, $u_i = \varepsilon_{ij}^0 x_j$; (ii) uniform traction, $t_i = \sigma_{ij}^0 n_j$. The first of these yields a stiffness tensor $\mathbf{C}^d(\omega)$ so that the volume averaged stress $\bar{\boldsymbol{\sigma}}(\omega)$ becomes

$$\bar{\boldsymbol{\sigma}}(\omega) = \mathbf{C}^d(\omega){:}\boldsymbol{\varepsilon}^0, \tag{4}$$

while the second one yields a compliance tensor $\mathbf{S}^t(\omega)$:

$$\bar{\boldsymbol{\varepsilon}}(\omega) = \mathbf{S}^t(\omega){:}\boldsymbol{\sigma}^0. \tag{5}$$

Now, recall the extremum principle of elasticity theory

$$\frac{1}{2}\int_B \sigma_{ij}^* \varepsilon_{ij}^* dV - \int_{\partial B_u} t_i^* u_i dS > \frac{1}{2}\int_B \sigma_{ij} \varepsilon_{ij} dV - \int_{\partial B_u} t_i u_i dS$$

$$> -\frac{1}{2}\int_B \sigma_{ij}^{**} \varepsilon_{ij}^{**} dV$$

$$+ \int_{\partial B_t} t_i u_i^{**} dS, \tag{6}$$

where the stars on the left-hand side indicate statically admissible fields, while the double stars on the right-hand side indicate kinematically admissible fields. Clearly, the expression in the middle takes the absolute minimum value of the function of the pair $(\sigma^*{:}\varepsilon^*)$ and the absolute maximum value of the function of the pair $(\sigma^{**}{:}\varepsilon^{**})$. Given that the crystal is homogeneous (although anisotropic), implies that $\mathbf{C}^d(\omega) = \mathbf{S}^t(\omega)^{-1}$, $\forall\ \omega \in \Omega$. Now, by assigning the single crystal orientation uniformly

on a sphere of 3D distributions, and upon ensemble averaging, one obtains an isotropic single crystal response. Thus, the ensemble averaged stiffness and compliance tensors can be expressed in terms of the shear modulus G and the bulk modulus K as follows:

$$\mathbf{C}^V = \langle\mathbf{C}^d\rangle = 2G^V\mathbf{K} + 3K^V\mathbf{J}, \tag{7a}$$

$$\mathbf{S}^R = \langle\mathbf{S}^t\rangle = \frac{1}{2G^R}\mathbf{K} + \frac{1}{3K^R}\mathbf{J}. \tag{7b}$$

In the above, $\mathbf{J}$ and $\mathbf{K}$ represent the spherical and the deviatoric parts of the unit fourth-order tensor. The superscripts V and R represent the Voigt and Reuss estimates, respectively. Note that $\mathbf{C}^V = (\mathbf{S}^R)^{-1}$ in the particular case when the crystal is perfectly isotropic. By contracting Eqs. (7a) and (7b), we obtain

$$\mathbf{C}^V{:}\mathbf{S}^R = \langle\mathbf{C}^d\rangle{:}\langle\mathbf{S}^t\rangle = 5\frac{G^V}{G^R} + \frac{K^V}{K^R}. \tag{8}$$

The above equation takes a minimum value of 6 when the crystal is locally isotropic. At this stage, we introduce the universal anisotropy index:

$$A^U = \mathbf{C}^V{:}\mathbf{S}^R - 6 = 5\frac{G^V}{G^R} + \frac{K^V}{K^R} - 6 \geq 0. \tag{9}$$

Equation (9) can also be interpreted as a generalization of the Zener anisotropy index. This is because instead of taking ratios of individual stiffness coefficients to define anisotropy, we take into account all the stiffness coefficients by recognizing the tensorial nature of the elastic stiffness. A^U is identically zero for locally isotropic single crystals. The departure of A^U from zero defines the extent of single crystal anisotropy and accounts for both the shear and the bulk contributions unlike all other existing anisotropy measures. Thus, A^U represents a universal measure to quantify the single crystal elastic anisotropy.

