A critical study of the elastic properties and stability of Heusler compounds: Cubic $\text{Co}_2YZ$ compounds with $L2_1$ structure

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Elastic constants and their derived properties of various cubic Heusler compounds were calculated using first-principles density functional theory. To begin with, $\text{Cu}_2\text{MnAl}$ is used as a case study to explain the interpretation of the basic quantities and compare them with experiments. The main part of the work focuses on $\text{Co}_2$-based compounds that are $\text{Co}_2\text{MnM}$ with the main group elements $M = \text{Al, Ga, In, Si, Ge, Sn, Pb, Sb, Bi}$, and $\text{Co}_2\text{TM}$ with the main group elements $\text{Si}$ or $\text{Ge}$, and the 3d transition metals $T = \text{Sc, Ti, V, Cr, Mn, and Fe}$. It is found that many properties of Heusler compounds correlate to the mass or nuclear charge $Z$ of the main group element.

Blackman’s and Every’s diagrams are used to compare the elastic properties of the materials, whereas Pugh’s and Poisson’s ratios are used to analyze the relationship between interatomic bonding and physical properties. It is found that the Pugh’s criterion on brittleness needs to be revised whereas Christensen’s criterion describes the ductile–brittle transition of Heusler compounds very well. The calculated elastic properties give hint on a metallic bonding with an intermediate brittleness for the studied Heusler compounds.

The universal anisotropy of the stable compounds has values in the range of $0.57 < A_U < 2.73$. The compounds with higher $A_U$ values are found close to the middle of the transition metal series. In particular, $\text{Co}_2\text{ScAl}$ with $A_U = 0.01$ is predicted to be an isotropic material that comes closest to an ideal Cauchy solid as compared to the remaining $\text{Co}_2$-based compounds. Apart from the elastic constants and moduli, the sound velocities, Debye temperatures, and hardness are predicted and discussed for the studied systems. The calculated slowness surfaces for sound waves reflect the degree of anisotropy of the compounds.

I. INTRODUCTION

There is a broad interest in Heusler compounds owing to the multitude of different thermal, electrical, magnetic, and transport properties that are realized in a rather simple crystalline structure. Owing to both applications and fundamental interests, such as superconductivity, heavy fermions, the Kondo effect, the Hall effect, and half-metallic ferromagnetism, these compounds are among the most studied materials. Regular Heusler compounds crystallize in a cubic fcc lattice with the space group $Fm\overline{3}m$. In certain cases, the cubic phases of regular Heusler compounds undergo a martensite-austenite phase transition to a tetragonal lattice, whereby the symmetry changes to $I4/mmm$. In fact, both the cubic and the tetragonal phases have attracted considerable attention owing to their half-metallic ferromagnetic and spin-transfer-torque applications. Knowledge of the stability of each of these phases is crucial for industrial applications as well as fundamental research.

New Heusler compounds have been suggested to be stable in many theoretical works. However, in many cases it is experimentally found that it is not possible to synthesize these compounds. A possible reason for this is that not all stability criteria are respected in theoretical calculations. In fact, mostly all the used stability criteria are necessary but not sufficient. This implies that a suggested compound that fulfills a particular criterion may not exist as it possibly violates other criteria. One of these necessary, but not sufficient, criteria is the total energy, or the energy of formation, satisfying the condition $E_{\text{compound}} < \sum E_{\text{elements}}$ for a compound to be stable. Further, the formation energy of the suggested compound needs to be the minimum on the “convex hull” taking into account all the competing phases. Otherwise, it would decompose into other compounds with lower energies. For example, the appearance of different binaries (XY, XZ, or other similar combinations) may lead to a lower total energy as compared to a single ternary ($X_2YZ$), and thus, hinder the formation of a Heusler compound.

Another important criterion is the mechanical stability of a predicted structure. According to Born,[5] a necessary condition for the thermodynamic stability of a crystal lattice is that the crystals have to be mechanically stable against arbitrary (small) homogeneous deformations. In fact, this is the main concept of elastic constants. Elastic constants provide important information concerning the strength of materials, and often act as stability criteria or order parameters in the study of the problem of structural transformation.[6,15] Further physical properties, such as hardness, velocity of sound, Debye temperature, and melting point are also related to the elastic constants.[6,12] The information is not only essential requirements for industrial applications but also for fundamental research. Examples of the latter case are the superconducting and heavy fermion systems, in which a drastic change of elastic constants and related properties have been obtained upon phase transition.[6,13]
There are several reports on calculations of elastic constants and phase stability. Members of the series of cubic Co$_2$TM (T = transition element, M = main group element) Heusler compounds have been studied previously to some extent by various authors. In many cases, only the three independent elastic constants and the bulk modulus are calculated. In only a few cases, experiments were carried out to measure the hardness and melting temperatures, and to compare them with the calculations. However, most works have been carried out for specific cases, and almost not all relevant properties have been calculated and compared with experiments or properties of other compounds.

The present report is intended to investigate the mechanical properties of a variety of Co$_2$-based Heusler compounds by calculating their elastic constants. To begin with, Cu$_2$MnAl, as a typical Heusler compound, is studied to explain the basic quantities and their interpretation. Results for Co$_2$MnM (M = main group element), and Co$_2$TM (T = 3d-metal, M = Al, Si) are listed and discussed with an in-depth analysis of the physical properties and chemical bonding. As elastic constants are derived from the second derivative of the energy with respect to the lattice displacements, the use of an accurate energy calculator is crucial. Here, the full-potential all-electron method was used to calculate the elastic constants, and related properties. The relationship between the interatomic bonding and the physical properties is considered using Pugh’s and Poisson’s ratios. The Blackman’s diagram provides complementary information about the bonding character of the Heusler compounds. A covalent to metallic bonding with an intermediate ductility or brittleness is found for the studied Heusler compounds. Several other physical properties have been extracted from the elastic constant calculations.

The present work concentrates on the half-metallic Co$_2$-based Heusler compounds that have a high impact on magnetoelectronics. The results for the elastic properties of tetragonal and phase change materials that exhibit magnetic shape memory and magnetocaloric effects will be published elsewhere. In many, only two more calculations are needed besides the bulk modulus that is found from the equation of state. A necessary supplementary condition in the calculation of the elastic constants is the conservation of volume when strain (or stress) is applied. Table I summarizes the applied strains used in the present work to determine the elastic constants. The charge density and relaxed lattice parameters are found by fitting the calculated energy–volume relation to the Birch–Murnaghan equation of state.

Most Co$_2$-based Heusler compounds are among the half-metallic ferromagnetic materials, thus only ferromagnetic ordering has been studied here. See References [22-26] for details of the electronic and magnetic structure of the investigated compounds. The basic equations for the calculations of the elastic constants are discussed in the following. More details are provided in Appendix. The bulk moduli and relaxed lattice parameters are found by fitting the calculated energy–volume relation to the Birch–Murnaghan equation of state.

There are numerous ways to apply different strains and their combinations to the crystal. For cubic crystals, there are three independent elastic constants and only two more calculations are needed besides the bulk modulus that is found from the equation of state. A necessary supplementary condition in the calculation of the elastic constants is the conservation of volume when strain (or stress) is applied. Table I summarizes the applied strains used in the present work to determine the elastic constants. The applied strains are illustrated in Figure 1. More details are found in Reference [21] and in the Appendix.

In the present calculations, four distortions of each type in the range of $-3\% \leq x \leq +3\%$ were applied to the relaxed structure with $V_0$ from the structural optimization, which is the equation of state fitted to $E_{tot}(V)$. The different distortions are sketched in Figure 1. The energies $E(x)$ of the monoclinic, orthorhombic and tetragonal strain types were fitted to a 4th order polynomial $E(x) = E_0 + a_2x^2 + a_3x^3 + a_4x^4$. Finally, the $B$ and the $a_2$ values were used to determine the elastic constants and their derived quantities. The elastic constants reported below are averaged over the values determined by applying tetragonal or orthorhombic strains. The equations for the properties calculated from the elastic con-
TABLE I. Applied strains to cubic structure are listed here.
The isotropic strain type (0) is not volume conserving, it is not directly used but used together with the lattice parameter optimization that yields the bulk modulus. The consequences of the strain types are sketched in Figure 1.

| Type | Strain | $\Delta E/V_0$ |
|------|--------|----------------|
| (0)  | isotropic | $e_1 = \delta$  
          |         | $e_2 = \delta$  
          |         | $e_3 = \delta$  |
| (1)  | tetragonal | $e_1 = \delta$  
          |         | $e_2 = \delta$  
          |         | $e_3 = \delta (2 + \delta)/(1 + \delta)^2$  |
| (2)  | orthorhombic | $e_1 = \delta$  
          |         | $e_2 = -\delta$  
          |         | $e_3 = \delta^2/(1 - \delta^2)$  |
| (3)  | monoclinic | $e_1 = \delta^2/(1 - \delta^2)$  |

FIG. 1. (Online color) Strain types for calculation of the elastic constants in cubic systems. (a) shows the cubic Heusler structure with $F m\overline{3}m$ symmetry. (0)...(3) sketch the strain types and resulting distortions according to Table I. Please note that (1) results in $x = y \neq z$ whereas in (2) all three lattice parameters are different: $x \neq y \neq z$. See Table II for details of the strain types.

