Necessary and sufficient elastic stability conditions in various crystal systems

Félix Mouhat and François-Xavier Coudert[*]

PSL Research University, Chimie ParisTech–CNRS, Institut de Recherche de Chimie Paris, 75005 Paris, France

While the Born elastic stability criteria are well known for cubic crystals, there is some confusion in the literature about the form they should take for lower-symmetry crystal classes. Here we present closed form necessary and sufficient conditions for elastic stability in all crystal classes, as a concise and pedagogical reference to stability criteria in noncubic materials.

PACS number(s): 62.20.D−, 63.10.+a

I. INTRODUCTION

The fundamental understanding of the conditions of the mechanical stability of unstressed crystalline structures dates back to the seminal work of Born and coauthors in the 1940's [1], and was consolidated in his 1954 book [2]. This and later textbooks [3–5] usually state the generic requirements for the elastic stability of crystal lattices, and give simplified equivalents of the generic conditions for some high-symmetry crystal classes. In particular, in the case of cubic crystals, the conditions of stability reduce to a very simple form:

$$C_{11} - C_{12} > 0, \quad C_{11} + 2C_{12} > 0, \quad C_{44} > 0. \quad (1)$$

The above equations for the cubic crystal system are well known, and are often called the "Born stability criteria." We noticed, however, through a review of the recent literature on the experimental measurements and first principles calculations of elastic constants of solids, that there is a large amount of confusion about the form that these conditions should take for other crystal classes, including hexagonal, tetragonal, rhombohedral, and orthorhombic classes. In more than a few cases, incorrect generalizations of the cubic criteria have been published [6–10]; this is particularly frequent for orthorhombic crystals [11–17]. In other papers, the authors rely on conditions that are necessary but not sufficient [18]. As long as the diagonal elastic constants C_{ii} are dominant, this leads to wrong quantitative analyses, but does not change the qualitative picture (whether or not a specific crystal is stable). However, we identified at least one case where accounting for the proper stability criteria did change the conclusions drastically, meaning that a system (MOF-74 material with CH_4 guest molecules) was identified as stable when it is not [10].

In this paper, we summarize the generic elastic stability conditions for crystals, and present necessary and sufficient conditions for each crystal class. We also detail the crystal classes where no analytical necessary and sufficient conditions exist.

II. GENERAL ELASTIC STABILITY CONDITION

The elastic behaviors of a lattice are described by its matrix of second-order elastic constants,

$$C_{ij} = \frac{1}{V_0}\left(\frac{\partial^2 E}{\partial \varepsilon_i \partial \varepsilon_j}\right), \quad (2)$$

where E is the energy of the crystal, V_0 its equilibrium volume, and $\boldsymbol{\varepsilon}$ denotes a strain [19]. This elastic matrix (also called the stiffness matrix) has size 6×6 and is symmetric: It is thus composed of 21 independent components. The crystal class of the material considered yields additional symmetry constraints, further reducing the number of independent elastic constants. For arbitrary homogeneous deformation by an infinitesimal strain, the energy of the crystal is therefore given by the following quadratic form:

$$E = E_0 + \frac{1}{2}V_0 \sum_{i,j=1}^{6} C_{ij}\varepsilon_i\varepsilon_j + O(\varepsilon^3). \quad (3)$$

A crystalline structure is stable, in the absence of external load and in the harmonic approximation [20], if and only if (i) all its phonon modes have positive frequencies for all wave vectors (*dynamical stability*), and (ii) the elastic energy, given by the quadratic form of Eq. (3), is always positive ($E > 0, \forall \varepsilon \neq 0$). This latter condition is called the *elastic stability* criterion. As first noted by Born [1], it is mathematically equivalent to the following *necessary and sufficient* stability conditions:

(1) The matrix $\boldsymbol{C}$ is definite positive;

(2) all eigenvalues of $\boldsymbol{C}$ are positive;

(3) all the leading principal minors of $\boldsymbol{C}$ (determinants of its upper-left k by k submatrix, $1 \leqslant k \leqslant 6$) are positive, a property known as Sylvester's criterion; and

(4) an arbitrary set of minors of $\boldsymbol{C}$ are all positive. It can be useful to choose, for example, the *trailing* minors, or any other set.

These are four possible formulations of the generic *Born elastic stability conditions* for an unstressed crystal. They are valid regardless of the symmetry of the crystal studied, and are not linear.