Discussion.—In the special case of cubic crystals, $K^V = K^R$ ([10]) and Eq. (9) takes the following form relating A^U and A:

$$A^U = \frac{6}{5}\left(\sqrt{A} - \frac{1}{\sqrt{A}}\right)^2. \tag{10}$$

Notice that A^U is a single-valued measure and remains unchanged if A is replaced by A^{-1}. Similarly, using Eqs. (2) and (10), we obtain the following relationship between A^U and A^C:

$$A^U = 10\left(\frac{A^C}{1 - A^C}\right). \tag{11}$$

We plot Eqs. (10) and (11) for a variety of cubic crystals as illustrated in Fig. 1. We observe from the plot that the Zener anisotropy index predicts much different anisotropies for Nb ($A \approx 0.49$) and β-Zr ($A \approx 2.07$) single crystals. On the contrary, our universal anisotropy index ($A^U \approx 0.63$) and the Chung-Buessem anisotropy index ($A^C \approx 0.06$) are single valued for these crystals. Notice
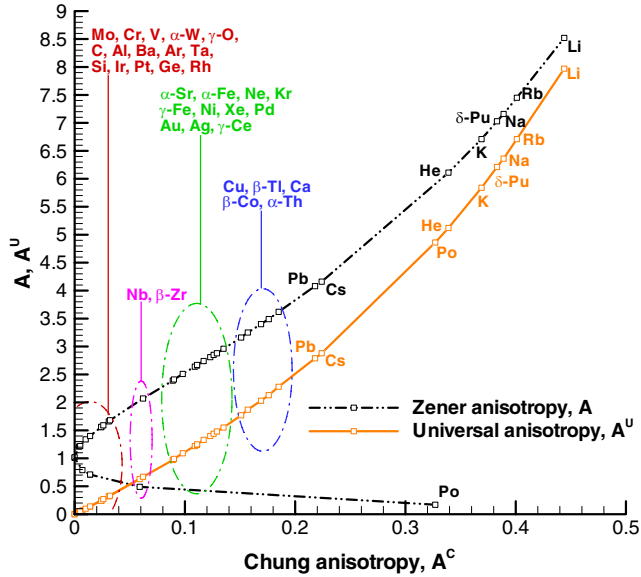
FIG. 1 (color online). Anisotropy measures for cubic crystals (Zener and Ledbetter-Migliori anisotropy measures coincide for cubic crystals). Single crystal data obtained from [3,11].

that the A^C predicts much lower value for anisotropy than all other measures. For a locally isotropic crystal, $A = 1$ but $A^U = A^C = 0$. Also, as A^U increases, it approaches A, albeit predicting lower values for anisotropy.

Let us now consider crystals of hexagonal type. The only measures that can now be used are A^U or A^*. It is indeed difficult to compare these measures directly since the former quantifies the extent of anisotropy based on the departure from zero while the latter quantifies it with the departure from unity. To enable some comparison, we introduce an equivalent Zener anisotropy measure A^{eq} by observing Eq. (10), so that

$$A^{eq} = \left(1 + \frac{5}{12}A^U\right) + \sqrt{\left(1 + \frac{5}{12}A^U\right)^2 - 1}. \quad (12)$$

Note that, for a locally isotropic crystal, $A^{eq} = 1$ while $A^U = 0$. We now compare the three measures, namely A^*, A^U, and A^{eq}, for a variety of hexagonal crystals as depicted in Table I. As expected, one cannot really compare A^* and A^U directly, but A^* and A^{eq} are strikingly identical. For most crystals, the margin of error is 1%. The differences between these measures are much greater in highly anisotropic Cd (16.5%) and Zn (25.5%) single crystals. Table I suggests that the errors are indeed associated with the contributions arising from the bulk part of the modulus.

Elastic anisotropy diagram (EAD).—Using Eq. (9), we construct an elastic anisotropy diagram in the $(\frac{G^V}{G^R}, \frac{K^V}{K^R})$ space as shown in Fig. 2. The advantage of such a diagram is that one can map all the known elastic crystals in one single plot and infer the following.

(i) Physically, the region $(\frac{G^V}{G^R} \leq 1, \frac{K^V}{K^R} \leq 1)$ is inadmissible and hence no crystals lie there.

TABLE I. Comparison of the equivalent Zener anisotropy measure and the Ledbetter-Migliori measure. Single crystal data obtained from [11].

