Classical turning surfaces in solids:  
When do they occur, and what do they mean? 

Aaron D. Kaplan
Department of Physics, Temple University, Philadelphia, PA 19122

Stewart J. Clark
Centre for Materials Physics, Durham University, Durham, DH1 3LE, United Kingdom

Kieron Burke
Departments of Chemistry and Physics, University of California, Irvine, CA 92697

John P. Perdew
Departments of Physics and Chemistry, Temple University, Philadelphia, PA 19122  
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Classical turning surfaces of Kohn-Sham potentials, separating classically-allowed regions (CARs) from classically-forbidden regions (CFRs), provide a useful and rigorous approach to understanding many chemical properties of molecules. Here we calculate such surfaces for several paradigmatic solids. Our study of perfect crystals at equilibrium geometries suggests that CFRs are absent in metals, rare in covalent semiconductors, but common in ionic and molecular crystals. A CFR can appear at a monovacancy in a metal. In all materials, CFRs appear or grow as the internuclear distances are uniformly expanded. Calculations with several approximate density functionals and codes confirm these behaviors. A classical picture of conduction suggests that CARs should be connected in metals, and disconnected in wide-gap insulators. This classical picture is confirmed in the limits of extreme uniform compression of the internuclear distances, where all materials become metals without CFRs, and extreme expansion, where all materials become insulators with disconnected and widely-separated CARs around the atoms.

I. INTRODUCTION

The most basic property of an ordered solid is whether or not it is metallic [1–3]. The Sommerfeld free electron model of metallic conduction [4], which involves quantum mechanics only via a Fermi distribution of velocities, assumes a homogeneous system (uniform electron gas), but we wish to understand the effect of inhomogeneity. A simple classical picture of conduction is to consider an electron of energy $\epsilon$ in a single-particle effective potential, $v_{\text{eff}}(r)$. If $\epsilon > v_{\text{eff}}(r)$ everywhere, this classical electron will move forever throughout the solid (or at least as far as its mean free path will allow), and the solid should be a metal. On the other hand, if the only classically allowed regions are disjoint regions bound to atoms, the solid should be strongly insulating. Unlike a classical electron, a quantum electron can tunnel into a classically-forbidden region.

The standard modern theory of conduction (for ordered solids) is that of Bloch bands, with insulators having filled bands below finite gaps in the spectrum [5]. At first glance, this appears to have little in common with the simple classical picture given above. But quantum theories derive from classical theories, and are connected to quantum mechanics via semiclassical approximations using classical trajectories. Consider what happens to a standard band structure as $\hbar \to 0$ keeping the Fermi energy fixed. For energies above the maximum of the potential everywhere, the bands become more free-electron like, as the inhomogeneity in the potential becomes less relevant. On the other hand, for energies below the maximum, the band becomes narrower and more localized as $\hbar$ shrinks. The importance of turning points to semiclassical (and density functional) approximations was prefigured in the cartoon of Fig. 1 of Ref. [6].

The Kohn-Sham (KS) potential [7] is the scalar potential that, acting on non-interacting electrons, yields a ground-state electron density equal to that of the real system. While not a physical observable, the KS potential is extremely useful as an interpretive tool. Inspired by earlier work that used the “potential acting on an electron in a molecule” (PAEM) [8, 9], Ospadov et al. [10] recently created a “periodic table of nonrelativistic classical turning radii” using the KS turning surface of the highest occupied KS orbital, defined as those points satisfying

$$v_s(r) = \epsilon_{\text{HO}}, \quad (1)$$

where $v_s(r)$ is the KS potential, and $\epsilon_{\text{HO}}$ is the energy of the highest occupied orbital (the Fermi energy $\epsilon_F$ in a metal). They demonstrated that a classical turning surface could characterize bond types in molecules numerically and visually [10]. At equilibrium geometries, covalent bonds as in $N_2$ have fused (roughly ellipsoidal) turning surfaces, ionic bonds as in NaCl often have seamed surfaces, hydrogen bonds as in $(H_2O)_2$ have necked sur-
faces, and van der Waals bonds as in Ne$_2$ have bifurcated surfaces (with each part nearly spherical). The ratio of an equilibrium bond length to the sum of its atomic radii is roughly 0.5 for a covalent bond, 1.0 for an ionic or hydrogen bond, and 1.5 for a van der Waals bond. More recently, Gould et al. [11] found that the classical turning surface of H$_2^+$, which is approximately ellipsoidal at the equilibrium bond length, bifurcates when the bond length is stretched to about twice the turning radius of one dissociation product H$_2^+$ (rigorously the same as the turning radius of a neutral hydrogen atom, ≈ 1.06 Å). Neutral atoms other than hydrogen typically have one or more electrons in the classically-forbidden region (CFR) outside their turning surfaces [12]. Earlier, Ref [14] had noted that a CFR emerges within the local density approximation (LDA) in stretched H$_2$ very near the Coulson-Fisher point, signaling the onset of strong correlation as the bond grows.

A turning surface in position space should not be confused with a Fermi surface in wavevector space. The turning surface defined here is the intersection in position-space of the KS potential with the Fermi level. If $\varepsilon_F > v_{\mu}(r)$ everywhere, there is no turning surface, whereas the Fermi surface is always well-defined. One could also define a turning surface in terms of the chemical potential $\mu \geq \varepsilon_F$ [14], which differs from $\varepsilon_{\text{HO}}$ for non-metals, but using $\varepsilon_{\text{HO}}$ in Eq. 1 is more practical and useful.

Here we present calculations of KS turning surfaces for a variety of simple solids. Our calculations are at the LDA and generalized gradient approximation (GGA) level of exchange-correlation approximations, which usually yield close approximations to more precise KS potentials in molecules (as both KS potential and $\varepsilon_{\text{HO}}$ are typically too shallow by about the same amount). In Kohn-Sham density functional theory (KS DFT) [7], the KS potential $v_{\mu}(r)$ is a multiplicative operator. In generalized KS theory, the exchange-correlation potential of a meta-GGA or a hybrid functional (using the Hartree-Fock exchange energy) is a non-multiplicative operator, but can be replaced [15] by the local one needed to define a classical turning surface. One can apply all the concepts of Ref. [10] to analyze bonding in solids from a chemical viewpoint, but here we focus on the most elementary property of materials: are they metallic? In our classical conduction argument above, the effective potential is clearly the KS potential, and the most energetic electron is the highest-occupied level. If $\varepsilon_{\text{HO}}$ is higher than the maximum value of the KS potential, there are no classical Fermi-energy turning surfaces and the system ought to be metallic. If not, and if $\varepsilon_{\text{HO}}$ is so low that the classically-allowed regions are disconnected, the system ought to be insulating (with a wide gap). We expect semiconductors to lie somewhere in between these extremes.

This work discusses classical turning surface analogs and semiclassical interpretations of them for a variety of simple solids. Section II describes the computational tools used to extract and analyze the KS potential for metals, as presented in Section III and band insulators presented in Section IV. Special attention is paid to the roles of strain and defects in forming CFRs within solids. Section V discusses how the CFR can be used to predict conduction properties. Section VI discusses the role of CFR connectedness in determining the conductive properties of solids. The Supplemental Materials section contains additional data.

To interpret our results correctly, we point out the following crucial points concerning gaps. It has long been known that the KS gap, i.e., the bandgap of the exact KS potential, does not match the true (fundamental charge) gap [14], and typically underestimates it. The KS gaps of strictly semilocal approximations like LDA or GGAs are typically close to the exact KS gap [16,18], and thus are often substantially less than the fundamental gap. Hybrid functionals and meta-GGAs yield larger gaps when treated in a generalized KS scheme [19]. When lattice parameters are stretched well beyond equilibrium, semilocal functionals may produce broken-symmetry solutions of lower energy, as is well-known in the paradigmatic case of stretched H$_2$ [20], but does not occur (at least for finite systems) with the exact functional. As all calculations in this paper use only semilocal functionals, they are in the KS scheme, yield gaps that are smaller than fundamental gaps, and can break symmetry.

II. COMPUTATIONAL METHODS

All ensuing calculations were performed with either the Vienna ab initio Simulation Package (VASP) [21], or the Castep code [22,23], or both. All GGA calculations used the Perdew-Burke-Ernzerhof GGA [24], and all LSDA calculations used the Perdew-Zunger parameterization of the uniform electron gas correlation energy [25]. The calculations in VASP were performed with a cutoff energy of 800 eV, a $\Gamma$-centered mesh of spacing 0.08 Å, and stress convergence at $10^{-6}$ eV/Å. To determine equilibrium geometries in VASP, for metals, first-order Methfessel-Paxton smearing with parameter of 0.2 was used, and for insulators, the Blöchl tetrahedron method was used. VASP’s internal methods were used to determine the relaxed cell volume. In Castep, a density-mixing algorithm was used to reach self-consistency, and geometries were determined with a BFGS (Broyden-Fletcher-Goldfarb-Shanno) energy minimization scheme with the finite basis set corrected for stress [26]. After relaxation, a calculation at the equilibrium volume using the Blöchl tetrahedron method was performed to accurately determine the density of states. Accurate [27] PAW on-the-fly pseudopotentials were used throughout. Tables S6 to S54 (in the Supplemental Materials) present all raw data; machine readable data will be made available upon reasonable request.

For monolayers, a $45 \times 45 \times 1$ k-point grid was used in conjunction with the Blöchl tetrahedron method. All
other parameters remain the same from bulk calculations. The c direction was padded with 30 Å of vacuum region to reduce interactions between image monolayers.

In density functional plane-wave codes, the densities and potentials are stored on a uniform grid \( \mathbf{R} \), the dimensions of which are determined by the size of the unit cell and the plane-wave cutoff energy. Acceptable convergence of the total energy relies on suitable convergence of the potentials and densities on this grid. The values of \( v_s(\mathbf{R}) \) are obtained from this grid. In core regions, the true potential is much deeper than the pseudopotential, so these are classically allowed. Thus the PAW pseudopotential core regions were excluded from the CFR (frozen-core pseudopotentials were used in both VASP and Castep). The self-consistent electronic eigenstates give \( \varepsilon_{\text{HO}} \) (the Fermi energy \( \varepsilon_F \) in a metal), and the regions where \( \varepsilon_{\text{HO}} - v_s(\mathbf{R}) < 0 \) define the CFR. We assign equal volume to each point relative to the primitive cell, as the real-space mesh is uniform. Suppose there are \( N_{\text{prim}} \) total real-space mesh points in the cell, and let the volume of the primitive cell be \( V_{\text{prim}} \). Then the volume of any point is \( V_{\text{prim}}/N_{\text{prim}} \). If there are \( N_{\text{CFR}} \) points at which \( \varepsilon_{\text{HO}} - v_s(\mathbf{r}) < 0 \), the volume of the CFR is

\[
V_{\text{CFR}} = V_{\text{prim}} N_{\text{CFR}} / N_{\text{prim}}. \tag{2}
\]

The dimensionless, “fractional volume” of the CFR, which will be used in the ensuing figures and fits, is defined as

\[
v \equiv V_{\text{CFR}} / V_{\text{prim}} = N_{\text{CFR}} / N_{\text{prim}}, \tag{3}
\]

the number of real-space mesh points within the CFR relative to the total number of mesh points in the primitive cell.