Finally, from these conditions we can deduce some *necessary but not sufficient* conditions. Fedorov [4], in particular, noted that the condition on principal minors implies that all diagonal elements are positive ($C_{ii} > 0, \forall i$), but this alone is not strong enough to ensure stability. Another example of a *necessary* condition is

$$(C_{ij})^2 < C_{ii}C_{jj} \quad \forall i,j. \quad (4)$$

III. EXPRESSIONS FOR SPECIFIC LAUE CLASSES

We now turn our attention to express closed form expressions of the *necessary and sufficient* elastic stability conditions

---

*fx.coudert@chimie-paristech.fr

TABLE I. Laue groups and number of independent second-order elastic constants C_{ij}. We follow the naming convention of Wallace [5] (I/II) to distinguish Laue classes within the same crystal system.

Crystal system	Laue class	Point groups	C_{ij}'s
Triclinic	$\bar{1}$	$1, \bar{1}$	21
Monoclinic	$2/m$	$2, m, 2/m$	13
Orthorhombic	mmm	$222, 2mm, mmm$	9
Tetragonal (II)	$4/m$	$4, \bar{4}, 4/m$	7
Tetragonal (I)	$4/mmm$	$4mm, 422, \bar{4}2m, 4/mmm$	6
Rhombohedral (II)	$\bar{3}$	$3, \bar{3}$	7
Rhombohedral (I)	$\bar{3}m$	$32, 3m, \bar{3}m$	6
Hexagonal (II)	$6/m$	$6, \bar{6}, 6/m$	5
Hexagonal (I)	$6/mmm$	$6mm, 622, \bar{6}2m, 6/mmm$	5
Cubic (II)	$m\bar{3}$	$23, m\bar{3}$	3
Cubic (I)	$m\bar{3}m$	$432, \bar{4}3m, m\bar{3}m$	3

for 11 Laue classes, as described in Table I. For each class, the table also gives the number of *independent* elastic constants in the stiffness matrix. We focus here on the crystalline systems in three dimensions, but this analysis can also be extended in very similar terms to other dimensions (e.g., one- and two-dimensional quasicrystals) [21,22].

For each crystal system and Laue class, closed form expressions of the necessary and sufficient elastic stability conditions can be found following a number of different approaches. The one we have chosen is to develop the series of minors of the stiffness matrix, in an order chosen to minimize the degree of the polynomials involved. For this, one reorders the matrix into block diagonal form and expresses the minors starting with the smallest blocks. Another way to view this approach is to express the quadratic form of the energy and reduce it by successively "completing the square" in the variables, taken in a sequence appropriate to the symmetries [23,24]. This is formally equivalent and gives identical expressions for the conditions. Finally, we also checked the results presented below by means of direct calculation with computer algebra software [25], expanding the characteristic polynomial of the stiffness matrix in each case, and factoring it.

A. Cubic crystal system

The cubic crystal system has the simplest form of elastic matrix, with only three independent constants, C_{11}, C_{12}, and C_{44}:

$$C_{\text{cubic}} = \begin{pmatrix} C_{11} & C_{12} & C_{12} & & & \\ . & C_{11} & C_{12} & & & \\ . & . & C_{11} & & & \\ & & & C_{44} & & \\ & & & & C_{44} & \\ & & & & & C_{44} \end{pmatrix} \quad (5)$$

(in this notation, dots are used to indicate nonzero elements constrained by the symmetric nature of the matrix). The three Born stability criteria for the cubic system are well known:

$$C_{11} - C_{12} > 0, \quad C_{11} + 2C_{12} > 0, \quad C_{44} > 0. \quad (6)$$

They are necessary and sufficient. Here we merely note that the first two conditions imply that $C_{11} > 0$, so it does not need to be noted as an extra condition, as is sometimes done. Also, the first condition can be equivalently stated as $C_{11} > |C_{12}|$.

B. Hexagonal and tetragonal classes

Both Laue classes of the hexagonal crystal system, as well as the tetragonal (I) class ($4/mmm$), have the same form for the elastic matrix:

$$C_{\text{hexa/tetra I}} = \begin{pmatrix} C_{11} & C_{12} & C_{13} & & & \\ . & C_{11} & C_{13} & & & \\ . & . & C_{33} & & & \\ & & & C_{44} & & \\ & & & & C_{44} & \\ & & & & & C_{66} \end{pmatrix}. \quad (7)$$

Crystals of the tetragonal (I) class thus have six independent elastic constants, while those with a hexagonal crystal system have only five, due to the added relation

$$C_{66} = (C_{11} - C_{12})/2. \quad (8)$$

By direct calculation of the eigenvalues of the stiffness matrix above, one can derive the following four *necessary and sufficient* conditions for elastic stability in the hexagonal and tetragonal (I) case:

$$C_{11} > |C_{12}|, \quad 2C_{13}^2 < C_{33}(C_{11} + C_{12}),$$
$$C_{44} > 0, \quad C_{66} > 0 \quad (9)$$

(where the condition on C_{66} is redundant with the first one, for the hexagonal case).