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TABLE II. Elastic properties of Cu$_2$MnAl.
The experimental and optimized lattice parameters ($a$) are given in Å. Elastic moduli $B$ (bulk), $G$ (shear), $E$ (Young), hardness parameter $H$, elastic constants $c_{ij}$, $C'$, and Cauchy pressure $p_C$ are given in GPa. $k$, $\nu$, $\zeta$, $A_e$ and $A_U$ are dimensionless quantities. Experimental values for the $c_{ij}$ are taken from Reference[34]. Values for 0 K are extrapolated from the temperature dependence shown in Figure 3 of Reference[34].

|          | Calculated | Experiment |
|----------|------------|------------|
|          | 0 K        | 300 K      |
| $a$ Å    | 5.934      | 5.9615     |
| $c_{11}$ GPa | 143.7      | 128.1      |
| $c_{12}$ GPa | 116.1      | 101.5      |
| $c_{44}$ GPa | 117.6      | 104.4      |
| $C'$ GPa | 27.6       | 26.6       |
| $p_C$ GPa | -1.5       | -1.3       |
| $B$ GPa  | 125.3      | 110.4      |
| $G$ GPa  | 52.5       | 47.9       |
| $E$ GPa  | 138.0      | 125.6      |
| $H$ GPa  | 6.43       | 6.06       |
| $k$      | 2.41       | 2.30       |
| $\nu$    | 0.32       | 0.31       |
| $\zeta$  | 0.866      |            |
| $A_e$    | 9.64       | 7.85       |
| $A_U$    | 7.97       | 7.17       |

III. RESULTS

To begin with, the results for the classical Heusler compound Cu$_2$MnAl are presented and compared with experiments, because it is one of the few Heusler compounds for which measured values of the elastic constants are available. Table II compares the calculated elastic properties with the experimental work of Michelutti et al.[34]. As seen in Table II, the calculated elastic constants agree well with the experiment, where the overestimation of about 10% that is observed could be due to the intrinsic properties of the calculational method[23] or due to uncertainties of the experimental set-up. The other calculated properties, such as elastic moduli, Cauchy pressure, and velocity of sound show excellent agreement with those found in the experiment.

The elastic constants of Cu$_2$MnAl are listed in Table II. $c_{11}$, which represents stiffness against principal strains, is higher than $c_{44}$, which represents shear deformation. The shear ($G$) and tetragonal shear $C'$ moduli are also low compared to the bulk modulus. This implies that Cu$_2$MnAl has the lowest resistance against shear deformations. The cross-sections on (110) and (001) crystallographic planes of Young’s moduli of Cu$_2$MnAl are shown in Figure 2.(c) and (f). It is seen that the anisotropy of Young’s modulus is noticeable in both the planes. The directions where the maxima appear correspond to the high-fracture energy directions, which are along (111) in the (110) plane and (110)-direction in the (001) plane.

The Kleinman’s parameter $\zeta$ describes the relative positions of the atoms under strain. The calculated value of $\zeta \approx 0.9$ for Cu$_2$MnAl suggests that the atomic positions are rather rigid against distortions of the lattice. The tetragonal shear modulus $C' = C'/2 \approx 13 \ldots 20$ GPa is the smallest modulus and thus it is the main constraint on stability. Both anisotropies, the Zener ratio $A_e$ and universal anisotropy $A_U$, are about 8. This is a rather large value and may suggest elastic instability of Cu$_2$MnAl in the $L2_1$ structure, as will be discussed in
the following.

The use of Blackman’s and Every’s diagrams is an efficient way to compare the elastic properties of cubic materials. In both types of diagrams, dimensionless quantities that are ratios of different moduli are correlated. Figure 2 summarizes the results of the present work in such diagrams. Blackman’s diagram compares in a simple way the ratios $F_{ij} = c_{ij}/c_{11}$ of the elastic constants in a plot of $F_{12}(F_{44})$, whereas Every’s diagram is more complicated. It compares $s_2 = (1 - F_{12} - 2F_{44})/(1 + 2F_{44})$ as a function of $s_3 = (1 - F_{14})/(1 + 2F_{44})$. Born’s shear criterion restricts $F_{14}$ to positive values and the spinodal and Born criteria restrict $F_{12}$ to the range $-0.5 < F_{12} < 1$. Therefore, no additional restrictions appear for Blackman’s diagram and all values within the entire range of Figure 2(a) are allowed. In Every’s diagram, the values for stable systems need to fall into the triangle with $(s_2, s_3)$ equal to $(-1/2, 1/2)$-(1,0)-(1,3/2) as is marked in Figure 2(b).

From both the diagrams shown in Figure 2 it is seen that the Heusler compounds calculated in the present work are close to the Cauchy line, where the Cauchy pressure vanishes. All the studied compounds are in the region where the anisotropy index is positive. Obviously, Cu2MnAl has one of the highest anisotropies and comes close to the line of $2/m$, $3m$ symmetry breaking phase transitions as marked in Every’s diagram. The Co2-based compounds will be discussed below in more detail.

Table III summarizes the physical properties of Cu2MnAl that are related or derived from the elastic properties. The measured sound velocities at room temperature are in the range from 4553 m/s to 6003 m/s for longitudinal modes depending on the direction of propagation and the polarization. The average sound velocity in the transverse modes is about $(3791 \pm 3)$ m/s. The values calculated with Anderson’s approach are in the same order of magnitude.

The Debye temperature was calculated from the mode averaged sound velocity $\bar{v}$ and in the quasi-harmonic approach. Both methods result in values of about 395 K. This is marginally higher than the experimental value. The slightly larger values are typical for the acoustical approaches that neglect the optical phonons.

![Figure 2](image1.png)

**FIG. 2.** (Online color) Crystalline, electronic, and elastic structure of Cu2MnAl. Compared are (a) Cubic structure with (110) plane highlighted. (b) The electron charge density plot in the (110) plane. (c) The (110) cross-sections of Young’s modulus. (d) Cubic structure with (001) plane highlighted. (e) The electron charge density plot in the (001) plane. (f) The (001) cross-sections of Young’s modulus.

![Figure 3](image2.png)

**FIG. 3.** (Online color) (a) Blackman and (b) Every diagrams. The full line refers to the isotropic case, where $A_e = 1$, the dashed line refers to the case where the Cauchy pressure $p_C$ vanishes: $c_{44} = c_{12}$. Ideal Cauchy solids appear at the point of intersection of the two lines. In Every’s representation (b), the stability triangle according to the Born criteria is marked by dash-dotted lines. Its three sides are given by the line of cubic isomorphous phase transitions (cipt) and two lines of symmetry breaking phase transitions (sbpt).

| Property          | Calculated  | Experiment  |
|-------------------|-------------|-------------|
|                  | 0 K         | 300 K       |
| $a$ (Å)          | 5.934       | 5.9615      |
| $\rho$ (kg/m$^3$) | 6637        | 6550        |
| $m$ (g/mol)      | 209.01      |             |
| $v_l$ (m/s)      | 5421        |             |
| $v_t$ (m/s)      | 2807        | see text    |
| $\bar{v}$ (m/s)  | 3142        |             |
| $\Theta_D^{CC}$ (K) | 397        | 372         |
| $\Theta_D^{CC}$ (K) | 1.88        | 0.72        |
| $f_{\nu}$        | 0.72        | 0.74        |
| $\Theta_D^{CC}$ (K) | 395        |             |
| $T_m^{CC}$ (K)   | 1402        |             |
| $H_V^{CC}$ (GPa) | 4.38        |             |

Figure 3 shows the three sheets of the slowness surface of Cu2MnAl. Pronounced extrema, arising from the
large anisotropy of the compound, are observed for all
the three modes. The pressure (p) wave has the highest
phase velocity. The minima of its slowness appear along
{111}-type directions that is along the space diagonals
and the maxima are found along the {001}-type, principal
axes. The maxima of the slowness of the fast shear
wave (s₂) are along the {111}-type directions. The slown-
ess surface of the slow shear wave (s₁) appears to be
more complicated; its maxima appear along {110}-type
directions. The two shear modes are sixfold degenerate,
that is, their slowness is the same at the six [00s]-type
points along the three principal axes.

One may roughly categorize materials as ductile (ma-
alleable) or as brittle with respect to mechanical character-
istics, as for example, machinability. For various applica-
tions, the materials need to be malleable, for instance if
they are required to be used as wires. Ductile materials
usually exhibit metallic bonding, whereas high brittleness
indicates a more covalent or ionic character of the bonds.
The transition region between these subjective criteria is
blurred. Because of the importance in various applica-
tions, the malleability criteria are of great significance.
Pugh’s and Poisson’s ratios are very helpful mechanical
parameters in the characterization of malleability (brittle
or ductile).

Pettifor[39] proposed the criterion that a positive
Cauchy pressure indicates metallic bonds whereas neg-
ative Cauchy pressures are typical in the case of coval-
ent bonds. Another older criterion is based on Pugh’s
criterea[38]. According to the so-called “Pugh’s criterion”,
many publications[39][40][41] indicate that the critical value
(kcr) that separates brittle (k ≤ kcr) and ductile (k ≥ kcr)
materials is around 1.75 or k−1 ≈ 0.571. It is worthwhile
to mention that Pugh’s ratio for a cubic, isotropic Cauchy
solid is kCauchy = 5/3 = 1.66, as shown in Figure 5(a). The
behavior of the Heusler compounds investigated in
the present work is summarized in Figure 5 where the
Cauchy pressure is plotted as a function of Pugh’s ra-
tio. The two criteria are drawn as vertical and horizontal
lines. Following the value for Pettifor’s and Pugh’s crite-
ria, most of the studied compounds should be classified as
ductile or metallic materials. There is an obvious contra-
bition between the empirical rules and the observation
that the CO₂-based Heusler compounds are all brittle
instead of ductile. In particular, it appears that Pugh’s
criterion needs to be modified.