As the fractional CFR volume \( v \to 0 \), our method requires ever finer real- and reciprocal-space meshes to resolve \( v \). This need is limited by the resolution determined by the plane-wave cutoff energy. Our data for \( v \ll 1 \) will necessarily be more noisy than for larger values of \( v \). Despite this, we show a posteriori that reasonable fits to \( v(V_{\text{prim}}) \) may be found.

Each code uses differently-generated pseudopotentials with different optimal basis set cutoff energies (and hence pseudopotential grid sizes, etc.), different energy minimization schemes, and different Brillouin zone integration methods. To ensure that our method is not dependent upon the numerical methods of a particular code, we have verified that the Castep and VASP results are consistent.

### III. OPENING CFRS IN METALS

As we see in Table 1, no CFR is present in certain defect-free metals (Al, Cu, and Pt) at their equilibrium geometries. This is in line with our initial hunch, but does not extend to metals with monovacancies. A Pt supercell with a monovacancy defect harbors a small CFR; plotting this CFR in Fig. 1, we see that the classically-forbidden region encapsulates the center of the vacancy perfectly. Relaxation of the supercell volume was performed two ways: direct minimization of the stress tensor, and keeping the supercell volume fixed to the bulk volume while allowing ion positions to change.

The vacancy defect formation energy can be recast as the energy needed to create a curved surface within a solid \[28\]. The localization of the CFR to the vacancy region is a clear manifestation of this. Carling et al. \[29\] found that the LDA is more accurate than GGAs for the Al monovacancy formation energy, in line with earlier results \[30\] for the jellium surface energy. They also found a very low electron density near the center of the vacancy, and large Friedel oscillations around it, consistent with a CFR near the center. Large voids and exterior surfaces would also give rise to extensive CFReRs in any material.

The definition of the monovacancy volume given in Carling et al. differs from ours. Their method used the liquid drop model of jellium from Ref. \[28\] to extract the vacancy’s volume from the vacancy defect formation energy. This method will generally yield larger volumes than the corresponding CFR volumes.

It is natural then to ask how far a metal needs to be stretched before a CFR emerges. In Fig. 2 we plot the fractional volumes of the LSDA and PBE CFReRs as a function of the primitive cell volume. To find accurate
critical primitive cell volumes $V_c$ for the emergence of a CFR, we perform a least squares fit to

$$v(V_{\text{prim}}, c) = \Theta(V_{\text{prim}} - V_c) \sum_{m=0}^{3} c_m \left( \frac{V_c}{V_{\text{prim}}} \right)^m$$

where $V_{\text{prim}}$ is the primitive cell volume, $0 \leq v < 1$ is the fractional CFR volume, $\Theta$ is a step function, and the dimensionless $c = (c_0, c_1, c_2, c_3)$ are derived from least-squares fit parameters. Note that $\sum_{m=0}^{3} c_m = 0$, and $\partial v / \partial V_{\text{prim}} > 0$ for $V_{\text{prim}} - V_c = 0^+$. Fit parameters for PBE and LSDA data, as calculated in VASP, are listed in Table I for the fit parameters of PBE data as calculated in Castep, refer to Table SI in the Supplemental Materials.

Despite the possibility of noisy data at small CFR fractions, $0 < v \ll 1$, no lower cutoff on the VASP data was needed in the fitting process. The Castep data required a cutoff of $v_c = 0.01$ (for which any data with $v \leq v_c$ was taken to have $v = 0$ instead), and a few elemental solids (Mg, Sr, Ba, Ra) required a higher cutoff, $v_c = 0.05$. The form of Eq. I is selected because it makes $v$ tend to zero as $V_{\text{prim}}$ tends to $V_c$ from above, and to a finite value ($c_0$) as $V_{\text{prim}}$ tends to infinity. (A perfect fit over the whole range $V_{\text{prim}} > V_c$, not needed here, would require $c_0 = 1$.) As the PAW core region is classically-allowed in an all-electron approach, there will always exist a classically-allowed region in this type of calculation. As our method has lower resolution for $v \ll 1$, we expect our fitted values of $V_c$ to be estimates of the “true” values. $V_c$ was determined by a root-finding algorithm. When possible, the value of $V_c$ was constrained to lie between the largest tabulated value of $V_{\text{prim}}$ for which $v = 0$, and the smallest tabulated value of $V_{\text{prim}}$ for which $v > 0$. When that was not possible, a tolerance of 3% was afforded, which we note in Table II as well. Many of the metals presented here exhibit more complex $V_{\text{prim}}$-dependence than the insulators, so we perform a piecewise fit

$$v_{\text{metal}}(V_{\text{prim}}) = v(V_{\text{prim}}, c)\Theta(V_0 - V_{\text{prim}}) + v(V_{\text{prim}}, d)\Theta(V_{\text{prim}} - V_0),$$

where both functions on the RHS are of the form of Eq. I. To perform the fit, we chose a value of $V_0$ to model the point of inflection of the curve, and the fitting procedure detailed above was followed for $V_{\text{prim}} \leq V_0$. We then required that $v(V_{\text{prim}})$ and $\partial v(V_{\text{prim}}) / \partial V_{\text{prim}}$ be continuous at $V_{\text{prim}} = V_0$, fixing $d_0$ and $d_1$. A least squares fit was then performed to yield $d_2$ and $d_3$. $V_0$ was modulated to minimize the sum of square residuals, $R^2 = \sum_{V_{\text{prim}}} [v_{\text{approx}}(V_{\text{prim}}) - v(V_{\text{prim}})]^2$. To prevent over-fitting, the lowest value of $V_0$ for which $R^2 < 10^{-3}$

| Solid (structure) | $\varepsilon_{\text{HO}}$ (eV) | $v^\text{max}$ (Å$^3$) | $V_{\text{prim}}$ (Å$^3$) | Lattice Const. (Å) | Expt. Lattice Const. (Å) |
|------------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| Al (fcc)         | 5.75            | 0%              | 16.48           | 4.04            | 4.02            |
| Cu (fcc)         | 5.56            | 0%              | 12.01           | 3.63            | 3.59            |
| Pt (fcc, bulk)   | 4.76            | 0%              | 15.61           | 3.97            | 3.91            |
| Pt monovacency   | -1.18           | 10.9%           | 15.39           | 3.95            | 3.91            |
| Pt (fcc)         | -1.29           | 12.1%           | 15.61           | 3.97            | 3.91            |
| Si (rs) LSDA     | 0.89            | -0.23           | -1.86           | 1.20            | 0.0002          | 30.70           |
| Si (rs) PBE      | 0.86            | -0.09           | -1.90           | 1.13            | 0.0002          | 28.88           |
| NiO PBE          | -0.14           | 4.45            | -8.49           | 4.17            | 0.0004          | 29.56           |

| Solid (structure) | DFA | $c_0$ | $c_1$ | $c_2$ | $c_3$ | $R^2$ | $V_c$ (Å$^3$) |
|------------------|-----|------|------|------|------|-------|-------------|
| Al LSDA          | 0.59| -1.32| 0.90 | -0.17| 0.0008| 48.08 |
| Al PBE           | 0.67| 1.25 | -7.22| 5.96 | 71.18 |
| Pt LSDA          | 7.37| -24.1| 27.18| -10.14| 0.0002| 25.61 |
| Pt PBE           | 1.04| -1.47| 2.31 | -3.14| 47.31 |
| Ne LSDA          | 0.97| -0.27| -0.25| -0.44| 0.0008| 5.85  |
| Ne PBE           | 1.02| -0.84| 1.04 | -1.22| 0.0015| 5.54  |
| NaCl PBE         | 0.89| -0.23| -1.86| 1.20 | 0.0002| 30.70 |
| Si PBE           | 0.86| -0.09| -1.90| 1.13 | 0.0002| 28.88 |
| Si (rs) PBE      | 0.92| -1.34| 0.52 | -0.10| 0.0005| 53.82 |
| NiO PBE          | -0.14| 4.45 | -8.49| 4.17 | 0.0004| 29.56 |
| NiO (rs) PBE     | 0.94| -0.68| -0.40| -0.07| 48.29 |
was deemed the optimal fit.

Let $a$ be the equilibrium lattice parameter for a given solid as given by Table I. From the critical lattice parameters in Table I, we see that a CFR opens in Cu at a lattice parameter of 1.43$a$ for PBE (about 2.9 times the equilibrium volume). The CFR appears before the KS gap opens. The fits predict that a CFR in Al opens at 1.49$a$ for PBE (about 3.3 times the equilibrium volume), also without a KS band gap opening. For Pt, the CFR opens at 1.20$a$ (about 1.7 times the equilibrium volume), without a gap opening. By bandgap, we always mean the gap determined from our approximate band structure or density of states, rather than the fundamental gap.

Note also that the LSDA and PBE curves in Figs. 2 and S4 for Al, Cu, and NaCl cross, whereas those for elemental insulators do not. For the elemental insulators, the difference between the LSDA and PBE curves is always of the same sign.

IV. CFRS IN INSULATORS

The situation for insulators, as shown in Table III, is more nuanced, and there are clear variations in the volumes of approximate CFRs found from different approximate exchange-correlation functionals. Our intuition that the presence of a CFR is accompanied by the opening of a band gap is not borne out.

