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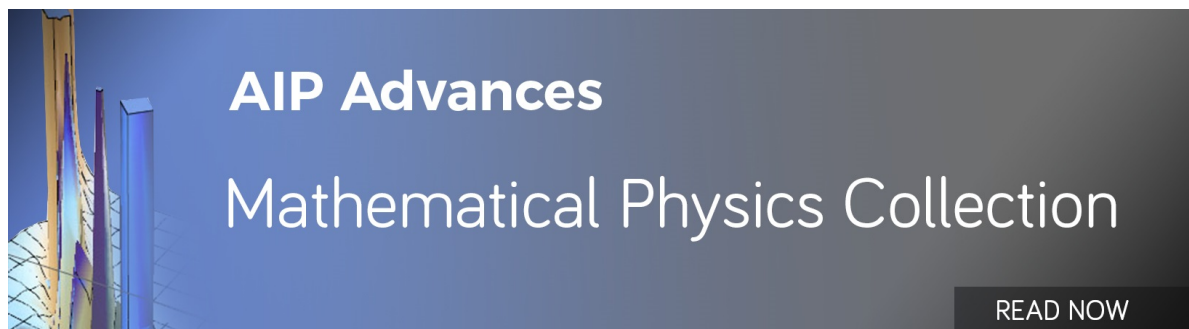
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Elastic anisotropy of crystals

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An anisotropy index seeks to quantify how directionally dependent the properties of a system are. In this article, the focus is on quantifying the elastic anisotropy of crystalline materials. Previous elastic anisotropy indices are reviewed and their shortcomings discussed. A new scalar log-Euclidean anisotropy measure A^L is proposed, which overcomes these deficiencies. It is based on a distance measure in a log-Euclidean space applied to fourth-rank elastic tensors. A^L is an absolute measure of anisotropy where the limiting case of perfect isotropy yields zero. It is a universal measure of anisotropy applicable to all crystalline materials. Specific examples of strong anisotropy are highlighted. A [supplementary material](#) provides an anisotropy table giving the values of A^L for 2,176 crystallite compounds. © 2016 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>). [<http://dx.doi.org/10.1063/1.4962996>]

I. INTRODUCTION

Elastic anisotropy¹ influences a variety of physical processes. Some of these processes include geophysical explorations of the Earth's interior,²⁻⁵ development of plastic deformation in crystals,⁶ enhanced positively charged defect mobility,⁷ microscale cracking in ceramics,⁸ alignment or misalignment of quantum dots,⁹ mechanical properties of nickel-based superalloys,¹⁰ fluid transport in proelastic materials,¹¹ focusing of phonons in crystallites,¹² ultrastructural properties of osteonal bone,¹³ texture in nanoscale shape-memory alloys,¹⁴ and plastic relaxation in thin-film metallics.¹⁵ Thus, it is crucial to be able to quantify the elastic anisotropy to observe effects on these and many other processes for a variety of materials.

Zener¹⁶ proposed an anisotropy factor for crystals of cubic symmetry defined as the ratio of the extreme values of the orientation-dependent shear moduli given by

$$A = \frac{2c_{44}}{c_{11} - c_{12}}, \quad (1)$$

where c_{11} , c_{12} , and c_{44} are the independent single-crystal elastic constants of a crystal having cubic symmetry. Zener's anisotropy factor yields unity when the crystallite is isotropic while a deviation less than or greater than unity signified the degree of anisotropy. Chung and Buessem¹⁷ observed that a crystal is isotropic when the Voigt average of the shear moduli μ^V over all possible orientations was equal to the inverse of the orientation averaged shear compliance μ^R (Reuss average), which motivated the adoption of the factor

$$A^C = \frac{\mu^V - \mu^R}{2\mu^H}, \quad (2)$$

where μ^H is the arithmetic or Hill average given by $\mu^H = (\mu^V + \mu^R) / 2$. A^C can be calculated for any crystal symmetry. However, Chung and Buessem refrained from extending A^C to lower symmetries because, in addition to the shear modulus, the bulk modulus influences the anisotropy of crystals other

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than cubic symmetry.¹⁷ Ledbetter and Migliori¹⁸ noticed that the ratio of the maximum and minimum long-wavelength shear wave velocities in a cubic crystal yielded Zener's anisotropy factor A . They extended this idea to evaluate the anisotropy of non-cubic crystals.¹⁸ Such a metric is appealing because of its connection to easily performed wave-based experiments, but their factor still does not include the influence of the bulk modulus to the anisotropy. Other anisotropy factors have been derived within specific symmetry planes and symmetry directions in a crystal. Lau and McCurdy¹⁹ derived factors based on in-plane phonon focusing for crystals of orthorhombic, tetragonal and hexagonal crystals. Thomsen³ used three anisotropy factors to quantify the anisotropy of transversely isotropic materials (applicable to hexagonal crystals). Tsvankin⁴ developed seven anisotropy parameters for orthorhombic crystals, which followed closely to Thomsen.³