Poisson’s ratio ν is related to Pugh’s ratio by[43]

\[ ν = \frac{3k - 2}{6k + 2}. \quad (1) \]

The valid range of Pugh’s ratio (0 < k ≤ ∞) restricts
Poisson’s ratio to -1 < ν ≤ 1/2. The Poisson ratio
ν0 = 0 is obtained for k₀ = 2/3. Materials with ν < 0 are
called “auxetic”. Typically, Poisson’s ratio of covalent
materials is small ν ≈ 0.1, whereas it is greater than 0.33
for metallic materials. Poisson’s ratio indicates the de-
gree of directionality of the covalent bonds. Smaller Pois-
on’s ratios indicate a stronger degree of covalent bonding
resulting in higher hardness. The so-called “Frantsevich
rule” is widely used as a criterion for brittleness, which
is based on the tables of elastic properties in the book by
Frantsevich et al[42]. According to this rule, compounds
with a Poisson ratio of νcr ≤ 0.33 are brittle and those
with νcr ≥ 0.33 are ductile or malleable. It should be
noted that this value is not given explicitly in Reference[43]. It is based on properties reported for the ma-
terials tabulated by Frantsevich and was later accepted
as empirical rule.

Referring to the original work[39], Pugh also did not
suggest a criterion. However, Pugh only mentioned that
Ir was the least malleable metal (k = 1.74) and Au was
the most malleable metal (k = 6.14). Based on present
knowledge, it is obvious that Ir is hard and brittlen[44]
and hence, the critical value could possibly be between
1.74 and 6.14. Based on the relation between k and ν,
it follows that the critical Poisson’s ratio νcr ≈ 1/3 of
Frantsevich’s rule corresponds to a critical Pugh’s ratio
of kcr ≈ 2.66 (kcr−1 ≈ 3/8), so that the two empirical
rules only differ in the exact number that distinguishes
between the two types of behaviors.

Generally, it may be considered that materials with
ν = 0 are absolutely brittle, whereas those with ν = 1/2
are perfectly ductile. Christensen[45] used the failure the-
ory to describe the mechanical properties. He introduced
a nanoscale variable κ, which characterizes the relative
size of the bond bending and the bond stretching effects.
Further, he related it to renormalized Poisson’s or Pugh’s
ratios and defined the ductility D by:

\[ D = (1 - κ)^2 \]

\[ κ = 1 - \frac{2ν}{1 + ν} = \frac{2}{3k}. \]

According to this relation, materials with D = 0 are
absolutely brittle, those with D = 1 are perfectly ductile,
and the brittle–ductile transition takes place at D_B/D =
1/2. The latter implies that

\[ κ_{B/D} = 1 - \sqrt{1/2}, \quad (3) \]

and thus, the critical values of the Poisson’s and Pugh’s
ratios defined as,

\[ ν_{B/D} = \frac{1}{3\sqrt{2} - 1} \]

\[ k_{B/D} = \frac{2}{3\sqrt{2} - 1}. \]

ν_{B/D} ≈ 0.31 is close to νcr of Frantsevich’s rule. Fur-
ther, it corresponds to a critical Pugh’s ratio of k_{B/D} ≈
2.3 (k_{B/D}−1 ≈ 0.44), that is clearly larger than the so-called
Pugh’s criterion. It is worthwhile to note that the ductil-
ity of a Cauchy solid becomes D_Cauchy = (3/5)^2 = 0.36,
from the extreme elements, which are elemental Au, and 2
ductility to Pugh’s ratio. It is obvious that the com-
ments from experiments, fcc–Au as respectively. Cu
vertical dash-dotted lines correspond to the metallic–covalent
Co–
2
−
−

is plotted in the range of the calculated compounds, whereas
D
is denoted by a spherical symbol. Note that Poisson’s ratio
ria of Frantsevich and Christensen, respectively. Cu
2
ure 2(a) and (d). Figure 2(b) and (c) show the charge
analysis was used to analyze the charge density
MnAl in the (110) plane. Bader’s QTAIM analysis was used to analyze the charge density
and magnetic moments. The results of the QTAIM anal-
ysis are listed in Table IV, where a charge transfer is
C in the form of diamond, as the most ductile and most
brittle elements, respectively.

The above discussion and the experimental observa-
tions on various Heusler compounds lead to the con-
clusion that Frantsevich’s rule (or better Christensen’s
ductility criterion) is suitable for the compounds studied
here, whose behaviors lie on the border-line between brittleness and ductility.

The crystalline structure of Cu2MnAl is shown in Figure 2(a) and (d). Figure 2(b) and (c) show the charge
densities and Young’s modulus of Cu2MnAl in the (110)
plane. Figure 2(e) and (f) show the charge densities and
Young’s modulus of Cu2MnAl in the (001) plane. Bader’s
QTAIM analysis was used to analyze the charge density
and magnetic moments. The results of the QTAIM anal-
ysis are listed in Table IV, where a charge transfer is
observed. On the average, about 0.9 electrons are trans-
ferred from the Mn and Al atoms to the Cu atoms with
relatively larger contribution from the Al atoms. The Mn
atoms carry a magnetic moment of 3.45 µB, whereas Cu
and Al exhibit only a negligible polarization.

which implies that Cauchy solids should be more brittle
than ductile.

The behavior of the Heusler compounds is shown in
Figure 6 that relates Poisson’s ratio and Christensen’s
ductility to Pugh’s ratio. It is obvious that the com-
pounds — and in particular Cu2MnAl — are far away
from the extreme elements, which are elemental Au, and

FIG. 4. (Online color) Calculated slowness surfaces of Cu2MnAl. The slowness is given in (km/s)−1.

FIG. 5. (Online color) Ductile-brittle diagram for various Co2-based Heusler compounds.
Values are given as open circles. The horizontal dashed and vertical dash-dotted lines correspond to the metallic–covalent and ductile-brittleness criteria of Petifor and Pugh, respectively. Cu2MnAl is marked by a spherical symbol and an ideal Cauchy solid by an asterix. The values for the extremal elements from experiments, fcc–Au as “most soft” and diamond–C as “most hard”, are given for comparison. The solid line connecting them is drawn for better comparison.

FIG. 6. (Online color) Poisson’s ratio and ductility for various Co2-based Heusler compounds. Values are given as open circles. The horizontal dashed and Dash-dotted lines correspond to the ductile-brittleness criteria of Frantsevich and Christensen, respectively. Cu2MnAl is denoted by a spherical symbol. Note that Poisson’s ratio is plotted in the range of the calculated compounds, whereas the ductility is shown in the entire intervals of D and k−1.
The QTAIM critical points of Cu$_2$MnAl and their properties are summarized in Table IV. There are, indeed, three different nuclei that act as attractors. The cage critical point $c$ is found between Mn and Al along the [001] axis and acts as a repeller, which is the absolute minimum of the charge density. Further, two bond critical points $b_{1,2}$ are located between Cu and Al ($b_1$), and between Cu and Mn ($b_2$). The third bond critical point is located in between the Mn atoms along the [001] direction. When the two ring critical points $r_{1,2}$ are also considered, the Morse sum of the numbers $n_i$ of the different critical points vanishes ($n_n - n_b + n_r - n_s = 0$), as expected for crystals.

### Table IV. QTAIM analysis of Cu$_2$MnAl

| $V$ [Å$^3$] | $n_e$ | $q$ | $m$ [µB] |
|------------|------|----|----------|
| Cu         | 15.547 | 29.86 | -0.86 | 0.03 |
| Mn         | 12.228 | 24.57 | 0.43 | 3.45 |
| Al         | 8.969  | 11.71 | 1.28 | 0.04 |

The QTAIM critical points of Cu$_2$MnAl and their properties are summarized in Table V. There are, indeed, three different nuclei that act as attractors. The cage critical point $c$ is found between Mn and Al along the [001] axis and acts as a repeller, which is the absolute minimum of the charge density. Further, two bond critical points $b_{1,2}$ are located between Cu and Al ($b_1$), and between Cu and Mn ($b_2$). The third bond critical point is located in between the Mn atoms along the [001] direction. When the two ring critical points $r_{1,2}$ are also considered, the Morse sum of the numbers $n_i$ of the different critical points vanishes ($n_n - n_b + n_r - n_s = 0$), as expected for crystals.

### Table V. QTAIM critical point analysis of Cu$_2$MnAl

| $pg$ | type | position | mult | name |
|------|------|----------|------|------|
| $O_h$ | nucleus | 0 | 0 | 4 | Mn |
| $O_h$ | nucleus | 1/2 | 1/2 | 1/2 | 4 | Al |
| $C_{3v}$ | ring | 0.397 | 0.397 | 0.397 | 1/2 | 4 | Cu |
| $C_{3v}$ | ring | 0.878 | 0.878 | 0.878 | 1/2 | 4 | Cu |
| $D_{2h}$ | ring | 0 | 0.302 | 0.302 | 1/2 | 4 | Mn |
| $C_{2v}$ | ring | 0 | 0.287 | 0.213 | 1/2 | 4 | Mn |
| $C_{4v}$ | cage | 0 | 0 | 0.254 | 1/2 | 4 | Mn |

The analysis of the bonding type with the properties of the critical points is discussed in Reference [22]. Metallic systems exhibit a flat electron density $\rho$ throughout the valence region. The flatness $f = \rho_{\text{min}}/\rho_{\text{max}}$ is a measure of the metallicity of the compounds. $\rho_{\text{min}}$ is the cage critical point, at which the density is minimum, and $\rho_{\text{max}}$ is the highest density among all the bond critical points. For Cu$_2$MnAl, if $f = 0.684$. This is of the same order of magnitude as the flatness in Cu or Fe (both ≈ 0.57; see Reference [22]), whereas compounds with covalent bonding typically have $f$ ratios of less than 0.1. From the large electronic flatness, the bonding in Cu$_2$MnAl is clearly metallic.