However in weakly interacting and van der Waals solids, like graphite and Ne, there are noticeable PBE CFRs. The small (1%) PBE CFR volume in graphite (hexagonal C) at its experimental lattice constants reflects the semimetallic nature of this material. The PBE CFR in graphite lies between monolayers, just as one might expect for few-layer graphene. The CFR volume is nearly 20% of the PBE equilibrium cell volume in graphite because PBE underestimates intermediate-range van der Waals interactions, and thus overestimates the equilibrium spacing. This fraction is reduced to 1% when the experimental cell volume is used instead. The LSDA finds no CFR in graphite, which may be related to the LSDA’s underestimation of equilibrium lattice constants. For the prototypical semiconducting layered material MoS$_2$, we see the same pattern. The LSDA underestimates the equilibrium $c$ lattice parameter, yielding no CFR. PBE dramatically overestimates the $c$ parameter, yielding a CFR encompassing 22.3% of the primitive cell volume. Note that the LSDA and PBE are similarly accurate for the sandwich-layer thickness $z$ (the distance between neighboring layers of sulfur atoms).

Consider instead a monolayer of graphite or MoS$_2$. For these sheets, we use the bulk $a$ and $z$ lattice parameters found by relaxing the equilibrium cell volume. We find no CFR within the monolayer region for graphene or monolayer MoS$_2$ using both the LSDA and PBE. Thus, no in-plane CFR is present in graphene, and no in-sandwich CFR is found in monolayer MoS$_2$.

Crystalline NaCl, just like its molecular form [10], also has large PBE and LSDA CFRs. Because NaCl is a prototypical ionic solid, we expect that many other ionic crystals and more weakly-bound crystals at equilibrium will exhibit CFRs.

Referring back to Table, III we see that a CFR emerges in $d$s C when the lattice is stretched to 1.21$a$ for PBE (about 1.8 times the equilibrium volume): for $d$s Si, a CFR is predicted to emerge at 1.06$a$ for PBE (about 1.2 times the equilibrium volume). Thus it appears that PBE predicts the emergence of a CFR in an insulator when the lattice is stretched not much further past its equilibrium point. For both Si and C, the band gap is substantial even when the CFR begins to emerge.

The classical radius of the free Ne atom is 0.87 Å, in both PBE [39], and with a more accurate Kohn-Sham potential [10], with a volume of 2.76 Å$^3$. The experimental lattice constant is 4.464 Å [85], corresponding to a cell volume per atom of 22.24 Å$^3$. The CFR predicted by Ref. [10] is then $(22.24 - 2.76)/22.24 \approx 88\%$ of the total cell volume, agreeing with the values in Table III. A Ne atom in solid Ne at the equilibrium lattice constant is very similar to a free Ne atom.

In the same vein as for C and Si, we can compress the Ne lattice until the CFR vanishes, as seen in Fig. 2. The Ne CFR is predicted to vanish at 0.62$a$ for PBE. One
might expect the bandgap to shrink as the CFR collapses, but the opposite is true. For the smallest lattice constant calculated here (2.85 Å), the band gap is roughly 18.57 eV, compared to a gap of about 11.51 eV (11.45 eV) at the PBE equilibrium (experimental) lattice constant, consistent with previous work that used PBE to study phases of Ne under pressure \[40\]. Intuition suggests that the Ne CFR should not be fully suppressed before the classical turning surface collapses completely, since it is itself a prototype for dichalcogenide structure, and is often referred to as the “MoS\(_2\) structure,” or by its point group 2\(H\)\(_6\), or by its polytype 2\(H\)\(_{6}\)\(_{2}\)\(_{3}\). The parameters of the fit functions (Tables \(S2\)\(_S5\)), as well as full strain curves for these solids (Figs. \(S6\)\(_S9\)) can be found in the Supplementary Materials. The figure shows that the existence of a CFR at equilibrium comes close to classifying a solid as an insulator or metal. No metal has an equilibrium CFR, but some insulators need a small expansion to produce one.

Moreover, we can see very clear trends in the strain curves of elemental solids as one goes down a column of the periodic table. In Fig. 4(a), we plot the strain curves of solid elements (alkaline earth metals, alkaline earth metals, 14 (Group IV, carbon group) and 18 (rare, inert, or noble gases). The parameters of the fit functions (Tables \(S2\)\(_S5\)) can be found in the Supplementary Materials. The figure shows that the existence of a CFR at equilibrium comes close to classifying a solid as an insulator or metal. No metal has an equilibrium CFR, but some insulators need a small expansion to produce one.

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FIG. 3. Contrasting the amount of strain needed to induce a CFR for various solids. The grey line is $V_{\text{prim}} = V_c$, so that solids lying above the grey line have no CFR at equilibrium, and solids below the line have a CFR at equilibrium. For the VASP data (circles for PBE and squares for LSDA), $V_{\text{prim}}$ is the equilibrium cell volume. For the Castep data (diamonds all with PBE), $V_{\text{prim}}$ is the equilibrium cell volume except for the rare gases, for which $V_{\text{prim}}$ takes the experimental value. Metals are shown in blue, and insulators in orange. Note that Sn and Pb were calculated in the cubic diamond structure ($\alpha$- or grey Sn), which are likely non-metallic phases. In particular, grey Sn has a 0.1 eV gap [5]. NiO is treated as spin-unpolarized.

the volume of a free atom as $V_{\text{at}} = 4\pi r_{TS}^2/3$, where $r_{TS}$ is the radius of the atom’s classical turning surface from Ref. [10], then the ratio $V_c/V_{\text{at}}$ is of order 1 and seems to approach a column-dependent large-$Z$ limit (with $Z$ the nuclear charge, see Tables S2-S5). The first ionization energies of the atoms exhibit similar behavior [41].

VI. CFR CONNECTEDNESS

In the introduction, we described a (semi-) classical model of solids that defined metals and insulators by their turning surface properties. In this model, a metal would have a connected classically-allowed region (CAR), and an insulator would have a disconnected CAR.

In Figure 5, we plot the evolution of the CFR and CAR in Si using PBE as a function of the lattice parameter. At equilibrium (panel (a)), there is no CFR. Just above $V_c$ (panel (b)), the CAR is clearly connected and the CFR is disconnected. As the lattice is stretched further (panel (c)), the CFR grows and connects. Under an even more extreme strain (panel (d)), the CFR dominates the primitive cell, but the CAR remains connected, albeit not simply. The bandgap increases from 0.55 eV at $a = 5.47$ Å to 0.81 eV at $a = 5.80$ Å, then decreases to 0.71 eV at $a = 5.87$ Å before rapidly falling toward zero. In Fig. 6, we show a three-dimensional view of the Si turning surface at $a = 7.11$ Å. Both the CFR and CAR are simultaneously fully connected. The geometry of Fig. 6 is very nearly identical to that of Fig. 5(c). To generate the plane of Fig. 5(c) from Fig. 6, one would make a diagonal cut from the front bottom left corner to the rear upper right corner of the cell in Fig. 6.

In the limit of extreme expansion, the CAR’s are always disconnected and well-separated. Electron tunneling is inhibited through energy barriers that are wide or high. The bands will narrow to atomic levels. For a solid built from closed-shell atoms like Ne, the bandgap will tend to a non-zero and typically large value, while for a...
FIG. 5. Evolution of the PBE CAR (grey) and CFR (blue) in ds Si as a function of the lattice parameter \( a \) plotted along the plane \( z = (x + y)/2 \) (i.e., the plane containing the line joining the origin and the point \((a, a, a)\)) within the primitive cell. Ions located in plane are labelled with a black circle, those above with a black triangle, and those below with an open triangle. The lattice constant \( a \) is 5.47 Å in (a), 5.87 Å in (b), 7.07 Å in (c), and 9.07 Å in (d).

VII. CONCLUSIONS

For the solids studied here, our calculations found no CFRs for metals, large CFRs for wide-gap insulators, and the emergence of CFRs when small-gap semiconductors are mildly expanded. Since standard density-gradient expansions are derived for slowly-varying densities without CFRs, the absence of CFRs in metals at equilibrium suggests that generalized gradient approximations (like or beyond PBE) should work especially well for them.

A monovacancy in a metal can induce a CFR, and an expansive strain in any material can induce a CFR or increase its volume. Moreover, the emergence of a CFR does not necessarily accompany the opening or closing of a band gap.

The volume of a CFR is a function of the lattice strain. Both metals and band insulators without a CFR at their equilibrium geometry can be stretched to introduce a CFR. Those wider-gap insulators with a CFR at equilibrium can be compressed until the CFR vanishes. Layered materials may have a CFR at equilibrium if a density functional approximation tends to stretch the \( c \) lattice parameter, as PBE does.

CFRs are also characteristic of perfect ionic and molecular crystals at equilibrium. Our analysis supports the conclusion that rare gas atoms in the crystalline phase are nearly free. Ionic crystals have large CFRs. We showed that graphite and MoS\(_2\), where intermediate-range van der Waals interactions dominate between monolayers, have CFRs located solely between monolayers, and that their corresponding monolayers have no in-plane CFR. Our work demonstrates that weakly-bound solids tend to have prominent CFRs. Hydrogen-bonded crystals like ice, while not tested here, can be expected to have substantial CFR volume fractions, as suggested by Fig. 8 for the water dimer in Ref. [10].

The connectedness of a CFR seems to play a role in a system’s conductivity. It was shown that the CFR in Si near the critical volume \( V_c \) is disconnected. As the lattice is stretched further, the CFR grows, eventually subsuming much of the primitive cell. The Si bandgap closes very nearly at the same \( V_{\text{prim}} \) that the CFR connects, indicating a semiclassical insulator-metal transi-
tion. A semiclassical picture suggests that a connected CAR and zero bandgap indicate a metallic state, found under extreme compression for any solid. As a corollary, disconnected and well-separated CAR’s indicate an insulating state, and are found under strong expansion of the lattice.

Interacting quantum mechanical electrons can insulate through the Mott mechanism. We looked for CFRs in zero-gap spin-unpolarized NiO, a paradigm Mott insulator, but did not find one at the equilibrium lattice constant \(a\). A CFR appeared at a lattice constant 1.18\(a\) (see Tables II and III). Our PBE calculations for NiO at equilibrium confirmed that a gap appears when the spin symmetry is allowed to break to antiferromagnetic order.

This is the first work to attempt to classify CFRs in solids, and without doubt more inquiry is needed to determine if CFRs are hallmarks of other phenomena in solids.

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SUPPLEMENTAL MATERIALS FOR
“CLASSICAL TURNING SURFACES IN SOLIDS:
WHEN DO THEY OCCUR, AND WHAT DO THEY MEAN?”

Here we include extra figures, fit parameters, and data tables that may prove useful for future work. A figure of the density of states for Si at its PBE equilibrium geometry, and at the critical lattice parameter, is given in Fig. S1. A contour plot of $\varepsilon_{HO} - v_s(r)$ in Si along the same plane as in Fig. 1 of the main text is included in Fig. S2.