Hexagonal crystal	$\frac{G^V}{G^R}$	$\frac{K^V}{K^R}$	A^*	A^U	A^{eq}	% error
α-Be	1.010	1.003	1.22	0.05	1.24	−1.61
Cd	1.197	1.207	2.18	1.19	2.61	−16.5
α-Co	1.039	1.000	1.49	0.20	1.50	−0.67
α-Dy	1.004	1.000	1.12	0.02	1.13	−0.88
α-Er	1.004	1.001	1.13	0.02	1.15	−1.74
α-Gd	1.006	1.000	1.17	0.03	1.18	−0.85
α-Hf	1.006	1.000	1.18	0.03	1.18	0.00
Ho	1.003	1.000	1.12	0.02	1.12	0.00
α-Lu	1.001	1.002	1.04	0.01	1.07	−2.80
Mg	1.007	1.000	1.19	0.04	1.19	0.00
α-Nd	1.026	1.000	1.37	0.13	1.39	−1.44
α-Pr	1.038	1.000	1.47	0.19	1.48	−0.68
Re	1.022	1.000	1.37	0.11	1.35	1.48
Ru	1.006	1.001	1.17	0.03	1.17	0.00
α-Se	1.017	1.000	1.33	0.08	1.30	2.31
α-Tb	1.005	1.000	1.15	0.02	1.15	0.00
α-Ti	1.038	1.000	1.49	0.19	1.48	0.68
α-Tl	1.396	1.003	3.39	1.98	3.35	1.19
Y	1.008	1.003	1.18	0.04	1.21	−2.48
Zn	1.320	1.222	2.39	1.82	3.21	−25.5
α-Zr	1.021	1.002	1.38	0.11	1.35	2.22

(ii) Lines of constant universal anisotropy index appear as straight lines with a slope of "−5." Thus, an increment along the $(\frac{G^V}{G^R})$ axis influences the anisotropy much more than an identical increment along the $(\frac{K^V}{K^R})$ axis. All crystals lying on the line $(A^U = c)$ are equally anisotropic. This implies that Zn (hexagonal), SnF_2 (monoclinic), bismuth (trigonal), and Cu (cubic) are equally anisotropic with $A^U \approx 1.8$. Similarly, for 1,3,5-triphenylbenzene (orthorhombic) and In (tetragonal), $A^U \approx 3$.

(iii) Almost locally isotropic crystals cluster around $(\frac{G^V}{G^R} \approx 1, \frac{K^V}{K^R} \approx 1)$, irrespective of their crystal class. For example, cubic: α-W, γ-O, etc.; hexagonal: α-Lu, Ho, etc.; monoclinic: diallage, $MgCaSi_2O_6$, etc.; orthorhombic: enstatite, bronzite, etc.; tetragonal: $Sr_4KLiNb_{10}O_{30}$, etc.; trigonal: $LiNbO_3$, Fe_2O_3, etc.

(iv) All cubic crystals lie scattered along the line $\frac{K^V}{K^R} = 1$. The anisotropy index increases as we move to our right along this line. For example, Cs is more anisotropic than Pb. Similarly, Cu is more anisotropic than γ-Ce.

(v) Although indium (In) and tin (Sn) belong to tetragonal crystal class, they are elastically similar to cubic crystals. One can draw similar conclusions for all other crystals lying along $\frac{K^V}{K^R} = 1$.