In the following, the results of the calculations for various Co$_2$Mn$_2$ compounds are discussed. These compounds, containing Mn in particular, are of much interest in spintronic applications. The Mn containing compounds are discussed in the first part. The second part discusses the variation of the 3d transition metal in Co$_2$Mn$_2$ when the main group element $M$ is attached to Al or Si, and the 3d transition metal $T$ ($T = $ Sc, Ti, V, Cr, Mn, Fe) is varied.

#### A. Results for Co$_2$Mn$M$ ($M =$ main group element)

In this section, the elastic and mechanical properties of the Mn containing Heusler compounds Co$_2$Mn$_2$M ($M =$ Al, Ga, In, Si, Ge, Sn, Pb, Sb, Bi) are discussed. Table VI compares the mechanical properties of the Co$_2$Mn$_2$M compounds. It may be noted that the compounds with main group heavy elements (In, Pb, Sb, Bi) have not been synthesized up to now and Heusler compounds with this composition most probably do not exist. They are used here to complete the trends when changing the main group elements. It should also be noted that those compounds are stable — at least from the Born-Huang criteria (These criteria are discussed in the Appendix).

The relaxed lattice parameters $a_{\text{opt}}$ agree well with the experimental values. They exhibit the typical trend that the lattice parameter increases with the nuclear charge $Z$ of the main group element. The elastic constants of the Co$_2$Mn$_2$M compounds follow the general inequality $B > c_{44} > G > c' > 0$. Here, the isotropic shear or rigidity modulus $G$ is not the main constraint on stability. The smallest values are obtained for $c' = C'/2$, that is the tetragonal shear modulus is the limiting parameter for the stability of the cubic $L2_1$ structure of the investigated compounds. The values of $c'$ are in the range of 37–68 GPa and thus far above the values required to force tetragonal instabilities in the vicinity of ambient pressure. The lowest values for the bulk moduli are obtained for the compounds containing the main group, heavy elements, which exhibit large lattice parameters. The Pb and Bi containing compounds exhibit comparably low values of the rigidity moduli. Within each group, the values of the hardness parameter decrease with increasing $Z$ of the main group elements. As compared to the previous calculations [25], the elastic constants and bulk modulus fit quite well for most compounds, and the differences are lesser than 20 GPa for each quantity.

Pugh’s ratio $k$ of the Co$_2$Mn$_2$M compounds ranges from about 1.83 to 2.46 with a mean value of $k = 2.1$. Similarly, Poisson’s ratio $\nu$ falls also in a narrow range 0.27–0.32 with a mean value of $\nu = 0.29$. A convenient way of quantifying the degree of off-axis anisotropy in the elastic constants for a cubic crystal is to use the Zener ratio. Here, Zener ratio ($A_{\text{z}}$) exhibits no trend of a dependency on the main group element. It exhibits the
TABLE VI. Elastic properties of Co$_2$MnM ($M =$ main group element). The experimental ($\alpha_{\text{exp}}$) and optimized ($\alpha_{\text{opt}}$) lattice parameters are given in Å. Elastic moduli $B$, $G$, $E$, hardness parameter $H$, elastic constants $c_{ij}$, $C^i$, and Cauchy pressure $p_c$ are given in GPa. $k$, $\zeta$, $A_e$, and $A_U$ are dimensionless quantities.

| $M$ | Al | Ga | In | Si | Ge | Sn | Pb | Sb | Bi |
|-----|----|----|----|----|----|----|----|----|----|
| $\alpha_{\text{exp}}$ | 5.755$^b$ | 5.770$^b$ | 5.654$^b$ | 5.743$^b$ | 6.000$^b$ | 5.643 | 5.730 | 5.987 | 6.102 |
| $\alpha_{\text{opt}}$ | 5.700 | 5.718 | 5.974 | 310.5 | 272.8 | 233.6 | 200.1 | 6.019 | 6.184 |
| $c_{11}$ | 267.3 | 242.2 | 194.4 | 151.9 | 112.0 | 74.7 | 63.8 | 106.4 | 72.4 |
| $c_{12}$ | 155.3 | 167.5 | 130.6 | 174.2 | 160.0 | 138.3 | 124.5 | 133.6 | 112.3 |
| $c_{44}$ | 160.4 | 150.5 | 131.8 | 156.9 | 137.9 | 125.0 | 103.8 | 106.4 | 72.4 |
| $C^i$ | 112.0 | 74.7 | 63.8 | 136.3 | 112.8 | 95.3 | 76.6 | 101.7 | 78.8 |
| $p_c$ | -5.1 | 17.0 | -1.2 | 17.3 | 22.1 | 13.3 | 20.7 | 27.2 | 40.8 |
| $B$ | 192.6 | 192.4 | 151.9 | 219.6 | 197.6 | 170.0 | 149.7 | 167.5 | 139.5 |
| $G$ | 105.3 | 86.6 | 75.2 | 112.3 | 96.4 | 85.0 | 69.3 | 79.1 | 56.7 |
| $E$ | 267.2 | 225.9 | 193.6 | 287.9 | 248.7 | 218.5 | 180.1 | 205.1 | 149.8 |
| $H$ | 16.2 | 11.3 | 10.7 | 16.4 | 13.5 | 12.1 | 8.0 | 10.8 | 6.8 |
| $k$ | 1.83 | 2.22 | 2.02 | 1.96 | 2.05 | 2.00 | 2.16 | 2.12 | 2.46 |
| $\nu$ | 0.27 | 0.30 | 0.29 | 0.28 | 0.29 | 0.29 | 0.30 | 0.30 | 0.32 |
| $\zeta$ | 0.692 | 0.779 | 0.764 | 0.676 | 0.696 | 0.701 | 0.767 | 0.681 | 0.699 |
| $A_e$ | 2.86 | 4.05 | 4.14 | 2.30 | 2.44 | 2.62 | 2.75 | 2.09 | 1.84 |
| $A_U$ | 1.46 | 2.73 | 2.85 | 0.88 | 1.02 | 1.21 | 1.88 | 0.68 | 0.46 |

$^b$Ref.11$^c$, Ref.12$^d$

The smallest value for Co$_2$MnBi (1.84) and the largest value for Co$_2$MnIn (4.14). The latter value is exceptional (compare with the next section) and points to a large elastic anisotropy in the material. Another method is the usage of the so-called universal anisotropy index ($A_U$), which shows the same tendency as the Zener ratio. Especially for Co$_2$MnSb, in spite of the value of the anisotropy being in a reasonable range, this compound does not exist, and tested samples exhibit phase separation. The anisotropy does only judge on the structural stability but not on the chemical stability.

Figure 7 shows the Blackman’s diagram using the elastic data of the Co$_2$MnM Heusler compounds given in Table VI. All $F$ ratios fall in the allowed range for mechanical stability. The values of $F_{12}$ appear close together in a region around the Cauchy line where the Cauchy pressure vanishes. They also fall in the region of positive Zener ratios ($A_e > 1$). The figure suggests the type of bonding, covalent or metallic. A positive Cauchy pressure is suggestive of greater degree of metallic bonding. On the contrary, when the Cauchy pressure is negative, there appears to be greater degree of covalent bonding. As discussed in the foregoing and in agreement with the Poisson’s ratio, all the studied compounds are metallic and on the borderline between brittleness and ductility. This is in agreement with experiments, in which all these compounds have a silvery metallic luster.

In fact, directional dependent plots of rigidity $G(\hat{r})$ and Young’s $E(\hat{r})$ moduli are an alternative visual way of showing the Zener anisotropy. The implication of the elastic anisotropy on the elastic moduli will be illustrated for the two borderline cases with the largest (In) and the smallest (Bi) anisotropy. The three dimensional distributions of $G(\hat{r})$ and $E(\hat{r})$ are shown in Figure 8 for Co$_2$MnIn and Co$_2$MnBi.

The anisotropy of Young’s modulus of the In and Bi containing compounds is displayed in Figures 8(a) and (b) that show the three dimensional distribution $E(\hat{r})$. The pronounced anisotropy of the In containing compound is clearly visible. Figures 8(c) and (d) show the three dimensional distribution $G(\hat{r})$ of the rigidity moduli of the two compounds. Again, the differences in the anisotropy of the moduli are clearly visible. Comparing the distribution of the moduli, it is obvious that Young’s modulus is largest in the (111)-type directions whereas the rigidity modulus is largest in the (100)-type directions, that is along the cubic axes. This behavior is generic for all compounds listed in Table VI and a direct consequence of the condition $A_e > 1$. These anisotropic compounds exhibit different responses to stress or strain when tested in different directions. The anisotropic property is particularly important for applications where mechanical stress is applied to the materials, directly or by thermal expansion and contraction.