The tetragonal (II) class ($4/m$) features an extra elastic constant, C_{16}, bringing the total of independent C_{ij}'s to seven:

$$C_{\text{tetra II}} = \begin{pmatrix} C_{11} & C_{12} & C_{13} & & & C_{16} \\ . & C_{11} & C_{13} & & & -C_{16} \\ . & . & C_{33} & & & \\ & & & C_{44} & & \\ & & & & C_{44} & \\ . & . & & & & C_{66} \end{pmatrix}. \quad (10)$$

The conditions for this are the same as for the tetragonal (I) class, Eq. (9), with the exception of the condition on C_{66} being replaced by $2C_{16}^2 < C_{66}(C_{11} - C_{12})$. Thus the complete *necessary and sufficient* Born stability criteria for tetragonal (II) class are

$$C_{11} > |C_{12}|, \quad 2C_{13}^2 < C_{33}(C_{11} + C_{12}),$$
$$C_{44} > 0, \quad 2C_{16}^2 < C_{66}(C_{11} - C_{12}). \quad (11)$$

C. Rhombohedral classes

Crystals in the rhombohedral (I) class (Laue class $\bar{3}m$) feature six independent elastic constants,

$$
\boldsymbol{C}_{\text{rhombo I}} = \begin{pmatrix}
C_{11} & C_{12} & C_{13} & C_{14} & & \\
\cdot & C_{11} & C_{13} & -C_{14} & & \\
\cdot & \cdot & C_{33} & & & \\
\cdot & \cdot & & C_{44} & & \\
& & & & C_{44} & C_{14} \\
& & & & \cdot & C_{66}
\end{pmatrix},
$$

(12)

where, as in the hexagonal case, $C_{66} = (C_{11} - C_{12})/2$. We therefore obtain the following four *necessary and sufficient* conditions:

$$
\begin{aligned}
C_{11} &> |C_{12}|, \quad C_{44} > 0, \\
C_{13}^2 &< \tfrac{1}{2} C_{33}(C_{11} + C_{12}), \\
C_{14}^2 &< \tfrac{1}{2} C_{44}(C_{11} - C_{12}) \equiv C_{44} C_{66}.
\end{aligned}
$$

(13)

For the rhombohedral (II) class, there is one more independent elastic constant, namely, C_{15}:

$$
\boldsymbol{C}_{\text{rhombo II}} = \begin{pmatrix}
C_{11} & C_{12} & C_{13} & C_{14} & C_{15} & \\
\cdot & C_{11} & C_{13} & -C_{14} & -C_{15} & \\
\cdot & \cdot & C_{33} & & & \\
\cdot & \cdot & & C_{44} & & -C_{15} \\
\cdot & \cdot & & & C_{44} & C_{14} \\
& & & \cdot & \cdot & C_{66}
\end{pmatrix}.
$$

(14)

The resolution leads to a case similar to the rhombohedral (I) class, but with a stricter version of the last criterion:

$$
\begin{aligned}
C_{11} &> |C_{12}|, \quad C_{44} > 0, \\
C_{13}^2 &< \tfrac{1}{2} C_{33}(C_{11} + C_{12}), \\
C_{14}^2 + C_{15}^2 &< \tfrac{1}{2} C_{44}(C_{11} - C_{12}) \equiv C_{44} C_{66}.
\end{aligned}
$$

(15)

The criteria presented in Eq. (15) are rigorously *necessary and sufficient*.

D. Orthorhombic systems

Finally, we come to the crystal systems with lower symmetry and a larger number of independent elastic constants. The stiffness matrix for an orthorhombic crystal has the following form, with nine constants and no relationships between them:

$$
\boldsymbol{C}_{\text{ortho}} = \begin{pmatrix}
C_{11} & C_{12} & C_{13} & & & \\
\cdot & C_{22} & C_{23} & & & \\
\cdot & \cdot & C_{33} & & & \\
& & & C_{44} & & \\
& & & & C_{55} & \\
& & & & & C_{66}
\end{pmatrix}.
$$

(16)