Fig. S3 is analogous to Fig. 2 of the main text, but emphasizes the shapes of the strain-CFR curves by plotting $V_{prim}/V_c$. A plot of the LSDA fractional CFR volumes in the same manner as Fig. 2 in the main text, with PBE curves superposed faintly, is included in Fig. S4. Table S1 enumerates fitted critical volumes $V_c$ for a comparison between VASP and Castep.

Last are a series of figures and tables for showing fitted strain curves for select main group elements as calculated with PBE in Castep, and using the fitting procedure in the main text, plotted separately for the Group 1, Group 2, Group 14, and Group 18 elemental solids.

We also include numerous tables (Tables S6-S54) enumerating the raw data used for fitting and generating figures. All data presented is available in this text, and machine-readable data will be made available at reasonable request.
FIG. S1. Intensive density of states plots for Si at both the PBE equilibrium lattice parameter 5.47 Å (blue), and at the critical lattice parameter 5.81 Å (orange).
FIG. S2. Contour plot of Si along the [110] (conventional cubic indices) direction at the PBE critical lattice constant $a_c = 5.79 \ \text{Å}$, analogous to Fig. 1 in the main text. The CFR (purple) is minute at this volume, about 0.53% of the primitive cell volume, and located in the interstice. While this indicates the fit is not perfect, it provides a reasonable upper bound to $a_c$. 

$\varepsilon_{\text{HO}} - \nu_s(r)$ for Si at PBE $V_c$
FIG. S3. Figure analogous to Fig. 2 in the main text, but plotting $V_{\text{prim}}/V_c$ to emphasize the shapes of the strain-CFR curves. Perhaps expectedly, the C and Si curves have exceedingly similar shapes. Quite unexpectedly, the NaCl and spin-unpolarized NiO curves are nearly identical.
FIG. S4. Emergence of LSDA (solid lines) and PBE (dashed faint lines) CFRs in Al (blue closed circles), Cu (yellow squares), Pt (green diamonds), C (red point-up triangles), Ne (purple point-down triangles), NaCl (brown crosses), and Si (olive stars) as a function of the lattice constant. All lines are fits given in Table II in the main text. For the elemental insulators, the difference between the LSDA and PBE curves has the same sign. For the metals and NaCl, the LSDA and PBE curves cross.
FIG. S5. Comparison between VASP and Castep results for PBE for the elements presented in the main text.

TABLE S1. Fits for the solids presented in the main text as calculated with PBE in Castep. Materials with two lines of fit parameters use a separate fit for \( V_{\text{prim}} < V_0 \) (first line) and \( V_{\text{prim}} \geq V_0 \) (second) line. For these, \( V_c \) is given on the first line, and \( V_0 \) is given on the second. For the fitting method and fit functions, refer to the main text. The fitted values of \( V_c \) for Ne and Si are too large, and the fitted value of \( V_c \) for Al is too small; all other values of \( V_c \) are within their respective bounds from the numerical calculations.
FIG. S6. Fitted CFR strain curves, in dimensioned (left) and dimensionless (right) forms, for Group 1 (alkali metals) elemental solids as calculated with PBE in Castep.

### TABLE S2. Fit parameters for the Group 1 (alkali metals) elemental solids as calculated with PBE in Castep.

Materials with two lines of fit parameters use a separate fit for $V_{\text{prim}} < V_0$ (first line) and $V_{\text{prim}} \geq V_0$ (second) line. For these, $V_c$ is given on the first line, and $V_0$ is given on the second. For the fitting method and fit functions, refer to the main text. All values of $V_c$ are within their respective bounds from the numerical calculations. When possible, we report the ratio $V_c/V_{\text{at}}$, with $V_{\text{at}} = 4\pi r_{TS}^3/3$, a sphere at the non-relativistic turning surface radius $r_{TS}$ as reported in Ref. [10].

| Solid (struct.) | $c_0$  | $c_1$  | $c_2$  | $c_3$  | $R^2$ | $V_c$ (Å³) | $V_c/V_{\text{at}}$ |
|-----------------|-------|-------|-------|-------|-------|------------|---------------------|
| Li (bcc)        | 13.64 | -48.14| 58.39 | -23.89| 0.0032 | 122.20     | 1.50                |
|                 | 1.51  | -4.29 | 7.92  | -5.93 |        | 178.48     |                     |
| Na (bcc)        | -20.95| 70.29 | -76.07| 26.73 | 0.0180 | 141.92     | 1.37                |
|                 | 0.66  | 1.52  | -4.77 | 2.85  |        | 173.11     |                     |
| K (bcc)         | 1.07  | -1.67 | 1.50  | -0.90 | 0.0056 | 228.89     | 1.18                |
| Rb (bcc)        | 0.79  | -0.11 | -1.14 | 0.46  | 0.0023 | 263.60     | 1.11                |
|                 | 0.89  | 1.54  | -15.46| 26.25 |        | 880.56     |                     |
| Cs (bcc)        | -1.43 | 5.39  | -5.04 | 1.08  | 0.0002 | 345.21     | 1.41                |
|                 | 1.41  | -2.72 | 2.41  | -1.08 |        | 391.33     |                     |
FIG. S7. Fitted CFR strain curves, in dimensioned (left) and dimensionless (right) forms, for Group 2 (alkali earth metals) elemental solids as calculated with PBE in Castep.

| Solid (struct.) | $c_0$ | $c_1$ | $c_2$ | $c_3$ | $R^2$ | $V_c$ ($\text{Å}^3$) | $V_c/V_{at}$ |
|-----------------|-------|-------|-------|-------|-------|----------------|---------------|
| Be (bcc)        | 1.67  | -4.08 | 5.56  | -3.15 | 0.0011 | 46.55          | 1.94          |
|                 | 1.06  | -1.39 | 2.89  | -3.84 |        | 108.23         |               |
| Mg (bcc)        | 1.86  | -4.58 | 5.56  | -2.84 | 0.0009 | 80.54          | 1.86          |
|                 | 1.06  | -1.97 | 3.52  | -2.98 |        | 133.80         |               |
| Ca (fcc)        | 43.72 | -147.30 | 167.62 | -64.05 | 0.0004 | 154.37         | 1.66          |
|                 | 0.97  | -0.59 | -0.17 | -0.09 |        | 178.96         |               |
| Sr (fcc)        | 38.23 | -129.32 | 147.96 | -56.87 | 0.0007 | 189.97         | 1.55          |
|                 | 1.24  | -2.16 | 2.63  | -1.67 |        | 229.06         |               |
| Ba (bcc)        | 17.22 | -56.94 | 65.16  | -25.43 | 0.0007 | 236.59         |               |
|                 | 1.20  | -1.68 | 1.62  | -1.09 |        | 275.68         |               |
| Ra (bcc)        | 8.39  | -30.21 | 38.44  | -16.62 | 0.0003 | 244.61         |               |
|                 | 0.71  | 0.57  | -2.02 | 0.89  |        | 285.89         |               |

TABLE S3. Fit parameters for the Group 2 (alkali earth metals) elemental solids as calculated with PBE in Castep. Materials with two lines of fit parameters use a separate fit for $V_{prim} < V_0$ (first line) and $V_{prim} \geq V_0$ (second) line. For these, $V_c$ is given on the first line, and $V_0$ is given on the second. For the fitting method and fit functions, refer to the main text. The fitted values of $V_c$ for Ba and Ra are too small; all other values of $V_c$ are within their respective bounds from the numerical calculations. When possible, we report the ratio $V_c/V_{at}$, with $V_{at} = 4\pi r_{TS}^3/3$, a sphere at the non-relativistic turning surface radius $r_{TS}$ as reported in Ref. [10].
FIG. S8. Fitted CFR strain curves, in dimensioned (left) and dimensionless (right) forms, for Group 14 (carbon group) elemental solids as calculated with PBE in Castep.

| Solid (struct.) | $c_0$ | $c_1$ | $c_2$ | $c_3$ | $R^2$ | $V_c$ (Å$^3$) | $V_c/V_{at}$ |
|-----------------|-------|-------|-------|-------|-------|--------------|--------------|
| C (ds)          | 1.04  | -1.73 | 1.18  | -0.50 | 0.0015 | 21.71        | 1.57         |
| Si (ds)         | 0.97  | -1.63 | 1.19  | -0.52 | 0.0014 | 51.54        | 1.24         |
| Ge (ds)         | 1.09  | -2.31 | 2.34  | -1.12 | 0.0001 | 55.17        | 1.22         |
| Sn (ds)         | 0.93  | -1.56 | 1.17  | -0.54 | 0.0003 | 71.37        | 1.17         |
| Pb (ds)         | 0.95  | -1.69 | 1.42  | -0.69 | 0.0003 | 77.56        |              |

TABLE S4. Fit parameters for the Group 14 (carbon group) elemental solids as calculated with PBE in Castep. The fitted $V_c$ for Si is too large; all other values of $V_c$ are within their respective bounds from the numerical calculations. When possible, we report the ratio $V_c/V_{at}$, with $V_{at} = 4\pi r_{TS}^3/3$, a sphere at the non-relativistic turning surface radius $r_{TS}$ as reported in Ref. [10].
FIG. S9. Fitted CFR strain curves, in dimensioned (left) and dimensionless (right) forms, for Group 18 (rare gases) elemental solids as calculated with PBE in Castep.

| Solid (struc.) | $c_0$ | $c_1$ | $c_2$ | $c_3$ | $R^2$ | $V_c$ (Å$^3$) | $V_c/V_{at}$ |
|----------------|-------|-------|-------|-------|-------|----------------|---------------|
| He (fcc)       | 1.00  | -0.40 | 0.27  | -0.87 | 0.0013 | 2.63           | 2.63          |
| Ne (fcc)       | 0.99  | -0.46 | -0.29 | -0.25 | 0.0061 | 5.49           | 1.99          |
| Ar (fcc)       | 0.98  | -0.45 | -0.19 | -0.34 | 0.0072 | 14.91          | 1.66          |
| Kr (fcc)       | 0.91  | 0.11  | -1.48 | 0.46  | 0.0139 | 20.43          | 1.54          |
| Xe (fcc)       | 0.74  | 1.03  | -3.15 | 1.38  | 0.0128 | 30.21          | 1.44          |