A singular anisotropy index is attractive because of its simplicity when compared to the plurality of anisotropy factors defined for specific planes in crystals. Ranganathan and Ostoja-Starzewski²⁰ derived a universal anisotropy index A^U to provide a singular measure of anisotropy. This index was termed *universal* because of its applicability to all crystal symmetries, a short-coming of all previous measures. A^U is based on the fractional difference between the upper (Voigt) and lower (Reuss) bounds on the bulk (κ^V , κ^R) and shear modulus (μ^V , μ^R), which introduced the influence of the bulk modulus for the first time. The Voigt and Reuss bounds arrive from the statistical elasticity models used to describe elastic deformation of polycrystalline materials. It has long been known that the Voigt and Reuss bounds converge when the crystallites within the aggregate are isotropic.²¹ Thus, the condition of crystallite isotropy exists when $\mathbf{C}^V = \mathbf{C}^R$ where $\mathbf{C}^V = \langle \mathbf{C} \rangle$, $\mathbf{C}^R = (\mathbf{S}^R)^{-1}$, and $\mathbf{S}^R = \langle \mathbf{S} \rangle$ with $\langle \mathbf{C} \rangle$ and $\langle \mathbf{S} \rangle$ being a tensor average operation over all possible orientations of the crystallite's elastic modulus \mathbf{C} and compliance \mathbf{S} , respectively. Recognizing this condition, Ranganathan and Ostoja-Starzewski²⁰ observed that the contraction of the tensor \mathbf{C}^V with \mathbf{S}^R led to a scalar parameter that yielded a minimum value of 6 when a crystallite is isotropic,

$$C_{ijkl}^V S_{ijkl}^R = \frac{\kappa^V}{\kappa^R} + 5 \frac{\mu^V}{\mu^R}, \quad (3)$$

where the summation convention over repeated indices is employed. A^U was then defined as

$$A^U = \frac{\kappa^V}{\kappa^R} + 5 \frac{\mu^V}{\mu^R} - 6 \quad (4)$$

in order for the minimum possible value of A^U to be null for the case of isotropy.²⁰ It was postulated that a larger fractional difference between the Voigt and Reuss estimated bulk or shear modulus would indicate a stronger degree of crystallite anisotropy.²⁰

The universal anisotropy index²⁰ was compared with the previous measures,¹⁶⁻¹⁸ which demonstrated the improved utility of A^U . Furthermore, the evaluation of A^U using tabulated elastic constant values was provided for a number of different crystalline materials, which provided a strong empirical argument for its validation.²⁰

However, similar to the other anisotropy indexes,¹⁶⁻¹⁸ A^U is a relative measure of anisotropy with respect to a limiting value. For example, A^U does not prove that a crystal having $A^U = 3$ has double the anisotropy of another crystal with $A^U = 1.5$. In Sec. II, this limitation is overcome by seeking an anisotropy *distance* measure between \mathbf{C}^V and \mathbf{C}^R , which will provide an absolute measure of anisotropy for different materials.

II. THEORY

A measure of distance between \mathbf{C}^V and \mathbf{C}^R is achieved by making use of distance formulas between tensors.²²⁻²⁴ Norris²² and Norris and Moakher²⁴ used Euclidean and Riemannian distances to determine how close a material of a given symmetry is to a material of another symmetry. These distance measures provide a physically appealing scalar parameter that quantifies the resemblance of materials possibly belonging to nearly disparate symmetry classes. Extending this idea, a measure of distance between \mathbf{C}^V and \mathbf{C}^R is sought in order to provide estimates to the degree of anisotropy present in the elastic constants. A proper distance will yield zero when the crystal is elastically isotropic and will scale correctly for increasing levels of anisotropy.