There are three trivial eigenvalues for this matrix, namely, C_{44}, C_{55}, and C_{66}, all of which need to be positive. However, the eigenvalues of the upper-left 3×3 block do not have a closed

form expression. They are the three roots of the following cubic polynomial:

$$
\begin{aligned}
\lambda^3 &- \lambda^2 (C_{11} + C_{22} + C_{33}) \\
&+ \lambda \big(C_{11}C_{22} + C_{11}C_{33} + C_{22}C_{33} - C_{12}^2 - C_{23}^2 - C_{13}^2 \big) \\
&+ C_{11}C_{23}^2 + C_{22}C_{13}^2 + C_{33}C_{12}^2 \\
&- C_{11}C_{22}C_{33} - 2C_{12}C_{13}C_{23}.
\end{aligned}
$$

(17)

One can, however, find a closed form expression equivalent to the generic criteria, through the requirement that leading principal minors be positive. This reduces to the following *necessary and sufficient* Born criteria for an orthorhombic system:

$$
\begin{aligned}
&C_{11} > 0, \quad C_{11}C_{22} > C_{12}^2, \\
&C_{11}C_{22}C_{33} + 2C_{12}C_{13}C_{23} - C_{11}C_{23}^2 - C_{22}C_{13}^2 - C_{33}C_{12}^2 > 0, \\
&C_{44} > 0, \quad C_{55} > 0, \quad C_{66} > 0.
\end{aligned}
$$

(18)

The conditions obtained are not all linear, but polynomial functions of the elastic constants (because the largest nondiagonal block in the stiffness matrix has size 3×3 and all coefficients are independent).

We feel it is important to note here that some authors have presented in the literature simpler stability conditions for orthorhombic crystals, many of them linear [6,11,12]. To quote only one, Wu *et al.* [6] claim that "it is known that for orthorhombic crystals, the mechanical stability requires the elastic stiffness constants satisfying the following conditions":

$$
\begin{aligned}
&C_{ii} > 0, \quad C_{ii} + C_{jj} - 2C_{ij} > 0, \\
&C_{11} + C_{22} + C_{33} + 2(C_{12} + C_{13} + C_{23}) > 0.
\end{aligned}
$$

(19)

These conditions seem to be a natural extension of the well-known cubic case, but they are incorrect. Indeed, it is easy to verify formally, with CAS (Computer Algebra System) software, that these conditions are *necessary but not sufficient* [26].

E. Monoclinic and triclinic systems

Monoclinic and triclinic crystal systems have 13 and 21 independent elastic constants, respectively. Given the complexity of the equations obtained, we will not show those here. When studying such low-symmetry crystals, it is usually more convenient to keep the stiffness coefficients in matrix form. In particular, the generic *necessary and sufficient* criterion that all eigenvalues of $\boldsymbol{C}$ be positive is easy to check with simple linear algebra routines.

If, nevertheless, one wishes to obtain closed form expressions for the stability conditions of monoclinic and triclinic systems, they can be obtained as six polynomials in the elastic constants by writing out that the leading principal minors of $\boldsymbol{C}$ be positive. For monoclinic systems, the polynomials will be of degree 4 (at most), while for triclinic crystals they will be of degree 6 (at most). Simpler forms, including fully linear or quadratic, that have sometimes been proposed in the literature [6], are incorrect.

IV. CONCLUSION

Here we gathered closed form expressions for necessary and sufficient elastic stability criteria (also called Born stability conditions) depending on the Laue classes of crystals. While high-symmetry crystal systems allow for simple formulas, these cannot be generalized trivially to lower-symmetry systems. In particular, the cubic system is the only one for which these conditions are all linear. Hexagonal, tetragonal, and rhombohedral systems have quadratic stability criteria. Conditions for orthorhombic crystals involve cubic polynomials, while monoclinic and triclinic systems can be expressed as quartic and sextic polynomials, respectively.

Finally, we note that the conditions of elastic stability described here for an unstressed system can be readily generalized to systems under an arbitrary external load σ by introducing an elastic stiffness tensor $\boldsymbol{B}$ under load, defined as (in tensorial notation) [20,27]

$$
\begin{aligned}
B_{ijkl} = C_{ijkl} + \tfrac{1}{2}(\delta_{ik}\sigma_{jl} + \delta_{jk}\sigma_{il} \\
+ \delta_{il}\sigma_{jk} + \delta_{jl}\sigma_{ik} - 2\delta_{kl}\sigma_{ij}).
\end{aligned} \tag{20}
$$

The resulting symmetry of the $\boldsymbol{B}$ tensor might be lower than that of $\boldsymbol{C}$, if the load is not isotropic. Elastic stability conditions can then be derived, as a function of crystal system and symmetry of the external load, by applying the formalism of this paper [28].