(vi) The majority of hexagonal crystals get clustered around $(\frac{G^V}{G^R} \approx 1, \frac{K^V}{K^R} \approx 1)$ and are thus the crystal class with least anisotropy consistent with [1].
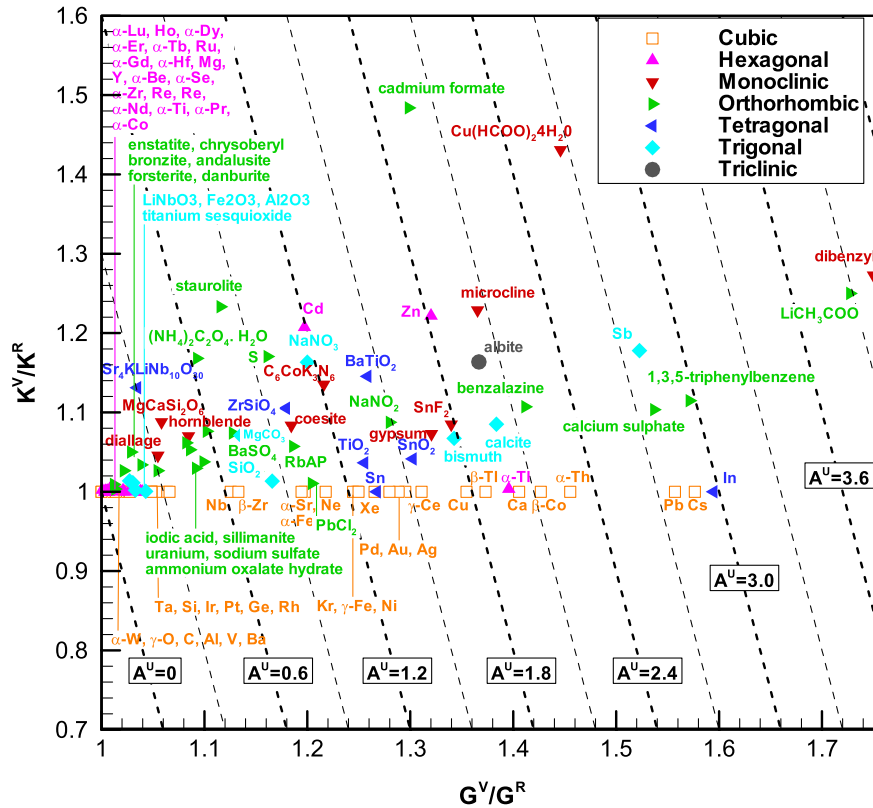
FIG. 2 (color online).   Elastic anisotropy diagram (EAD). Single crystal data obtained from [11,13–17].

Conclusion.—We have proposed a new universal index, A^U, to quantify the single crystal elastic anisotropy. One can readily generalize the concept to characterize the extent of anisotropy in tensors of second rank, third rank, and so on. For example, the anisotropy index for the second order thermal conductivity tensor [12] can be defined as $3(\frac{c^V}{c^R} - 1)$, where c^V and c^R are the Voigt and Reuss estimates in conductivity. Finally, the EAD offers a simple, yet powerful framework to unify and map all the known elastic crystals onto a single diagram. One can further attempt to relate the vacant regions, outliers, and local clustering in the EAD to the atomic lattice with a goal to unveil other mysteries in material elasticity.

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[1] H. Ledbetter and A. Migliori, J. Appl. Phys. **100**, 063516 (2006).
[2] P. Lloveras, T. Castán, M. Porta, A. Planes, and A. Saxena, Phys. Rev. Lett. **100**, 165707 (2008).
[3] D. Legut, M. Friák, and M. Šob, Phys. Rev. Lett. **99**, 016402 (2007).
[4] C. Zener, *Elasticity and Anelasticity of Metals* (University of Chicago, Chicago, 1948).
[5] D. H. Chung and W. R. Buessem, J. Appl. Phys. **38**, 5 (1967).
[6] M. Ostoja-Starzewski, *Microstructural Randomness and Scaling in Mechanics of Materials* (CRC Press, Boca Raton, FL, 2008).
[7] M. Ostoja-Starzewski and J. Schulte, Phys. Rev. B **54**, 278 (1996).
[8] S. I. Ranganathan and M. Ostoja-Starzewski, J. Mech. Phys. Solids **56**, 2773 (2008).
[9] P. S. Spoor, J. D. Maynard, and A. R. Kortan, Phys. Rev. Lett. **75**, 3462 (1995).
[10] R. Hill, Proc. Phys. Soc. London Sect. A **65**, 349 (1952).
[11] H. Ledbetter and S. Kim, *Handbook of Elastic Properties of Solids, Liquids, and Gases* (Academic, New York, 2001).
[12] S. I. Ranganathan and M. Ostoja-Starzewski, Phys. Rev. B **77**, 214308 (2008).
[13] J. G. Berryman, J. Mech. Phys. Solids **53**, 2141 (2005).
[14] J. P. Watt, J. Appl. Phys. **50**, 6290 (1979).
[15] J. P. Watt, J. Appl. Phys. **51**, 1520 (1980).
[16] J. P. Watt and L. Peselnick, J. Appl. Phys. **51**, 1525 (1980).
[17] J. M. Brown, E. H. Abramson, and R. J. Angel, Phys. Chem. Miner. **33**, 256 (2006).