The log-Euclidean distance first defined by Arsigny et al.²³ is used to define the distance between \mathbf{C}^V and \mathbf{C}^R , which is given by

$$d_L(\mathbf{C}^V, \mathbf{C}^R) = \|\text{Log}(\mathbf{C}_{ijkl}^V) - \text{Log}(\mathbf{C}_{ijkl}^R)\|, \quad (5)$$

where $\|\mathbf{H}\| \equiv (\mathbf{H}_{ijkl}\mathbf{H}_{ijkl})^{1/2}$ is the norm of a general fourth-rank tensor \mathbf{H} .²² The log-Euclidean distance is chosen over the standard Euclidean measures because it is less sparse in the limit of strong anisotropy and it is invariant under inversion, that is, $d_L(\mathbf{C}^V, \mathbf{C}^R) = d_L(\mathbf{C}^{V^{-1}}, \mathbf{S}^R)$.²² In order to simplify Eq. (5), Walpole's decomposition²⁵ is employed to define

$$\mathbf{C}_{ijkl}^V = 2\mu^V J_{ijkl} + 3\kappa^V K_{ijkl}, \quad (6)$$

$$\mathbf{C}_{ijkl}^R = 2\mu^R J_{ijkl} + 3\kappa^R K_{ijkl}. \quad (7)$$

A number of identities based on the contraction of indices illustrates the utility of Walpole's decomposition, such as $J_{ijij} = 5$, $K_{ijij} = 1$, $K_{ijmn}K_{mnlk} = K_{ijkl}$, $J_{ijmn}J_{mnlk} = J_{ijkl}$, $J_{ijmn}K_{mnlk} = 0$, and $K_{ijmn}J_{mnlk} = 0$.²⁵ Upon substituting into Eq. (5), the log-Euclidean distance d_L can be simplified and then defined as the anisotropy index

$$A^L(\mathbf{C}^V, \mathbf{C}^R) = \sqrt{\left[\ln\left(\frac{\kappa^V}{\kappa^R}\right)\right]^2 + 5\left[\ln\left(\frac{\mu^V}{\mu^R}\right)\right]^2}, \quad (8)$$

where \ln is the standard base-e logarithm. The evaluation of the log-Euclidean distance A^L requires the evaluation of the bulk (κ^V, κ^R) and shear (μ^V, μ^R) moduli. Equating the invariants of the fourth-rank elastic moduli and compliance tensors leads to

$$9\kappa^V = c_{11} + c_{22} + c_{33} + 2c_{12} + 2c_{13} + 2c_{23}, \quad (9)$$

$$15\mu^V = c_{11} + c_{22} + c_{33} - c_{12} - c_{13} - c_{23} + 3c_{44} + 3c_{55} + 3c_{66}, \quad (10)$$

$$1/\kappa^R = s_{11} + s_{22} + s_{33} + 2s_{12} + 2s_{13} + 2s_{23}, \quad (11)$$

$$15/\mu^R = 4(s_{11} + s_{22} + s_{33} - s_{12} - s_{13} - s_{23} + 3s_{44} + 3s_{55} + 3s_{66}), \quad (12)$$

which are valid for any crystallite symmetry. The stiffness/compliance relations are obtained from the set of equations produced by the inner product $C_{ijmn}S_{mnlk} = (\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})/2$. The general analytical forms for κ^R and μ^R are too lengthy to be reproduced here. A [supplementary material](#) is provided for the analytical expressions for $\kappa^R, \kappa^V, \mu^R, \mu^V$, and A^L valid for any symmetry (from cubic to triclinic). The [supplementary material](#) provides a detailed procedure with a *Matlab* script for calculating the anisotropy for any material assuming the elastic constants are well known. For the unique case of **cubic** crystallite symmetry, the log-Euclidean anisotropy parameter simplifies into a convenient form and can be written in terms of other previously used anisotropy parameters,^{16,17,20}

$$A^L(\mathbf{C}^V, \mathbf{C}^R) = \sqrt{5} \ln\left(\frac{\mu^V}{\mu^R}\right), \quad (13a)$$

$$= \sqrt{5} \left[\ln\left(\frac{(2+3A)(3+2A)}{25A}\right) \right], \quad (13b)$$

$$= \sqrt{5} \ln\left(\frac{1+A^C}{1-A^C}\right), \quad (13c)$$

$$= \sqrt{5} \ln\left(1 + \frac{A^U}{5}\right). \quad (13d)$$

Equation (13a) shows that A^L , for crystals of cubic symmetry, depends on the ratio of the Voigt and Reuss shear moduli and is independent of the bulk moduli. For lower symmetry, the anisotropy depends on both the bulk and shear modulus.