ACKNOWLEDGMENT

[1] M. Born, Math. Proc. Cambridge Philos. Soc. **36**, 160 (1940).

[2] M. Born and K. Huang, *Dynamics Theory of Crystal Lattices* (Oxford University Press, Oxford, UK, 1954).

[3] J. F. Nye, *Physical Properties of Crystals* (Oxford University Press, Oxford, UK, 1957).

[4] F. I. Fedorov, *Theory of Elastic Waves in Crystals* (Springer, Berlin, 1968).

[5] D. C. Wallace, *Thermodynamics of Crystals* (Wiley, New York, 1972).

[6] Z.-J. Wu, E.-J. Zhao, H.-P. Xiang, X.-F. Hao, X.-J. Liu, and J. Meng, Phys. Rev. B **76**, 054115 (2007).

[7] X. F. Li and Z. L. Liu, J. At. Mol. Sci. **3**, 78 (2011).

[8] C.-Y. Niu and J.-T. Wang, Phys. Lett. A **378**, 2303 (2014).

[9] Y.-Y. Fu, Y.-W. Li, and H.-M. Huang, Chin. Phys. Lett. **31**, 116201 (2014).

[10] P. Canepa, K. Tan, Y. Du, H. Lu, Y. J. Chabal, and T. Thonhauser, arXiv:1409.7124.

[11] S. K. R. Patil, S. V. Khare, B. R. Tuttle, J. K. Bording, and S. Kodambaka, Phys. Rev. B **73**, 104118 (2006).

[12] Z. Wu, X. Hao, X. Liu, and J. Meng, Phys. Rev. B **75**, 054115 (2007).

[13] C. Jiang, S. G. Srinivasan, A. Caro, and S. A. Maloy, J. Appl. Phys. **103**, 043502 (2008).

[14] P. Vajeeston, P. Ravindran, B. Hauback, and H. Fjellvåg, Int. J. Hydrogen Energy **36**, 10149 (2011).

[15] P. Vajeeston, P. Ravindran, and H. Fjellvåg, RSC Adv. **2**, 11687 (2012).

[16] J. Feng, APL Mater. **2**, 081801 (2014).

[17] C. Fan, Y. Jin, J. Li, and X. Dong, Sci. Rep. **4**, 6993 (2014).

[18] E. Calderon, M. Gauthier, F. Decremps, G. Hamel, G. Syfosse, and A. Polian, J. Phys.: Condens. Matter **19**, 436228 (2007).

[19] Here we use the standard Voigt notation for the indices of strains, stresses, and elastic tensors, contracting a pair of Cartesian indices into a single integer $1 \leqslant i \leqslant 6$: $xx \to 1$, $yy \to 2$, $zz \to 3$, $yz \to 4$, $xz \to 5$, $xy \to 6$. A 3×3 symmetric strain or stress matrix thus becomes a vector of length 6, and the stiffness tensor of rank four becomes a 6×6 matrix.

[20] G. Grimvall, B. Magyari-Köpe, V. Ozoliņš, and K. A. Persson, Rev. Mod. Phys. **84**, 945 (2012).

[21] R. Wang, W. Yang, C. Hu, and D.-H. Ding, J. Phys.: Condens. Matter **9**, 2411 (1997).

[22] J. Wang, R. Zhang, D. H. Ding, and R. Wang, Acta Crystallogr., Sect. A: Found. Crystallogr. **55**, 558 (1999).

[23] R. Hill and F. Milstein, Phys. Rev. B **15**, 3087 (1977).

[24] F. Milstein, R. Hill, and K. Huang, Phys. Rev. B **21**, 4282 (1980).

[25] Wolfram Research, Inc., Mathematica, Version 10.0.1, Champaign, IL (2014).

[26] In fact, let us take the example of $C_{11} = 1/2$, $C_{22} = C_{33} = 1$, $C_{12} = -1$, and $C_{13} = C_{23} = 0$. It fulfills all the conditions of Wu *et al.* [6], yet fails both of our *necessary and sufficient* orthorhombic criteria.

[27] T. H. K. Barron and M. L. Klein, Proc. Phys. Soc. **85**, 523 (1965).

[28] Additionally, we note that in experimental measurements under finite load, this external load is in most cases frame dependent and often deformation sensitive. On the other hand, Eq. (20) applies only to situations of constant external load, and needs to be adapted if that is not the case [29, 23] .

[29] R. Hill, Math. Proc. Camb. Philos. Soc. **77**, 225 (1975).