TABLE S5. Fit parameters for the Group 18 (rare gas) elemental solids as calculated with PBE in Castep. All fitted values of $V_c$ are too large, except for He. When possible, we report the ratio $V_c/V_{at}$, with $V_{at} = 4\pi r_{TS}^3/3$, a sphere at the non-relativistic turning surface radius $r_{TS}$ as reported in Ref. [10].
### S1. RAW DATA FOR ELEMENTS EMPHASIZED IN MAIN TEXT

**TABLE S6:** Raw data for Al as calculated with PBE in VASP.

| $V_{\text{prim}}$ (Å$^3$) | CFR fraction | Band gap from DOS (eV) |
|--------------------------|--------------|------------------------|
| 16.4848                  | 0.0000       | 0.0000                 |
| 19.0563                  | 0.0000       | 0.0000                 |
| 21.8821                  | 0.0000       | 0.0000                 |
| 24.9743                  | 0.0000       | 0.0000                 |
| 28.3450                  | 0.0000       | 0.0000                 |
| 32.0060                  | 0.0000       | 0.0000                 |
| 35.9695                  | 0.0000       | 0.0000                 |
| 40.2473                  | 0.0000       | 0.0000                 |
| 44.8515                  | 0.0000       | 0.0000                 |
| 49.7942                  | 0.0000       | 0.0000                 |
| 55.0872                  | 0.0081       | 0.0000                 |
| 60.7427                  | 0.0264       | 0.0000                 |
| 66.7725                  | 0.0491       | 0.0000                 |
| 73.1887                  | 0.0756       | 0.0000                 |
| 80.0034                  | 0.1056       | 0.0000                 |
| 87.2284                  | 0.1461       | 0.0000                 |
| 94.8759                  | 0.2427       | 0.0000                 |
| 102.9577                 | 0.3472       | 0.0000                 |
| 111.4859                 | 0.4219       | 0.0000                 |
| 120.4726                 | 0.4801       | 0.0000                 |
| 129.9296                 | 0.5185       | 0.0000                 |
| 139.8691                 | 0.5466       | 0.0000                 |
| 150.3029                 | 0.5656       | 0.0000                 |
| 161.2431                 | 0.5840       | 0.0000                 |
| 172.7018                 | 0.6022       | 0.0000                 |
| 184.6908                 | 0.6202       | 0.0000                 |
| 197.2223                 | 0.6378       | 0.0000                 |
| 210.3081                 | 0.6550       | 0.0000                 |
| 223.9603                 | 0.6712       | 0.0000                 |
| 238.1910                 | 0.6872       | 0.0000                 |
| 253.0120                 | 0.7024       | 0.0000                 |
TABLE S7: Raw data for Cu as calculated with PBE in VASP.

| $V_{\text{prim}}$ (Å$^3$) | CFR fraction | Band gap from DOS (eV) |
|----------------------------|--------------|-----------------------|
| 11.9580                    | 0.0000       | 0.0000                |
| 14.0455                    | 0.0000       | 0.0000                |
| 16.3627                    | 0.0000       | 0.0000                |
| 18.9217                    | 0.0000       | 0.0000                |
| 21.7346                    | 0.0000       | 0.0000                |
| 24.8132                    | 0.0000       | 0.0000                |
| 28.1696                    | 0.0000       | 0.0000                |
| 31.8159                    | 0.0000       | 0.0000                |
| 35.7639                    | 0.0140       | 0.0000                |
| 40.0258                    | 0.0523       | 0.0000                |
| 44.6134                    | 0.1074       | 0.0000                |
| 49.5388                    | 0.2328       | 0.0000                |
| 54.8141                    | 0.4168       | 0.0000                |
| 60.4511                    | 0.5237       | 0.0000                |
| 66.4619                    | 0.5701       | 0.0000                |
| 72.8586                    | 0.6038       | 0.0000                |
| 79.6530                    | 0.6353       | 0.0000                |
| 86.8572                    | 0.6651       | 0.0000                |
| 94.4833                    | 0.6925       | 0.0000                |
| 102.5431                   | 0.7171       | 0.0000                |
| 111.0487                   | 0.7396       | 0.0000                |
| 120.0122                   | 0.7599       | 0.0000                |
| 129.4454                   | 0.7782       | 0.0000                |
| 139.3604                   | 0.7944       | 0.0000                |
| 149.7693                   | 0.8086       | 0.0000                |
| 160.6839                   | 0.8219       | 0.0000                |
| 172.1163                   | 0.8337       | 0.0000                |
| 184.0786                   | 0.8446       | 0.0000                |
| 196.5826                   | 0.8541       | 0.0000                |
| 209.6405                   | 0.8632       | 0.0000                |
| 223.2641                   | 0.8713       | 0.0000                |
| 237.4655                   | 0.8785       | 0.0000                |
| 252.2568                   | 0.8855       | 0.0000                |
| V_{\text{prim}} (\text{Å}^3) | CFR fraction | Band gap from DOS (eV) |
|--------------------------|--------------|----------------------|
| 15.6427                  | 0.0000       | 0.0000               |
| 18.1279                  | 0.0000       | 0.0000               |
| 20.8634                  | 0.0000       | 0.0000               |
| 23.8610                  | 0.0000       | 0.0000               |
| 27.1328                  | 0.0059       | 0.0000               |
| 30.6909                  | 0.0411       | 0.0000               |
| 34.5471                  | 0.0890       | 0.0000               |
| 38.7135                  | 0.1846       | 0.0000               |
| 43.2022                  | 0.3464       | 0.0000               |
| 48.0250                  | 0.4608       | 0.0000               |
| 53.1940                  | 0.5255       | 0.0000               |
| 58.7213                  | 0.5652       | 0.0000               |
| 64.6187                  | 0.5994       | 0.0000               |
| 70.8983                  | 0.6311       | 0.0000               |
| 77.5722                  | 0.6605       | 0.0000               |
| 84.6522                  | 0.6871       | 0.0000               |
| 92.1505                  | 0.7105       | 0.0000               |
| 100.0789                 | 0.7317       | 0.0000               |
| 108.4495                 | 0.7510       | 0.0000               |
| 117.2744                 | 0.7687       | 0.0000               |
| 126.5654                 | 0.7844       | 0.0000               |
| 136.3346                 | 0.7990       | 0.0000               |
| 146.5941                 | 0.8121       | 0.0000               |
| 157.3557                 | 0.8244       | 0.0000               |
| 168.6315                 | 0.8355       | 0.0000               |
| 180.4336                 | 0.8457       | 0.0000               |
| 192.7738                 | 0.8550       | 0.0000               |
| 205.6642                 | 0.8637       | 0.0000               |
| 219.1169                 | 0.8718       | 0.0000               |
| 233.1437                 | 0.8793       | 0.0000               |
| 247.7567                 | 0.8863       | 0.0000               |
TABLE S9: Raw data for C as calculated with PBE in VASP.

| $V_{\text{prim}}$ (Å$^3$) | CFR fraction | Band gap from DOS (eV) |
|-----------------------------|-------------|------------------------|
| 11.3748                     | 0.0000      | 4.1395                 |
| 13.3957                     | 0.0000      | 3.7061                 |
| 15.6427                     | 0.0000      | 3.4592                 |
| 18.1279                     | 0.0000      | 3.0474                 |
| 20.8634                     | 0.0212      | 2.3060                 |
| 23.8610                     | 0.1116      | 0.5534                 |
| 27.1328                     | 0.1794      | 0.0000                 |
| 30.6909                     | 0.2407      | 0.0000                 |
| 34.5471                     | 0.3036      | 0.0000                 |
| 38.7135                     | 0.3615      | 0.0000                 |
| 43.2022                     | 0.4113      | 0.0000                 |
| 48.0250                     | 0.4548      | 0.0000                 |
| 53.1940                     | 0.4946      | 0.0000                 |
| 58.7213                     | 0.5302      | 0.0000                 |
| 64.6187                     | 0.5637      | 0.0000                 |
| 70.8983                     | 0.5943      | 0.0000                 |
| 77.5722                     | 0.6228      | 0.0000                 |
| 84.6522                     | 0.6492      | 0.0000                 |
| 92.1505                     | 0.6747      | 0.0000                 |
| 100.0789                    | 0.6986      | 0.0000                 |
| 108.4495                    | 0.7214      | 0.0000                 |
| 117.2744                    | 0.7436      | 0.0000                 |
| 126.5654                    | 0.7654      | 0.0000                 |
| 136.3346                    | 0.7839      | 0.0000                 |
| 146.5941                    | 0.7981      | 0.0000                 |
| 157.3557                    | 0.8104      | 0.0000                 |
| 168.6315                    | 0.8217      | 0.0000                 |
| 180.4336                    | 0.8321      | 0.0000                 |
| 192.7738                    | 0.8421      | 0.0000                 |
| 205.6642                    | 0.8509      | 0.0000                 |
| 219.1169                    | 0.8593      | 0.0000                 |
| 233.1437                    | 0.8669      | 0.0000                 |
| 247.7567                    | 0.8743      | 0.0000                 |
TABLE S10: Raw data for Ne as calculated with PBE in VASP.

| $V_{prim}$ (Å$^3$) | CFR fraction | Band gap from DOS (eV) |
|---------------------|--------------|------------------------|
| 5.7873              | 0.0933       | 18.5758                |
| 6.0972              | 0.1847       | 17.9035                |
| 6.4181              | 0.3122       | 17.5081                |
| 6.7500              | 0.3719       | 16.9491                |
| 8.1920              | 0.5404       | 14.9407                |
| 9.8260              | 0.6535       | 13.8288                |
| 11.6640             | 0.7240       | 12.9718                |
| 13.7180             | 0.7763       | 12.4658                |
| 16.0000             | 0.8088       | 12.1625                |
| 18.5220             | 0.8356       | 11.7600                |
| 21.2960             | 0.8570       | 11.5588                |
| 24.3340             | 0.8745       | 11.4974                |
| 27.6480             | 0.8899       | 11.2233                |
| 31.2500             | 0.9024       | 11.2782                |
| 35.1520             | 0.9131       | 11.2431                |
| 39.3660             | 0.9227       | 11.3382                |
| 43.9040             | 0.9306       | 11.3177                |
| 48.7780             | 0.9373       | 11.3249                |
| 54.0000             | 0.9436       | 11.3459                |
TABLE S11: Raw data for NaCl as calculated with PBE in VASP.