B. Results for Co$_2$TM ($T =$ 3$d$-metal, $M =$ Al, Si)

In this section, the influence of the 3$d$ transition metal on the elastic and mechanical properties of selected Co$_2$ based Heusler compounds is discussed. Table VII compares the elastic properties of the Co$_2$TM ($T =$ Sc, Ti,
FIG. 7. (Online color) Blackman diagram for Heusler compounds based on Co and Mn. The full line is for the isotropic case, where $A_e = 1$, the dashed line is for the limit where the Cauchy pressure $p_C$ vanishes: $c_{44} = c_{12}$. Different symbols are used for the main group elements from different groups: IIIa (Al, Ga, In), IVa (Si, Ge, Sn, Pb), Va (Sb, Bi), V, Cr, Mn, Fe and $M$ = Al, Si compounds. Al and Si were selected as the main group elements, because they exhibit the most complete series over the 3$d$ transition metals that exist in reality. The Sc compounds as well as $Co_2CrSi$ have not been synthesized up to now and do not possibly exist. They are used here to complete the trends of the properties when changing the transition metal. Similar to the compounds with heavy main group elements reported above, those compounds are stable — at least from the Born-Huang criteria.

The elastic constants of the $Co_2TM$ compounds follow the general inequality $B > c_{44} > G > C'/2 > 0$ as was also observed above for the Mn-containing compounds with varying main group elements. As in the earlier case, the tetragonal shear modulus $c' = C'/2$ is the most critical of the moduli for crystal stability. The bulk moduli are slightly greater in the Si containing compounds as compared to the Al- containing compounds. The Young’s and rigidity moduli fall in the ranges (237.2–287.9) GPa and (93.4–112.3) GPa, respectively. Our calculated values of the bulk moduli and elastic constants for $Co_2TSi$

(except $T = Sc$) agree well with those reported by Chen et al. Only the value of $c_{11}$ of $Co_2VSi$ exhibits a large deviation of 20%, whereas all others deviate by less than 7%.

As observed above for the Mn-containing compounds, all $Co_2TM$ compounds are between brittle and ductile from Frantsevich’s rule based on Poisson’s ratio ($\nu_{cr} \leq 0.33$) and Pugh’s criterion ($k_{cr} \leq 1.75$). The $Co_2$-based compounds synthesized in our laboratories turned out to be mostly brittle in accordance with the prediction of our calculations. Further discussion on the elastic properties will be presented using the Blackman’s diagram that is shown in Figure 9 for the $Co_2TM$ Heusler compounds. The values of $F_{12}$ fall in a very narrow range about the Cauchy line. All Si-containing compounds exhibit a positive Cauchy pressure, whereas $p_C < 0$ for the Al compounds with Sc, Mn, or Fe. It is worthwhile to note that the Al-containing $Co_2$ compounds tend to antistructural disorder, that is, they exhibit a $B2$ type rather than a $L2_1$ type crystalline structure. It is interesting to note that the hypothetical compound $Co_2ScAl$ is assumed to be nearly isotropic and very similar to an ideal Cauchy solid. Its universal anisotropy of only 1% is remarkable. Even though the values of $c_{44}/c_{11}$, $k$, and $\nu$ still deviate from the ideal Cauchy values by about 10%, out of all the compounds investigated here, it is closest to a
TABLE VII. Elastic properties of Co₂TM (T = Sc to Fe and M = Al, Si).
The experimental (a_{exp}) and optimized (a_{opt}) lattice parameters are given in Å. Elastic moduli B, G, E, hardness parameter H, elastic constants c_{ij}, C', and Cauchy pressure p_c are given in GPa. k, ν, ζ, and A_c are dimensionless quantities.

| M  | T | Sc  | Ti  | V   | Cr  | Mn  | Fe  | Sc  | Ti  | V   | Cr  | Mn  | Fe  |
|----|---|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
|    |   | a_{exp} |   | a_{opt} |   |   |   |   |   |   |   |   |   |
|    |   | 5.848 | 5.780 | 5.726 | 5.755 | 5.730 | 5.870 | 5.758 | 5.679 | 5.638 | 5.643 | 5.630 |
| T  |   | 5.780 | 5.758 | 5.711 | 5.700 | 5.702 | 5.870 | 5.758 | 5.679 | 5.638 | 5.643 | 5.630 |
| c_{11} | 277.8 | 286.0 | 290.0 | 265.0 | 267.3 | 268.2 | 267.4 | 295.4 | 297.4 | 287.5 | 310.5 | 273.6 |
| c_{12} | 83.8 | 129.7 | 150.5 | 169.2 | 155.3 | 150.1 | 123.5 | 158.3 | 183.5 | 194.2 | 174.2 | 168.5 |
| c_{44} | 105.0 | 126.7 | 140.7 | 156.1 | 160.4 | 150.5 | 113.1 | 134.8 | 148.7 | 162.4 | 156.9 | 144.7 |
| C'   | 194.0 | 156.3 | 139.5 | 95.8  | 112.0 | 118.1 | 143.9 | 137.1 | 113.9 | 103.3 | 136.3 | 105.1 |
| p_c  | -21.2 | 3.0   | 9.8   | 13.1  | -5.1  | -0.4  | 12.2  | 23.5  | 34.8  | 31.8  | 17.3  | 23.8  |
| B    | 148.4 | 181.8 | 197.0 | 201.5 | 192.6 | 189.5 | 171.5 | 204.0 | 221.5 | 225.3 | 219.6 | 203.5 |
| G    | 101.7 | 104.4 | 106.2 | 97.2  | 105.3 | 103.5 | 93.4  | 102.8 | 101.2 | 98.8  | 112.3 | 96.4  |
| E    | 248.4 | 262.9 | 270.0 | 251.3 | 267.2 | 262.6 | 237.2 | 264.0 | 263.5 | 258.6 | 287.9 | 249.8 |
| H    | 18.9  | 16.8  | 16.2  | 13.5  | 16.2  | 15.9  | 14.4  | 14.8  | 13.4  | 12.6  | 16.4  | 13.1  |
| k    | 1.46  | 1.74  | 1.86  | 2.07  | 1.83  | 1.83  | 1.84  | 1.99  | 2.19  | 2.28  | 1.96  | 2.11  |
| ν    | 0.22  | 0.26  | 0.27  | 0.29  | 0.27  | 0.27  | 0.27  | 0.28  | 0.28  | 0.30  | 0.31  | 0.28  |
| ζ    | 0.499 | 0.585 | 0.641 | 0.738 | 0.692 | 0.675 | 0.593 | 0.655 | 0.721 | 0.767 | 0.676 | 0.720 |
| A_c  | 1.08  | 1.62  | 2.02  | 3.28  | 2.86  | 2.55  | 1.55  | 1.97  | 2.61  | 3.48  | 2.30  | 2.76  |
| A_{21} | 0.01  | 0.29  | 0.62  | 1.88  | 1.46  | 1.13  | 0.23  | 0.57  | 1.19  | 1.75  | 0.88  | 1.34  |

Cauchy solid.

The elastic Zener (universal) anisotropy ranges from 1.08 (0.01) for Co₂ScAl to 3.48 (1.88) for Co₂CrSi (Co₂CrAl). Comparing the elastic anisotropy of compounds that are well known to crystallize in an ordered L₂₁ structure and those that are known to tend to disorder or where no successful synthesis is reported up to now, the Zener ratios for the most stable compounds are in the range 1.62 < A_c < 2.86.

The anisotropy of Young’s moduli and the rigidity moduli of Co₂ScAl, Co₂CrAl, Co₂ScSi and Co₂CrSi are displayed in Figures 10 and 11 that show the three dimensional distribution E(̂r) and G(̂r) as was also plotted above for the Mn containing compounds. The more pronounced anisotropy of the Cr containing compound is clearly visible. The differences in the anisotropy of the moduli between the two compounds are clearly visible, as observed above.

Comparing the distribution of the moduli, the Young’s modulus is largest in the ⟨111⟩-type directions, whereas the rigidity modulus is largest in the ⟨100⟩-type directions for most compounds, which are listed in Table VII. The only exception is Co₂ScAl, where A_c is close to unity and thus the distributions are nearly spherical, which shows a tendency to distortion in the ⟨100⟩ direction.

IV. DERIVED PROPERTIES

This section summarizes the physical properties of the compounds that are derived from the calculated elastic constants. Table VIII summarizes the results for the Co₂-based, Mn containing Heusler compounds with variation of the main group element Co₂MnM (M = main group element). Properties of the Co₂-Based Heusler compounds Co₂TM with varying 3d transition metals T, and Al and Si as the main group elements T are summarized in Table IX.

The Debye temperature Θ_D and Grünisen parameters ζ_ac are estimated in the acoustical approximation from the mode averaged sound velocities ̂v. These quantities depend, in addition to the elastic constants, on the mass density of the materials. The other two properties, melting temperature and hardness are exclusively based on the elastic constants. The melting temperature T_m is roughly estimated from c_{11}, and the hardness H_C results from Pugh’s and rigidity moduli. The underlying ideas and equations are given in the Appendix. Table VIII and Table IX summarize the properties derived for various Co₂TM Heusler compounds. In addition, the density ρ and the molecular mass m are given for completeness. Interestingly, the remaining physical properties do not appear to depend much on the composition. However, some clear trends are recognized on closer inspection.