For all crystallite symmetries, the universal anisotropy index defined by Ranganathan and Ostoja-Starzewski²⁰ is related to the standard Euclidean distance between the averaged stiffness \mathbf{C}^V and averaged compliance \mathbf{S}^R ,

$$d_E(\mathbf{C}^V, \mathbf{S}^R) = \sqrt{A^U + 6}. \quad (14)$$

On the other hand, $A^L(\mathbf{C}^V, \mathbf{C}^R)$ is based on the distance between the averaged stiffnesses \mathbf{C}^V and \mathbf{C}^R , which is more appropriate. Clearly, $A^L(\mathbf{C}^V, \mathbf{C}^R)$ is zero when the crystallite is isotropic (when $\kappa^V = \kappa^R$ and $\mu^V = \mu^R$). Unlike A^U , A^L does not need a normalization factor to indicate a zero-value for the limiting case of isotropy.

III. RESULTS

A survey and comparison of the anisotropy indexes A^L , A^U , A^C , and A was conducted on a dataset of 2,176 crystalline materials. The dataset spans all crystallographic symmetries and includes a variety of species and compounds. Each of the anisotropy measures were evaluated using full elastic tensors generated from first-principle density functional theory.²⁶ A description of the procedure used to generate the elastic constants can be found elsewhere.²⁶ Figure 1 gives a comparison of the proposed parameter A^L to A^U for all 2,176 materials. The materials were sorted in an order of increasing anisotropy defined by A^L as seen in Fig. 1(a); A^U as seen in Fig. 1(b) also follows this ordering for most materials. However, some cases indicate that a given material is more anisotropic than another for A^U and less anisotropic for A^L . This feature can be observed in the close-up subplot for A^U . The greater sparsity of A^U compared to A^L is easily noticed where the most anisotropic crystals are about three orders of magnitude greater than the majority of the materials in the dataset. The materials were color coded according to their symmetry. Their frequency of occurrence in the dataset are as follows: 749 cubic materials, 504 hexagonal materials, 176 trigonal materials, 369 tetragonal materials, 298 orthorhombic materials, 76 monoclinic materials, and 9 triclinic materials. Cubic and hexagonal crystals dominate for the cases extremely low levels of anisotropy (Material #'s;50). The extremely anisotropic materials contain entries belonging to each of the symmetry classes. Notable bands of orthorhombic and monoclinic symmetries exist for larger values of A^L and A^U . An *Excel* file is provided in the [supplementary material](#) for the numerical data contained in Fig. 1. The top 25 most anisotropic materials from the dataset is included in Table I.

MP-ID refers to the *Materials Project* identification numbers assigned to each material.²⁷ Further information on each of the entries in Table I can be accessed by visiting <https://materialsproject.org>. Table I contains entries belonging to each of the 7 crystal symmetry groups. The allotropes of carbon are the most anisotropic materials examined, which causes great sparsity in the calculated values of A^U ranging from 210 to 397.3. Conversely, A^C indicates that the variation in the anisotropy of carbon is far less sparse (from 0.95 to 0.97). A^L assigns anisotropy values from 8.77 to 10.27. The greater variation present in A^L and A^U highlights the influence of the bulk modulus in the calculations, which is neglected in A^C .

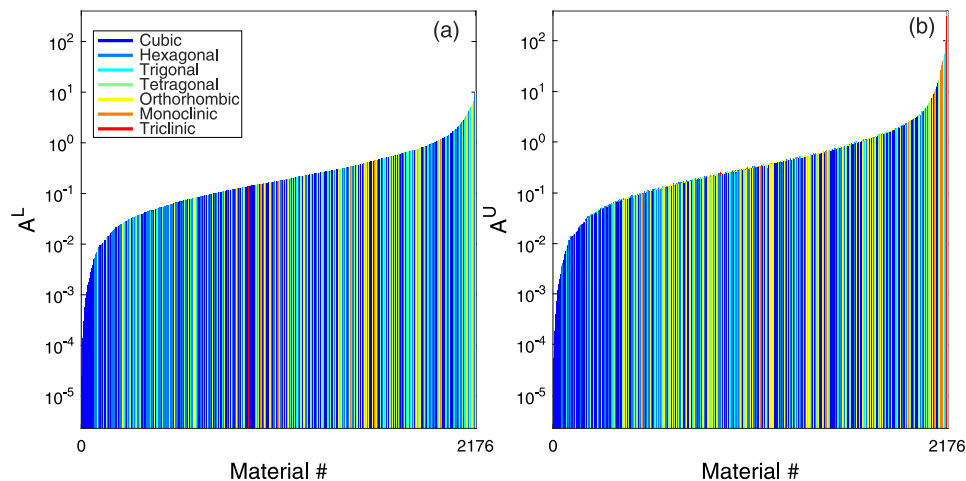


FIG. 1. Visual comparison between A^L (a) and A^U (b) for the 2,176 materials considered in the dataset. The values are sorted by increasing A^L and the corresponding crystal symmetries are color coded.