| $V_{prim}$ (Å$^3$) | CFR fraction | Band gap from DOS (eV) |
|---------------------|--------------|------------------------|
| 12.6633             | 0.0000       | 0.2540                 |
| 14.8297             | 0.0000       | 2.3167                 |
| 17.2302             | 0.0000       | 3.8483                 |
| 19.8767             | 0.0000       | 4.9494                 |
| 22.7813             | 0.0000       | 5.9849                 |
| 25.9558             | 0.0134       | 6.6959                 |
| 29.4123             | 0.0809       | 7.1671                 |
| 33.1627             | 0.1739       | 6.5604                 |
| 37.2193             | 0.2617       | 5.9844                 |
| 41.5938             | 0.3438       | 5.3491                 |
| 46.2983             | 0.4150       | 4.9802                 |
| 51.3447             | 0.4699       | 4.5501                 |
| 56.7452             | 0.5212       | 4.3312                 |
| 62.5117             | 0.5700       | 4.0477                 |
| 68.6562             | 0.6093       | 3.7484                 |
| 75.1907             | 0.6404       | 3.6438                 |
| 82.1273             | 0.6765       | 3.3609                 |
| 89.4778             | 0.6987       | 3.3206                 |
| 97.2542             | 0.7181       | 3.0762                 |
| 105.4688            | 0.7342       | 3.0424                 |
| 114.1333            | 0.7463       | 2.9033                 |
| 123.2598            | 0.7611       | 2.6795                 |
| 132.8603            | 0.7705       | 2.6415                 |
| 142.9468            | 0.7796       | 2.5432                 |
| 153.5312            | 0.7896       | 2.4179                 |
| 164.6258            | 0.7971       | 2.2904                 |
| 176.2422            | 0.8059       | 2.1310                 |
| 188.3928            | 0.8094       | 2.1254                 |
| 201.0893            | 0.8160       | 1.9031                 |
| 214.3438            | 0.8215       | 1.7761                 |
| 228.1683            | 0.8291       | 1.6488                 |
| 242.5748            | 0.8291       | 1.6157                 |
TABLE S12: Raw data for Si as calculated with PBE in VASP.

| V_{prim} (Å³) | CFR fraction | Band gap from DOS (eV) |
|---------------|--------------|------------------------|
| 40.9168       | 0.0000       | 0.5507                 |
| 45.5711       | 0.0000       | 0.7547                 |
| 48.7857       | 0.0064       | 0.8074                 |
| 50.5655       | 0.0242       | 0.7107                 |
| 55.9121       | 0.0849       | 0.0000                 |
| 61.6230       | 0.1457       | 0.0000                 |
| 67.7100       | 0.1885       | 0.0000                 |
| 74.1852       | 0.2351       | 0.0000                 |
| 81.0607       | 0.2779       | 0.0000                 |
| 88.3483       | 0.3186       | 0.0000                 |
| 96.0601       | 0.3554       | 0.0000                 |
| 104.2082      | 0.3890       | 0.0000                 |
| 112.8044      | 0.4199       | 0.0000                 |
| 121.8609      | 0.4493       | 0.0000                 |
| 131.3895      | 0.4764       | 0.0000                 |
| 141.4023      | 0.5022       | 0.0000                 |
| 151.9114      | 0.5264       | 0.0000                 |
| 162.9286      | 0.5491       | 0.0000                 |
| 174.4660      | 0.5709       | 0.0000                 |
| 186.5357      | 0.5916       | 0.0000                 |
| 199.1495      | 0.6114       | 0.0000                 |
| 212.3195      | 0.6304       | 0.0000                 |
| 226.0578      | 0.6488       | 0.0000                 |
| 240.3762      | 0.6661       | 0.0000                 |
| 255.2868      | 0.6834       | 0.0000                 |
TABLE S13: Raw data for spin-unpolarized NiO as calculated with PBE in VASP.

| $V'_{prim}$ (Å$^3$) | CFR fraction | Band gap from DOS (eV) |
|---------------------|--------------|------------------------|
| 17.9978             | 0.0000       | 0.0000                 |
| 20.7205             | 0.0000       | 0.0000                 |
| 23.7047             | 0.0000       | 0.0000                 |
| 26.9625             | 0.0000       | 0.0000                 |
| 30.5060             | 0.0007       | 0.0000                 |
| 34.3470             | 0.0691       | 0.0000                 |
| 38.4977             | 0.1572       | 0.0000                 |
| 42.9699             | 0.2711       | 0.0000                 |
| 47.7757             | 0.3531       | 0.0000                 |
| 52.9272             | 0.4213       | 0.0000                 |
| 58.4362             | 0.4829       | 0.0000                 |
| 64.3149             | 0.5394       | 0.0000                 |
| 70.5751             | 0.5882       | 0.0000                 |
| 77.2289             | 0.6259       | 0.0000                 |
| 84.2884             | 0.6593       | 0.0000                 |
| 91.7654             | 0.6881       | 0.0000                 |
| 99.6721             | 0.7116       | 0.0000                 |
| 108.0203            | 0.7293       | 0.0000                 |
| 116.8221            | 0.7454       | 0.0000                 |
| 126.0896            | 0.7604       | 0.2030                 |
| 135.8346            | 0.7739       | 0.0000                 |
| 146.0693            | 0.7866       | 0.0000                 |
| 156.8055            | 0.7986       | 0.2018                 |
| 168.0553            | 0.8097       | 0.1669                 |
| 179.8308            | 0.8203       | 0.0000                 |
| 192.1438            | 0.8300       | 0.0000                 |
| 205.0065            | 0.8389       | 0.1942                 |
| 218.4307            | 0.8476       | 0.1132                 |
| 232.4285            | 0.8553       | 0.1173                 |
| 247.0120            | 0.8628       | 0.0000                 |
TABLE S14: Raw data for Al as calculated with LSDA in VASP.

| $V_{pol}$ (Å$^3$) | CFR fraction | Band gap from DOS (eV) |
|-------------------|--------------|------------------------|
| 15.7612           | 0.0000       | 0.0000                 |
| 18.2587           | 0.0000       | 0.0000                 |
| 21.0069           | 0.0000       | 0.0000                 |
| 24.0180           | 0.0000       | 0.0000                 |
| 27.3038           | 0.0000       | 0.0000                 |
| 30.8765           | 0.0000       | 0.0000                 |
| 34.7480           | 0.0000       | 0.0000                 |
| 38.9302           | 0.0000       | 0.0000                 |
| 43.4353           | 0.0000       | 0.0000                 |
| 48.2751           | 0.0002       | 0.0000                 |
| 53.4618           | 0.0081       | 0.0000                 |
| 59.0073           | 0.0215       | 0.0000                 |
| 64.9235           | 0.0390       | 0.0000                 |
| 71.2226           | 0.0597       | 0.0000                 |
| 77.9164           | 0.0865       | 0.0000                 |
| 85.0171           | 0.1297       | 0.0000                 |
| 92.5366           | 0.1997       | 0.0000                 |
| 100.4868          | 0.2644       | 0.0000                 |
| 108.8799          | 0.3272       | 0.0000                 |
| 117.7277          | 0.3868       | 0.0000                 |
| 127.0424          | 0.4432       | 0.0000                 |
| 136.8359          | 0.4850       | 0.0000                 |
| 147.1201          | 0.5199       | 0.0000                 |
| 157.9072          | 0.5494       | 0.0000                 |
| 169.2090          | 0.5759       | 0.0000                 |
| 181.0377          | 0.5993       | 0.0000                 |
| 193.4052          | 0.6207       | 0.0000                 |
| 206.3234          | 0.6403       | 0.0000                 |
| 219.8045          | 0.6585       | 0.0000                 |
| 233.8603          | 0.6751       | 0.0000                 |
| 248.5030          | 0.6908       | 0.0000                 |
TABLE S15: Raw data for Cu as calculated with LSDA in VASP.

| $V_{\text{point}}$ (Å$^3$) | CFR fraction | Band gap from DOS (eV) |
|-----------------------------|--------------|------------------------|
| 10.9036                     | 0.0000       | 0.0000                 |
| 12.8697                     | 0.0000       | 0.0000                 |
| 15.0591                     | 0.0000       | 0.0000                 |
| 17.4836                     | 0.0000       | 0.0000                 |
| 20.1554                     | 0.0000       | 0.0000                 |
| 23.0864                     | 0.0000       | 0.0000                 |
| 26.2885                     | 0.0000       | 0.0000                 |
| 29.7739                     | 0.0020       | 0.0000                 |
| 33.5544                     | 0.0225       | 0.0000                 |
| 37.6422                     | 0.0562       | 0.0000                 |
| 42.0492                     | 0.1090       | 0.0000                 |
| 46.7873                     | 0.2345       | 0.0000                 |
| 51.8687                     | 0.3557       | 0.0000                 |
| 57.3052                     | 0.4634       | 0.0000                 |
| 63.1090                     | 0.5328       | 0.0000                 |
| 69.2920                     | 0.5834       | 0.0000                 |
| 75.8661                     | 0.6249       | 0.0000                 |
| 82.8435                     | 0.6593       | 0.0000                 |
| 90.2360                     | 0.6889       | 0.0000                 |
| 98.0558                     | 0.7143       | 0.0000                 |
| 106.3148                    | 0.7367       | 0.0000                 |
| 115.0249                    | 0.7564       | 0.0000                 |
| 124.1983                    | 0.7742       | 0.0000                 |
| 133.8468                    | 0.7902       | 0.0000                 |
| 143.9826                    | 0.8047       | 0.0000                 |
| 154.6176                    | 0.8180       | 0.0000                 |
| 165.7637                    | 0.8299       | 0.0000                 |
| 177.4331                    | 0.8408       | 0.0000                 |
| 189.6376                    | 0.8506       | 0.0000                 |
| 202.3894                    | 0.8600       | 0.0000                 |
| 215.7004                    | 0.8685       | 0.0000                 |
| 229.5825                    | 0.8763       | 0.0000                 |
| 244.0479                    | 0.8835       | 0.0000                 |
TABLE S16: Raw data for Pt as calculated with LSDA in VASP.