Table VIII reveals, for the Mn containing compounds, the trend that the sound velocities calculated from the elastic constants and the Debye temperature in the acous-
TABLE VIII. Physical properties of Co$_2$Mn$M$ ($M$ = main group element). The Debye temperature $\Theta_D^{ac}$ is estimated from the sound velocity $v$.

| $M$ | Al | Ga | In | Si | Ge | Sn | Pb | Sb | Bi |
|-----|----|----|----|----|----|----|----|----|----|
| $\rho$ kg/m$^3$ | 7166 | 8617 | 8959 | 7424 | 8665 | 9023 | 11110 | 8972 | 10726 |
| $m$ g/mol | 199.8 | 242.5 | 287.6 | 200.9 | 245.4 | 291.5 | 380.0 | 294.6 | 381.8 |
| $v_l$ m/s | 6817 | 5977 | 5305 | 7054 | 6135 | 5604 | 4668 | 5516 | 4478 |
| $\Theta_D^{ac}$ K | 561 | 465 | 406 | 576 | 487 | 429 | 343 | 413 | 312 |
| $\zeta^{ac}$ | 1.59 | 1.80 | 1.70 | 1.66 | 1.71 | 1.69 | 1.77 | 1.75 | 1.91 |
| $T_m$ K | 2133 | 1984 | 1702 | 2388 | 2165 | 1934 | 1736 | 1944 | 1688 |
| $H_V^C$ GPa | 12.0 | 7.7 | 8.0 | 11.4 | 9.5 | 8.9 | 6.7 | 7.7 | 4.4 |

FIG. 9. (Online color) Blackman diagram for Co$_2T$M Heusler compounds containing a 3$d$ transition metal $T$ and Al or Si as the main group element $M$.

tical approach decrease with $Z$ of the main group element, whereas the Gr"uneisen parameters are all nearly the same for the different compounds. The average values of $\zeta^{ac}$ is about 1.7. The acoustical Debye temperatures are in the range of 312 K–576 K with average values of about (440 ± 70) K. The range of validity for the melting temperature $T_m$ is ±300 K for the approximation used here, and thus is on the same order as that of the spread of the calculated values. This leads to the estimate that the melting temperatures of the compounds are about (1960 ± 200) K. The Vickers's hardness has average values of the order of (8.5 ± 1.8) GPa.

From Table IX, it is found that the sound velocities calculated from the elastic constants exhibit only small changes among the different transition metals. As a direct consequence, the values for the Debye temperature or Gr"uneisen parameters in the acoustical approach are
values of (565 ± 1.1) K for the Si containing compounds. The only two reported experimental values of hardness for Co$_2$MnGe and Co$_2$MnSi are known to be 7.3 and 7.9, respectively. Although the experimental values are smaller than the predicted values, the tendency is the same. This is expected from the approximate nature of the model used.

Figure [12] compares the slowness surfaces of Co$_2$ScAl and Co$_2$MnSi. The latter was considered, since it is known from experiments to be very stable and to exhibit very low disorder. Further, it is the Heusler material with highest tunneling magneto resistance (TMR) ratios. The isotropic elastic behavior of Co$_2$ScAl is reflected in the nearly spherical distributions describing its three slowness surfaces. The shear modes are nearly degenerate and exhibit kiss singularities in the high symmetry directions. The behavior of the slowness surfaces of Co$_2$MnSi is typical for most of the investigated Co$_2$-based Heusler compounds, and its shape is similar to that of Cu$_2$MnAl. Its lower asymmetry as compared to Cu$_2$MnAl results in less pronounced differences between minima and maxima of the slowness.

To use slowness as a parameter for determination of the elastic constants, measurements along different high symmetry directions may be used. For the pressure wave, it may be found, for example, the following slowness eigenvalues: $s_0^{001} = \sqrt{\rho/c_{11}}$, $s_1^{110} = \sqrt{\rho/c_{44}}$, and $s_2^{111} = \sqrt{3\rho/(c_{11} + 4c_{44} + 2c_{12})}$ (superscript indices indicate the high symmetry direction [hkl], subscript index indicates the direction of polarization). The shear waves are degenerate in the [001] and [111] directions with values $s_{1,2}^{011} = \sqrt{\rho/c_{44}}$, and $s_{1,2}^{111} = \sqrt{3\rho/(c_{11} + 4c_{44} + 2c_{12})}$, respectively. The two non degenerate values for the [110] direction are $s_{10}^{110} = \sqrt{2\rho/(c_{11} - c_{12})}$ and $s_{20}^{110} = \sqrt{2\rho/(c_{11} + 2c_{44} + c_{12})}$.

V. SUMMARY

The elastic and accompanying physical properties of Heusler compounds, Cu$_2$MnAl as well as the Co$_2$YZ family, have been calculated. The present results for Cu$_2$MnAl are in good agreement with experiments. The directions of the largest Young’s modulus indicates the high-fracture energy directions. According to the Bader’s QTAIM analysis, the bonding in Cu$_2$MnAl is clearly metallic. However, Cu$_2$MnAl has one of the highest anisotropies as compared to the Co$_2$-based compounds. The Debye temperature, where only acoustic vibrational modes contribute, is about 397 K, which is lower than that of most of the studied compounds based on Co$_2$.

Based on the calculation of their elastic properties, the crystalline stability of Co$_2$-based cubic Heusler materials was assessed. The elastic constants of all the studied compounds follow the general inequality $B > c_{44} > G > c’ > 0$ such that the rigidity modulus $G$ is the main
TABLE IX. Physical properties of Co₂TM (T = Sc, Ti, V, Cr, Mn, Fe; M = Al, Si). The Debye temperature Θ_D^{ac} is estimated from the sound velocity v.

| M  | Al | Si |
|----|----|----|
| ρ  kg/m³ | 5919 | 6436 | 6814 | 7019 | 7166 | 7189 | 6269 | 6742 | 7142 | 7336 | 7424 | 7511 |
| m  g/mol | 189.8 | 192.7 | 195.8 | 196.8 | 199.8 | 200.7 | 190.9 | 193.8 | 197.9 | 200.9 | 201.8 |
| v_l m/s | 6928 | 7062 | 7049 | 6868 | 6817 | 6748 | 6872 | 7112 | 7064 | 6978 | 7054 | 6649 |
| v_t m/s | 4145 | 4027 | 3947 | 3722 | 3833 | 3794 | 3860 | 3904 | 3764 | 3670 | 3890 | 3583 |
| v m/s | 4587 | 4476 | 4394 | 4153 | 4265 | 4221 | 4295 | 4352 | 4206 | 4104 | 4335 | 4000 |
| Θ_D^{ac} K | 576 | 575 | 572 | 546 | 561 | 555 | 549 | 567 | 556 | 546 | 576 | 533 |
| ζ^{ac} | 1.37 | 1.54 | 1.67 | 1.73 | 1.59 | 1.60 | 1.60 | 1.68 | 1.78 | 1.83 | 1.66 | 1.74 |
| T_m K | 2195 | 2244 | 2267 | 2119 | 2133 | 2138 | 2133 | 2299 | 2311 | 2252 | 2388 | 2170 |
| H^C_V GPa | 16.2 | 12.9 | 11.9 | 9.4 | 12.0 | 11.9 | 11.0 | 10.5 | 8.9 | 8.2 | 11.4 | 9.1 |

FIG. 12. (Online color) Calculated slowness surfaces of Co₂ScAl (a)…(c) and Co₂MnSi (d)…(f). The slowness is given in (km/s)^{-1}.

constraint on stability. The results of our calculations demonstrate that all the studied compounds are close to the borderline between brittle and ductile. From the elastic point of view, they mainly exhibit bonding behavior between those of covalent and metallic. For most of the studied stable compounds, the universal anisotropy index is in the range 0.57 < A_U < 2.73. All the studied stable compounds are most stiff in the ⟨111⟩-type directions. The detailed analysis of all the compounds revealed that Pugh’s criterion for the ductile–brittle transition (k_{Pugh} ≈ 1.75) should be replaced by Christensen’s criterion (k_{cr} ≈ 2.27 or k_{cr}^{-1} ≈ 0.44).

For Co₂-based compounds, when the nuclear charge Z of the main group element increases, the lattice parame-
ters also increase but the values of the hardness parameter decrease. Here, the Zener ratio \( (A_e) \) and the universal anisotropy index \( (A_T) \) show the same tendency. It should be mentioned however, that the anisotropy does only judge on the structural stability but not on the chemical stability, as is seen especially for the case of Co\(_2\)MnSb, that does not exist as a pure compound. The Debye temperature in the acoustical approach decreases with \( Z \), but there is no evidence for a distinguishable dependence on the \( T \) element for the Co\(_2\)TM compounds. The Grüneisen parameters in the acoustical approach are all nearly the same for the different compounds. The hardness shows the same tendency. Finally, it is found that Co\(_2\)ScAl is close to an ideal Cauchy solid and is predicted to be the most hard material in the investigated series.

The calculated material properties can be applied quite reliably to bulk materials. On the other hand, the prediction of stability could be exploited in any compounds in particular to those when the possibility of structural phase transition in crystals is investigated.