TABLE I. The top 25 most anisotropic crystalline materials.

MP-ID	Crystal Symmetry	Formula	A^L	A^U	A^C	A
mp-568286	Orthorhombic	C	10.27	397.3	0.97	–
mp-606949	Triclinic	C	9.77	320.5	0.97	–
mp-169	Trigonal	C	9.50	284.0	0.96	–
mp-629015	Hexagonal	BN	9.44	276.8	0.96	–
mp-984	Hexagonal	BN	8.86	217.1	0.95	–
mp-48	Hexagonal	C	8.77	210.0	0.95	–
mp-559976	Trigonal	Ta ₂ CS ₂	7.09	106.1	0.91	–
mp-25587	Monoclinic	LiNiO ₂	7.09	111.5	0.92	–
mp-543096	Trigonal	NiO ₂	6.61	85.1	0.89	–
mp-7581	Trigonal	MoSe ₂	6.54	80.7	0.87	–
mp-5745	Trigonal	Nb ₂ CS ₂	6.49	79.1	0.87	–
mp-540793	Orthorhombic	VCl ₂ O	6.24	72.4	0.87	–
mp-34134	Trigonal	MnO ₂	5.93	61.9	0.85	–
mp-27850	Orthorhombic	TiNCl	5.92	62.0	0.85	–
mp-27863	Orthorhombic	AlClO	5.79	58.3	0.82	–
mp-7868	Hexagonal	PtO ₂	5.75	56.8	0.82	–
mp-12910	Orthorhombic	GeS	5.66	57.7	0.85	–
mp-568346	Orthorhombic	HfBrN	5.45	51.8	0.83	–
mp-32450	Orthorhombic	VBr ₂ O	5.43	49.3	0.79	–
mp-27848	Orthorhombic	TiIN	5.37	47.8	0.79	–
mp-505531	Tetragonal	FeS	5.33	47.2	0.81	–
mp-18717	Cubic	SrVO ₃	5.30	48.6	0.83	3.14
mp-694	Trigonal	VSe ₂	5.22	44.8	0.80	–
mp-765892	Monoclinic	MnCoO ₄	5.21	45.2	0.81	–
mp-617	Trigonal	PtO ₂	5.17	43.4	0.79	–

TABLE II. Comparison of A^L between cubic single crystals and triclinic crystalline compounds. The Zener anisotropy index A is also included for comparison to A^L for the cubic crystals.

Cubic	A	A^L	Triclinic	A^L
Cu	3.21	0.69	CuSO ₄ · 5H ₂ O	0.32
K	6.71	1.73	KH ₃ C ₂ O ₄ · 2H ₂ O	1.06
Na	7.48	1.76	NaHC ₂ O ₄ · H ₂ O	1.07
Li	15.76	2.25	LiHC ₂ O ₄ · H ₂ O	1.37

The anisotropy index A^L allows comparisons between very different materials from various symmetry groups as illustrated in Table I. As an additional example, a comparison between cubic single crystals and triclinic crystalline compounds is shown in Table II.

The values in Table II were calculated from experimentally measured single-crystal elastic constants.²⁸ The triclinic compounds CuSO₄ · H₂O, KH₃C₂O₄ · 2H₂O, NaHC₂O₄ · H₂O, LiHC₂O₄ · H₂O each have one of the cubic crystallite constituents Cu, K, Na, and Li. Increasing levels of anisotropy are present in Cu, K, Na, and Li, which correlates with increasing levels of anisotropy in the corresponding triclinic compounds.

IV. CONCLUSION

The anisotropy index A^L , defined in Eq. (8), provides an absolute measure of anisotropy in crystalline materials. A^L is an improvement to previous measures A and A^C because it is valid for all of the crystallite symmetries. Conceptually, A^L is closely related to A^U . However, A^L differs from A^U in that it is an absolute distance measure between C^V and C^R .

SUPPLEMENTARY MATERIAL

See [supplementary material](#) for (1) A *Matlab* function *anisotropy.m* that calculates A^L from 21 elastic constant inputs. Symmetry relations for higher symmetry than triclinic is included. (2) A tabulation of the anisotropy measures, A^L , A^U , and A^C , is given for 2,176 inorganic crystal compounds where the elastic constants were generated by first-principle calculations based on density functional theory.²⁶ The table corresponds with the data that is illustrated in Fig. 1.

ACKNOWLEDGMENTS

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