| V_{pmm} (Å³) | CFR fraction | Band gap from DOS (eV) |
|--------------|--------------|------------------------|
| 14.8297      | 0.0000       | 0.0000                 |
| 17.2302      | 0.0000       | 0.0000                 |
| 19.8767      | 0.0000       | 0.0000                 |
| 22.7813      | 0.0000       | 0.0000                 |
| 25.9558      | 0.0052       | 0.0000                 |
| 29.4123      | 0.0314       | 0.0000                 |
| 33.1627      | 0.0724       | 0.0000                 |
| 37.2193      | 0.1653       | 0.0000                 |
| 41.5938      | 0.2868       | 0.0000                 |
| 46.2983      | 0.3966       | 0.0000                 |
| 51.3447      | 0.4821       | 0.0000                 |
| 56.7452      | 0.5396       | 0.0000                 |
| 62.5117      | 0.5843       | 0.0000                 |
| 68.6562      | 0.6213       | 0.0000                 |
| 75.1907      | 0.6525       | 0.0000                 |
| 82.1273      | 0.6802       | 0.0000                 |
| 89.4778      | 0.7042       | 0.0000                 |
| 97.2542      | 0.7253       | 0.0000                 |
| 105.4688     | 0.7445       | 0.0000                 |
| 114.1333     | 0.7620       | 0.0000                 |
| 123.2598     | 0.7777       | 0.0000                 |
| 132.8603     | 0.7928       | 0.0000                 |
| 142.9468     | 0.8059       | 0.0000                 |
| 153.5312     | 0.8182       | 0.0000                 |
| 164.6258     | 0.8295       | 0.0000                 |
| 176.2422     | 0.8400       | 0.0000                 |
| 188.3928     | 0.8494       | 0.0000                 |
| 201.0893     | 0.8584       | 0.0000                 |
| 214.3438     | 0.8667       | 0.0000                 |
| 228.1683     | 0.8746       | 0.0000                 |
| 242.5748     | 0.8817       | 0.0000                 |
TABLE S17: Raw data for C as calculated with LSDA in VASP.

| $V_{prim}$ (Å$^3$) | CFR fraction | Band gap from DOS (eV) |
|-------------------|--------------|------------------------|
| 10.9967           | 0.0000       | 4.0588                 |
| 12.9738           | 0.0000       | 3.7563                 |
| 15.1746           | 0.0000       | 3.3818                 |
| 17.6112           | 0.0000       | 3.0743                 |
| 20.2957           | 0.0000       | 2.5228                 |
| 23.2399           | 0.0097       | 1.0881                 |
| 26.4560           | 0.0827       | 0.0000                 |
| 29.9558           | 0.1643       | 0.0000                 |
| 33.7514           | 0.2401       | 0.0000                 |
| 37.8549           | 0.3093       | 0.0000                 |
| 42.2781           | 0.3676       | 0.0000                 |
| 47.0331           | 0.4171       | 0.0000                 |
| 52.1320           | 0.4621       | 0.0000                 |
| 57.5866           | 0.5012       | 0.0000                 |
| 63.4090           | 0.5374       | 0.0000                 |
| 69.6113           | 0.5703       | 0.0000                 |
| 76.2053           | 0.6005       | 0.0000                 |
| 83.2031           | 0.6294       | 0.0000                 |
| 90.6168           | 0.6555       | 0.0000                 |
| 98.4582           | 0.6803       | 0.0000                 |
| 106.7394          | 0.7041       | 0.0000                 |
| 115.4725          | 0.7258       | 0.0000                 |
| 124.6693          | 0.7471       | 0.0000                 |
| 134.3419          | 0.7665       | 0.0000                 |
| 144.5024          | 0.7837       | 0.0000                 |
| 155.1626          | 0.7981       | 0.0000                 |
| 166.3347          | 0.8106       | 0.0000                 |
| 178.0305          | 0.8224       | 0.0000                 |
| 190.2621          | 0.8327       | 0.0000                 |
| 203.0416          | 0.8422       | 0.0000                 |
| 216.3808          | 0.8512       | 0.0000                 |
| 230.2918          | 0.8591       | 0.0000                 |
| 244.7867          | 0.8666       | 0.0000                 |
TABLE S18: Raw data for Ne as calculated with LSDA in VASP.

| $V_{\text{prim}}$ (Å³) | CFR fraction | Band gap from DOS (eV) |
|-------------------------|--------------|------------------------|
| 5.7873                  | 0.0261       | 18.3436                |
| 6.0972                  | 0.0576       | 17.6710                |
| 6.4181                  | 0.1407       | 17.0007                |
| 6.7500                  | 0.2294       | 16.4522                |
| 8.1920                  | 0.5164       | 14.4664                |
| 9.8260                  | 0.6455       | 13.4261                |
| 11.6640                 | 0.7178       | 12.4733                |
| 13.7180                 | 0.7691       | 11.9869                |
| 16.0000                 | 0.8060       | 11.6004                |
| 18.5220                 | 0.8354       | 11.3754                |
| 21.2960                 | 0.8570       | 11.3931                |
| 24.3340                 | 0.8745       | 11.3476                |
| 27.6480                 | 0.8899       | 11.2333                |
| 31.2500                 | 0.9024       | 11.3534                |
| 35.1520                 | 0.9131       | 11.3287                |
| 39.3660                 | 0.9227       | 11.4308                |
| 43.9040                 | 0.9306       | 11.5933                |
| 48.7780                 | 0.9373       | 11.6459                |
| 54.0000                 | 0.9436       | 11.7969                |
| \(V_{ \text{prim} } (\AA^3)\) | CFR fraction | Band gap from DOS (eV) |
|-----------------|--------------|---------------------|
| 10.4455         | 0.0000       | 0.0000              |
| 12.3577         | 0.0000       | 0.0000              |
| 14.4902         | 0.0000       | 1.7634              |
| 16.8548         | 0.0000       | 3.2939              |
| 19.4636         | 0.0000       | 4.4587              |
| 22.3287         | 0.0000       | 5.4665              |
| 25.4619         | 0.0000       | 6.1151              |
| 28.8753         | 0.0000       | 6.7088              |
| 32.5810         | 0.0286       | 6.2297              |
| 36.5908         | 0.0884       | 5.5120              |
| 40.9168         | 0.1780       | 5.0743              |
| 45.5711         | 0.2580       | 4.5858              |
| 50.5655         | 0.3391       | 4.2189              |
| 55.9121         | 0.4023       | 3.9347              |
| 61.6230         | 0.4586       | 3.7337              |
| 67.7100         | 0.5119       | 3.5726              |
| 74.1852         | 0.5587       | 3.4197              |
| 81.0607         | 0.5957       | 3.1547              |
| 88.3483         | 0.6348       | 3.0390              |
| 96.0601         | 0.6682       | 2.9438              |
| 104.2082        | 0.6958       | 2.8269              |
| 112.8044        | 0.7157       | 2.7118              |
| 121.8609        | 0.7331       | 2.5950              |
| 131.3895        | 0.7504       | 2.4746              |
| 141.4023        | 0.7623       | 2.3570              |
| 151.9114        | 0.7769       | 2.3220              |
| 162.9286        | 0.7869       | 2.2251              |
| 174.4660        | 0.7982       | 2.1907              |
| 186.5357        | 0.8045       | 2.0956              |
| 199.1495        | 0.8142       | 1.9712              |
| 212.3195        | 0.8224       | 1.9687              |
| 226.0578        | 0.8293       | 1.8462              |
TABLE S20: Raw data for Si as calculated with LSDA in VASP.

| $V_{prim}$ (Å$^3$) | CFR fraction | Band gap from DOS (eV) |
|---------------------|--------------|------------------------|
| 39.3660             | 0.0000       | 0.3872                 |
| 43.9040             | 0.0000       | 0.6069                 |
| 48.7780             | 0.0000       | 0.6498                 |
| 54.0000             | 0.0105       | 0.1719                 |
| 59.5820             | 0.0497       | 0.0000                 |
| 65.5360             | 0.1094       | 0.0000                 |
| 71.8740             | 0.1685       | 0.0000                 |
| 78.6080             | 0.2202       | 0.0000                 |
| 85.7500             | 0.2672       | 0.0000                 |
| 93.3120             | 0.3090       | 0.0000                 |
| 101.3060            | 0.3468       | 0.0000                 |
| 109.7440            | 0.3819       | 0.0000                 |
| 118.6380            | 0.4135       | 0.0000                 |
| 128.0000            | 0.4430       | 0.0000                 |
| 137.8420            | 0.4716       | 0.0000                 |
| 148.1760            | 0.4970       | 0.0000                 |
| 159.0140            | 0.5221       | 0.0000                 |
| 170.3680            | 0.5455       | 0.0000                 |
| 182.2500            | 0.5678       | 0.0000                 |
| 194.6720            | 0.5892       | 0.0000                 |
| 207.6460            | 0.6091       | 0.0000                 |
| 221.1840            | 0.6285       | 0.0000                 |
| 235.2980            | 0.6472       | 0.0000                 |
| 250.0000            | 0.6648       | 0.0000                 |
TABLE S21: Raw data for Al as calculated with PBE in Castep.

| $V_{\text{cell}}$ (Å³) | CFR fraction  |
|---------------|-------------|
| 3.9062        | 0.0000      |
| 4.3940        | 0.0000      |
| 4.9207        | 0.0000      |
| 5.4880        | 0.0000      |
| 6.0972        | 0.0000      |
| 6.7500        | 0.0000      |
| 7.4478        | 0.0000      |
| 8.1920        | 0.0000      |
| 8.9842        | 0.0000      |
| 9.8260        | 0.0000      |
| 10.7188       | 0.0000      |
| 11.6640       | 0.0000      |
| 12.6633       | 0.0005      |
| 13.7180       | 0.0000      |
| 14.8297       | 0.0000      |
| 16.0000       | 0.0000      |
| 17.2302       | 0.0000      |
| 18.5220       | 0.0003      |
| 19.8767       | 0.0003      |
| 21.2960       | 0.0003      |
| 22.7812       | 0.0003      |
| 24.3340       | 0.0000      |
| 25.9558       | 0.0003      |
| 27.6480       | 0.0015      |
| 29.4123       | 0.0015      |
| 31.2500       | 0.0015      |
| 33.1627       | 0.0053      |
| 35.1520       | 0.0053      |
| 37.2192       | 0.0053      |
| 39.3660       | 0.0018      |
| 41.5938       | 0.0062      |
| 43.9040       | 0.0107      |
| 46.2983       | 0.0153      |
| 48.7780       | 0.0184      |
| 51.3448       | 0.0222      |
| 54.0000       | 0.0262      |
| 56.7452       | 0.0297      |
| 59.5920       | 0.0330      |
| 62.5117       | 0.0371      |
| 65.5360       | 0.0412      |
| 68.6562       | 0.0484      |
| 71.8740       | 0.0587      |
| 75.1908       | 0.0685      |
| 78.6080       | 0.0744      |
| 82.1273       | 0.0897      |
| 85.7500       | 0.1089      |
| 89.4777       | 0.1378      |
| 93.3120       | 0.1732      |
| 97.2542       | 0.2452      |
| 101.3060      | 0.3023      |
| 105.4690      | 0.3382      |
| 109.7440      | 0.3814      |
| 114.1330      | 0.4149      |
| 118.6380      | 0.4464      |
| 123.2600      | 0.4718      |
### TABLE S22: Raw data for Cu as calculated with PBE in Castep.