### Appendix A: Cubic elastic constants, elastic moduli, and elastic stability.

The basics of the elastic properties of solids are described in the book by Nye\(^{[23]}\). Here the focus is on the equations for cubic compounds, remarks on tetragonal and hexagonal compounds are found in Reference\(^{[2]}\). A lattice \( A \) can be transformed to a new deformed lattice \( A' \) by the strain matrix \( \epsilon \). The strain matrix \( \epsilon \) is symmetric and contains six different strains \( \epsilon_i \). By Hooke’s law, the elastic relation between strain \( (\epsilon) \) and stress \( (\sigma) \) is:

\[
\sigma = C \epsilon, \quad (A1)
\]

where \( C \) is the elastic stiffness matrices, and the relations between the compliance matrix \( (S) \) and the stiffness matrix is

\[
S = C^{-1}. \quad (A2)
\]

In the most general case, the elastic matrix is symmetric and on the order of \( 6 \times 6 \). In triclinic lattices, it contains 21 independent elastic constants. This number is largely reduced in highly symmetric lattices. In cubic lattices, only the three elastic constants \( c_{11}, c_{12}, \) and \( c_{44} \) are independent. The elastic matrix for all classes of cubic crystals has the form (zero elements are denoted by dots):

\[
C^{\text{cubic}} = \begin{pmatrix}
  c_{11} & c_{12} & c_{12} & \cdot & \cdot & \cdot \\
  c_{12} & c_{11} & c_{12} & \cdot & \cdot & \cdot \\
  c_{12} & c_{12} & c_{11} & \cdot & \cdot & \cdot \\
  \cdot & \cdot & c_{44} & \cdot & \cdot & \cdot \\
  \cdot & \cdot & \cdot & c_{44} & \cdot & \cdot \\
  \cdot & \cdot & \cdot & \cdot & c_{44} & \cdot \\
\end{pmatrix} \quad (A3)
\]

The matrix has 6 eigenvalues out of which only three are different. These three different values of the eigenvalues are:

- \( C_1^\ell = 2c_{12} + c_{11} \) (nondegenerate),
- \( C_{2,3}^\ell = c_{11} - c_{12} \) (twofold), and
- \( C_{4,\ldots,6}^\ell = c_{44} \) (threelfold degenerate).

They correspond to the bulk, the tetragonal shear, and the shear moduli as will be shown below. The crystal becomes unstable when one of the eigenvalues becomes zero or negative.

For an isotropic system, the elastic matrix \( C^{\text{iso}} \) contains only the two constants \( c_{11} \) and \( c_{12} \), whereas the remaining diagonal elements of the matrix are determined by \( c_{44} = (c_{11} - c_{12})/2 \).

In cubic systems, the relations between the elastic constants \( c_{ij} \) and the elements of the compliance matrix \( s_{ij} \) are given by,

\[
s_{11} = \frac{c_{11} + c_{12}}{c},
\]

\[
s_{12} = -\frac{c_{12}}{c},
\]

\[
c = (c_{11} - c_{12})(c_{11} + 2c_{12}),
\]

\[
s_{44} = \frac{1}{c_{44}}.
\]

The bulk modulus is defined by the elastic constants. For cubic materials it is given by,

\[
B = \frac{1}{3}(c_{11} + 2c_{12}). \quad (A5)
\]

Born and co-workers developed the theory of stability of crystal lattices\(^{[27]}\). The Born–Huang\(^{[25]}\) elastic stability criteria for a cubic crystal at ambient conditions\(^{[29]}\) are given by,

- \( c_{11} + 2c_{12} > 0 \)
- \( c_{44} > 0 \)
- \( c_{11} - c_{12} > 0 \)

that is, the bulk, \( c_{44} \), shear, and tetragonal shear moduli have to be all positive. The criteria are referred to as spinodal, Born’s shear\(^{[25]}\) and Born criteria, respectively. The first criterion defines the spinodal pressure,
whereas the last criterion is often used to define an additional elastic constant,

\[ C' = c_{11} - c_{12}. \]  

This constant is also called tetragonal shear modulus. In some studies, \( c' = \frac{(c_{11} - c_{12})}{2} \) is used instead, because the tetragonal instability is observed when the hydrostatic pressure becomes \( 2p > C' \), that is \( p > c' \). In detail, \( G_{110} = c' \) is the single-crystal shear modulus for the (110) plane along the [110] direction. The single-crystal shear modulus for the (100) plane along the [010] direction is \( G_{100} = c_{44} \). It is related to a tetragonal deformation and large values denote high stability of the crystal with respect to tetragonal shear.

The Cauchy pressure for cubic crystals is defined using the Cauchy relation as,

\[ p_C = c_{12} - c_{44}. \]  

For single cubic crystals, the shear modulus \( G \), Pugh’s ratio \( k = B/G \), and the Poisson’s ratio \( \nu \) are calculated from the elastic constants using the following relations:

\[ G = \frac{3c_{44} + c_{11} - c_{12}}{5}, \]  

\[ k = \frac{5}{3} \frac{(c_{11} + 2c_{12})}{(3c_{44} + c_{11} - c_{12})}, \]  

\[ \nu = \frac{c_{12}}{c_{11} + c_{12}}. \]  

The first and third Born criteria restrict the range of Poisson’s ratio to \(-1 \leq \nu \leq 1/2\).

Polycrystalline materials consist of randomly oriented crystals and thus a description of their elastic properties requires only two independent elastic moduli: the bulk modulus \( B \), and the shear modulus \( G \). The relationships between the single-crystal elastic constants and the polycrystalline elastic moduli are given by the Voigt\(^{[23]} \) or Reuß\(^{[24]} \) averages. Voigt’s approach uses the elastic stiffnesses \( c_{ij} \), whereas Reuß’s approach uses the compliances \( s_{ij} \). The bulk moduli in Voigt’s \( (B_V) \) and Reuß’s \( (B_R) \) approach are equal for cubic crystals and given by:

\[ B = \frac{1}{3} \frac{(c_{11} + 2c_{12})}{3(s_{11} + 2s_{12})}, \]  

\[ = \frac{B_V}{B_R}. \]  

The isotropic shear or rigidity modulus \( G = \frac{[G_V + G_R]}{2} \) is defined by Voigt’s \( G_V \) and Reuß’s \( G_R \) shear moduli, where,

\[ G_V = \frac{1}{5} (c_{11} - c_{12} + 3c_{44}) \]  

\[ G_R = \frac{5}{4(s_{11} - s_{12} + 3s_{44})} \]  

\[ = \frac{5}{3(c_{11} - c_{12}) + 4c_{44}}. \]  

Accordingly, Poisson’s ratio \( \nu \) and Young’s modulus \( E \) of polycrystalline cubic materials are calculated from the equations using the averaged bulk and rigidity moduli as,

\[ \nu = \frac{1}{2} \frac{3B - 2G}{3B + G}, \]  

\[ E = 2G(1 + \nu). \]  

In cubic crystals, the bulk modulus is isotropic. However, rigidity and Young’s moduli not isotropic. The directional dependence of Young’s modulus \( E(\hat{r}) \) is defined by the ratio of longitudinal stress to strain. For cubic systems, the three dimensional distribution is given by,

\[ E^{-1}(\hat{r}) = s_{11} - 2s_{12}F_{lmn}, \]  

where \( s = s_{11} - s_{12} - s_{44}/2 \), and \( F_{lmn} = (\hat{x}^2\hat{y}^2 + \hat{y}^2\hat{z}^2 + \hat{z}^2\hat{x}^2) \) is the orientation function of a cubic single crystal specimen given in terms of the direction cosines \( (l := \hat{x}, \text{ etc.}) \). It is obvious that \( E(\hat{r}) \) becomes isotropic for \( s = 0 \). Hence, Zener ratio or the elastic anisotropy is defined for cubic crystals as,

\[ A_c = \frac{2(s_{11} - s_{12})}{s_{44}} = \frac{2c_{44}}{c_{11} - c_{12}}. \]  

The cubic elastic anisotropy may be used as another important physical quantity for the description of structural stability. Materials exhibiting large \( A_c \) ratios occasionally show a tendency to deviate from the cubic structure. Materials with negative Zener ratio \( (A_c < 0) \) violate at least one of Born’s criteria and are mechanically instable.

Ranganathan and Ostoja-Starzewski\(^{[22]} \) summarized the existing anisotropy theories and developed a so-called universal anisotropy index \( A_U \) that is calculated for cubic crystals, using the condition \( B_V = B_R \) by the simplified equation,

\[ A_U = 5 \frac{G_V}{G_R} + \frac{B_V}{B_R} - 6 \]  

\[ A_U^{cubic} = 5 \left( \frac{G_V}{G_R}, 1 \right). \]  

Similar to the case of the Young’s modulus, the directional dependence of the rigidity modulus \( G(\hat{r}) \) is defined by\(^{[23]} \).
where \( \chi_{lmn} = \hat{a}^2 \hat{b}^2 \hat{c}^2 \). The last term in Eq. \((A16)\) is the so-called bending–torsion correction (or difference) if \( G_0 \) is defined as the “true” rigidity modulus. \( G_0 \) becomes isotropic for \( A_e = 1 \). The bending–torsion correction vanishes for the highly symmetric Cauchy solid with cubic symmetry is \( c_{12} = c_{44} \). The conditions required to satisfy this Cauchy relation are:

- Only central forces take part in the interaction between the atoms.
- Only harmonic forces exist between the atoms. Anharmonicity will destroy the Cauchy relations.
- The atoms are located at the centers of symmetry.
- Thermal effects and initial stress are neglected.

From the isotropy \((c_{12} = c_{11} - 2c_{44})\) and Cauchy \((c_{12} = c_{44})\) relations, only one independent elastic constant \((c_{11} = 3c_{12} = 3c_{44})\) would remain for cubic crystals. This has the result that Pugh’s ratio of a cubic, isotropic solid following Cauchy’s relation becomes \( k_{\text{Cauchy}} = 5/3 = 1.67 \approx 1.7 \). At the same time, Poisson’s ratio simplifies to \( \nu_{\text{Cauchy}} = 1/4 \). The elastic matrix of such an ideal Cauchy solid has the form (zero elements are denoted by dots),

\[
C_{\text{Cauchy}}^{\text{cubic}} = \frac{1}{3} \begin{pmatrix}
  3c_{11} & c_{11} & c_{11} & \cdots & \cdots \\
  c_{11} & 3c_{11} & c_{11} & \cdots & \cdots \\
  c_{11} & c_{11} & 3c_{11} & \cdots & \cdots \\
  \cdots & \cdots & \cdots & c_{11} & \cdots \\
  \cdots & \cdots & \cdots & \cdots & c_{11} \\
\end{pmatrix}
\]  \( (A17) \)

The three different eigenvalues of \( C_{\text{Cauchy}}^{\text{cubic}} \) are \( 5c_{11}, 2c_{11}, \) and \( c_{11} \), which are nondegenerate, twofold degenerate, and threefold degenerate, respectively.

Apart from the elastic moduli, a few more important physical quantities can be derived from the elastic constants. The volume \( (\kappa) \) and linear \( (\beta) \) compressibilities of cubic crystals are isotropic and given by,

\[
\beta = s_{11} + 2s_{12}, \quad (A18)
\]

\[
\kappa = 3\beta = B^{-1}.
\]

**Appendix B: Derived physical properties from cubic elastic constants.**

In the bond-orbital model, Kleinman’s internal displacement parameter is defined by \( \Theta_{\text{ca}} \)

\[
\zeta = \frac{c_{11} + 8c_{12}}{7c_{11} + 2c_{12}}.
\]  \( (B1) \)

It describes the relative positions of atoms in different sublattices under volume conserving strain distortions for which the positions are not fixed by symmetry anymore. \( \zeta \) vanishes if no internal displacements appear. \( \zeta = 1 \) when the bond lengths are unchanged and \( \zeta = -1/2 \) when the bond angles are unchanged, both for linear strain.

In the quasi-harmonic approach, the Debye temperature \( \Theta_{\text{D}}^{\text{qha}} \) depends upon the volume of the crystal. For every volume \( V \), \( \Theta_{\text{D}}^{\text{qha}}(V) \) is rigorously defined in terms of the elastic constants through a spherical average of the three components of the sound velocity. The isotropic approximation, allows to evaluate \( \Theta_{\text{D}}^{\text{qha}} \) using the expression \( \Theta_{\text{D}}^{\text{Heusler}} \)

\[
\Theta_{\text{D}}^{\text{qha}} = \frac{\hbar}{k_B} \sqrt{n \cdot 6\pi^2 V \sqrt{\frac{B}{M} f_{\nu}}}, \quad (B2)
\]

\[
\Theta_{\text{D}}^{\text{Heusler}} = \frac{\hbar}{k_B} \sqrt{9\pi^2 V \sqrt{a \frac{B}{M} f_{\nu}}}, \quad (B2)
\]

where \( \hbar \) is Planck’s constant, \( k_B \) is Boltzmann’s constant and \( n \) is the number of atoms in the primitive cell with volume \( V \) unit \( n = 4 \) in the case of Heusler compounds and the volume of the primitive cell or 16 for the cubic cell with lattice parameter \( a \). \( B \) is the adiabatic bulk modulus of the crystal and \( M \) the mass of the compound corresponding to \( V \). Finally, \( f_{\nu} \) is a function of the Poisson ratio \( \nu \)

\[
f_{\nu} = \frac{\frac{3}{2} \left( \frac{2 + \nu}{3 - 2\nu} \right)^{3/2} + \left[ \frac{2 + \nu}{3 - 2\nu} \right]^{3/2}}{\sqrt{2 + \nu}}.
\]  \( (B3) \)

Another important mechanical property is the hardness of a material. Other than for the elastic moduli, there is no straightforward theory to calculate the hardness directly from the elastic constants. However, several models were developed to relate the hardness of a material to the elastic moduli. Pugh related the Brinell hardness \( H_B \) of pure metals to their shear modulus \( G \) by \( H_B = G/b/c \), where \( b \) is the Burger’s vector of the dislocation and \( c \) is a constant for all metals of the same structure. Tote obtained the semi-empirical relation \( H_B^T \approx 0.151G \) between Vicker’s hardness \( H_V \) and the rigidity modulus \( G \). Recently, Chen et al gave a semi-empirical relation between Vicker’s hardness and the product of the squared Pugh’s modulus \( (k = B/G) \) ratio and the shear modulus \( G \) as,
\[ H_V^C = 2(k^{-2} G)^{0.585} - 3. \]  
(B4)

Also for cubic metals only, Fine et al.\(^2\) obtained an approximate linear relationship between the melting temperature \( T_m \) and the elastic constant \( c_{11} \). The \( T_m \) of various cubic metals was \( \pm 300 \, \text{K} \) within a linear dependence when estimated from the following empirical equation:

\[ T_m^c = 553 \, \text{K} + 5.91 \, \frac{\text{K}}{\text{GPa}} \cdot c_{11}. \]  
(B5)

The elastic constants and moduli also allow estimation of the averaged sound velocity \( \bar{v} \),

\[ \bar{v} = \left[ \frac{3}{v_i^{-3} + 2v_i^{-3}} \right]^{1/3}. \]  
(B6)

From the longitudinal \( (v_l) \) and transverse \( (v_t) \) wave velocities of isotropic materials the Debye temperature can be estimated, where \( v_l \) and \( v_t \) are,

\[ v_l = \sqrt{\frac{3B + 4G}{3\rho}} \]  
(B7)

\[ v_t = \sqrt{\frac{G}{\rho}}, \]

where, \( \rho \) is the mass density of the material. From the average sound velocity at low temperatures, the Debye temperature can be estimated by using the relation\(^3\)

\[ \Theta^\text{ac}_D = \frac{\hbar}{k_B} \sqrt{\frac{n \, N_A \rho}{4\pi M}} = \frac{\hbar}{k_B} \sqrt{\frac{n \, 1}{4\pi V}}, \]  
(B8)

where \( N_A \) is Avogadro’s number. Other parameters are the same as in the case of the Debye temperature calculation in the quasi-harmonic approach.

In solids, the Grüneisen parameter \( \zeta^\text{ac} \) is also related to the sound velocities. Belomestnykh\(^4\) derived this Grüneisen parameter using,

\[ \zeta^\text{ac} = \frac{3 \left( 3v_l^{-2} - 4v_t^{-2} \right)}{2 \left( v_l^{-2} + 2v_t^{-2} \right)}. \]  
(B9)

The above described acoustical properties concern averages and may be used for polycrystalline materials. Acoustical spectroscopy is used, indeed, also for investigation of the single crystal elastic constants. The directional dependence of the phase velocity \( v \) is found from Christoffel’s equation:

\[ (\Gamma_{ij} - \rho \varepsilon^2 \delta_{ij}) U_j = 0, \]  
(B10)

where \( \Gamma_{ij} = c_{ijkl} l_i l_j \) is the Christoffel tensor built from the elastic constants and the direction cosines \( l_i \) (\( i = 1 \ldots 3 \)). \( \rho \) is the density, \( \delta_{ij} \) is the Kronecker symbol and \( U \) is the polarization vector.

In cubic systems, only 3 elastic constants are independent and the components of the Christoffel tensor are reduced to,

\[ \Gamma_{ij} = \begin{cases} \left( c_{111} + c_{444} l_1 l_1 \right) i \neq j \\ c_{111} l_1^2 + c_{444} (1 - l_2^2) \ i = j \end{cases} \]  
(B11)

with \( l_1 = \sin(\theta) \cos(\phi), \) \( l_2 = \sin(\theta) \sin(\phi), \) and \( l_3 = \cos(\theta) \) in polar co-ordinates. In greater detail, the elements of the Christoffel tensor are (note that: \( \Gamma_{ij} = \Gamma_{ji} \)),

\[ \begin{align*}
\Gamma_{11} &= c_{111} l_1^2 + c_{444} l_1^2 + c_{444} l_2^2 \\
\Gamma_{22} &= c_{444} l_2^2 + c_{111} l_2^2 + c_{444} l_3^2 \\
\Gamma_{33} &= c_{444} l_3^2 + c_{444} l_2^2 + c_{111} l_3^2 \\
\Gamma_{12} &= (c_{111} + c_{444}) l_1 l_2 \\
\Gamma_{13} &= (c_{111} + c_{444}) l_1 l_3 \\
\Gamma_{23} &= (c_{111} + c_{444}) l_2 l_3
\end{align*} \]  
(B12)

In case of an ideal Cauchy solid the Christoffel tensor is further reduced to,

\[ \Gamma = \frac{c_{111}}{3} \begin{pmatrix}
2l_1^2 + 1 & 2l_1 l_2 & 2l_1 l_3 \\
2l_1 l_2 & 2l_2^2 + 1 & 2l_2 l_3 \\
2l_1 l_3 & 2l_2 l_3 & 2l_3^2 + 1
\end{pmatrix}. \]  
(B13)

The results for ideal Cauchy solids are three eigenvalues: \( \rho s_1^2 = c_{111} \) for the compression wave, and \( \rho s_2^2 \) for the twofold degenerate shear wave. Both the modes, shear \( s \) and pressure \( p \), are independent of the propagation direction and their slowness surfaces appear spherical with \( s_p = \sqrt{\rho/c_{111}} \) and \( s_s = \sqrt{3\rho/c_{111}} \).

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