| $V_{prim}$ (Å$^3$) | CFR fraction |
|---------------------|--------------|
| 3.9062              | 0.0000       |
| 4.3940              | 0.0000       |
| 4.9207              | 0.0000       |
| 5.4880              | 0.0000       |
| 6.0972              | 0.0000       |
| 6.7500              | 0.0000       |
| 7.4478              | 0.0000       |
| 8.1920              | 0.0000       |
| 8.9842              | 0.0000       |
| 9.8260              | 0.0000       |
| 10.7188             | 0.0000       |
| 11.6640             | 0.0000       |
| 12.6633             | 0.0000       |
| 13.7180             | 0.0000       |
| 14.8297             | 0.0000       |
| 16.0000             | 0.0000       |
| 17.2302             | 0.0000       |
| 18.5220             | 0.0000       |
| 19.8767             | 0.0000       |
| 21.2960             | 0.0000       |
| 22.7812             | 0.0000       |
| 24.3340             | 0.0000       |
| 25.9558             | 0.0000       |
| 27.6480             | 0.0000       |
| 29.4123             | 0.0000       |
| 31.2500             | 0.0000       |
| 33.1627             | 0.0019       |
| 35.1520             | 0.0095       |
| 37.2192             | 0.0239       |
| 39.3660             | 0.0448       |
| 41.5938             | 0.0718       |
| 43.9040             | 0.0959       |
| 46.2983             | 0.1387       |
| 48.7780             | 0.2028       |
| 51.3448             | 0.3226       |
| 54.0000             | 0.4044       |
| 56.7452             | 0.4711       |
| 59.5820             | 0.5177       |
| 62.5117             | 0.5479       |
| 65.5360             | 0.5663       |
| 68.6562             | 0.5832       |
| 71.8740             | 0.6008       |
| 75.1908             | 0.6181       |
| 78.6080             | 0.6329       |
| 82.1273             | 0.6500       |
TABLE S23: Raw data for Pt as calculated with PBE in Castep.

| \( V_{\text{prim}} (\text{Å}^3) \) | CFR fraction |
|-------------------------------|--------------|
| 4.3940                        | 0.0000       |
| 4.9207                        | 0.0000       |
| 5.4880                        | 0.0000       |
| 6.0972                        | 0.0000       |
| 6.7500                        | 0.0000       |
| 7.4478                        | 0.0000       |
| 8.1920                        | 0.0000       |
| 8.9842                        | 0.0000       |
| 9.8260                        | 0.0000       |
| 10.7188                       | 0.0000       |
| 11.6640                       | 0.0000       |
| 12.6633                       | 0.0000       |
| 13.7180                       | 0.0000       |
| 14.8297                       | 0.0000       |
| 16.0000                       | 0.0000       |
| 17.2302                       | 0.0000       |
| 18.5220                       | 0.0000       |
| 19.8767                       | 0.0000       |
| 21.2960                       | 0.0000       |
| 22.7812                       | 0.0000       |
| 24.3340                       | 0.0000       |
| 25.9558                       | 0.0000       |
| 27.6480                       | 0.0040       |
| 29.4123                       | 0.0209       |
| 31.2500                       | 0.0402       |
| 33.1627                       | 0.0680       |
| 35.1520                       | 0.0870       |
| 37.2192                       | 0.1105       |
| 39.3660                       | 0.2034       |
| 41.5938                       | 0.2942       |
| 43.9040                       | 0.3549       |
| 46.2983                       | 0.4063       |
| 48.7780                       | 0.4611       |
| 51.3448                       | 0.4893       |
| 54.0000                       | 0.5179       |
| 56.7452                       | 0.5427       |
| 59.5820                       | 0.5593       |
| 62.5117                       | 0.5743       |
| 65.5360                       | 0.5953       |
| 68.6562                       | 0.6127       |
| 71.8740                       | 0.6310       |
| 75.1908                       | 0.6457       |
| 78.6080                       | 0.6598       |
| 82.1273                       | 0.6692       |
| 85.7500                       | 0.6799       |
| 89.4777                       | 0.6891       |
TABLE S24: Raw data for C as calculated with PBE in Castep.

| $V_{\text{prim}}$ (Å$^3$) | CFR fraction |
|---------------------------|--------------|
| 3.9062                    | 0.0000       |
| 4.3940                    | 0.0000       |
| 4.9208                    | 0.0000       |
| 5.4880                    | 0.0000       |
| 6.0972                    | 0.0000       |
| 6.7500                    | 0.0000       |
| 7.4478                    | 0.0000       |
| 8.1920                    | 0.0000       |
| 8.9842                    | 0.0000       |
| 9.8260                    | 0.0000       |
| 10.7188                   | 0.0000       |
| 11.6640                   | 0.0000       |
| 12.6633                   | 0.0000       |
| 13.7180                   | 0.0000       |
| 14.8297                   | 0.0000       |
| 16.0000                   | 0.0000       |
| 17.2302                   | 0.0000       |
| 18.5220                   | 0.0000       |
| 19.8767                   | 0.0000       |
| 21.2960                   | 0.0015       |
| 22.7812                   | 0.0270       |
| 24.3340                   | 0.0986       |
| 25.9558                   | 0.1420       |
| 27.6480                   | 0.1723       |
| 29.4122                   | 0.2062       |
| 31.2500                   | 0.2465       |
| 33.1628                   | 0.2781       |
| 35.1520                   | 0.3125       |
| 37.2192                   | 0.3388       |
| 39.3660                   | 0.3700       |
| 41.5938                   | 0.3923       |
| 43.9040                   | 0.4157       |
| 46.2982                   | 0.4402       |
| 48.7780                   | 0.4602       |
| 51.3448                   | 0.4800       |
| 54.0000                   | 0.4969       |
| 56.7452                   | 0.5169       |
| 59.5820                   | 0.5373       |
| 62.5118                   | 0.5536       |
| 65.5360                   | 0.5724       |
| 68.5662                   | 0.5900       |
| 71.8740                   | 0.6077       |
| 75.1908                   | 0.6258       |
| 78.0800                   | 0.6447       |
| 82.1272                   | 0.6581       |
| 85.7500                   | 0.6731       |
| 89.4778                   | 0.6911       |
| 93.3120                   | 0.7033       |
| 97.2542                   | 0.7137       |
| 101.3060                  | 0.7253       |
| 105.4688                  | 0.7362       |
| 109.7440                  | 0.7474       |
| 114.1332                  | 0.7576       |
| 118.6380                  | 0.7664       |
| 123.2598                  | 0.7776       |
| 128.0000                  | 0.7847       |
| 132.8602                  | 0.7922       |
| 137.8420                  | 0.8000       |
| 142.9468                  | 0.8063       |
| ...                       | ...          |
TABLE S25: Raw data for Ne as calculated with PBE in Castep.

| $V_{\text{prim}}$ (Å³) | CFR fraction |
|-------------------------|--------------|
| 3.9062                  | 0.0000       |
| 4.3940                  | 0.0000       |
| 4.9207                  | 0.0000       |
| 5.4880                  | 0.0359       |
| 6.0972                  | 0.1041       |
| 6.7500                  | 0.2904       |
| 7.4478                  | 0.4156       |
| 8.1920                  | 0.4829       |
| 8.9842                  | 0.5534       |
| 9.8260                  | 0.6158       |
| 10.7188                 | 0.6594       |
| 11.6640                 | 0.6909       |
| 12.6633                 | 0.7259       |
| 13.7180                 | 0.7556       |
| 14.8297                 | 0.7745       |
| 16.0000                 | 0.7939       |
| 17.2302                 | 0.8111       |
| 18.5220                 | 0.8253       |
| 19.8767                 | 0.8388       |
| 21.2960                 | 0.8494       |
| 22.7812                 | 0.8594       |
| 24.3340                 | 0.8702       |
| 25.9558                 | 0.8772       |
| 27.6480                 | 0.8868       |
| 29.4123                 | 0.8949       |
| 31.2500                 | 0.9007       |
| 33.1627                 | 0.9049       |
| 35.1520                 | 0.9111       |
| 37.2192                 | 0.9157       |
| 39.3660                 | 0.9197       |
| 41.5938                 | 0.9249       |
| 43.9040                 | 0.9280       |
| 46.2983                 | 0.9307       |
| 48.7780                 | 0.9364       |
| 51.3448                 | 0.9396       |
| 54.0000                 | 0.9422       |
| 56.7452                 | 0.9436       |
| 59.5820                 | 0.9470       |
| 62.5117                 | 0.9497       |
| 65.5360                 | 0.9525       |
| 68.6562                 | 0.9535       |
| 71.8740                 | 0.9565       |
| 75.1908                 | 0.9583       |
| 78.6080                 | 0.9599       |
| 82.1273                 | 0.9621       |
| 85.7500                 | 0.9634       |
| 89.4777                 | 0.9650       |
| 93.3120                 | 0.9661       |
| 97.2542                 | 0.9682       |
| 101.3060                | 0.9694       |
| 105.4690                | 0.9704       |
| 109.7440                | 0.9725       |
| 118.6380                | 0.9737       |
| 123.2600                | 0.9746       |
| 128.0000                | 0.9755       |
| 132.8600                | 0.9762       |
| 137.8420                | 0.9773       |
| 142.9470                | 0.9785       |
| 148.1760                | 0.9787       |

| $V_{\text{prim}}$ (Å³) | CFR fraction |
|-------------------------|--------------|
| 153.5310                | 0.9795       |
| 159.0140                | 0.9802       |
| 164.6260                | 0.9812       |
TABLE S26: Raw data for NaCl as calculated with PBE in Castep.

| $V_{\text{prim}}$ (Å$^3$) | CFR fraction |
|---------------------------|--------------|
| 4.3940                    | 0.0000       |
| 4.9208                    | 0.0000       |
| 5.4880                    | 0.0000       |
| 6.0972                    | 0.0000       |
| 6.7500                    | 0.0000       |
| 7.4478                    | 0.0000       |
| 8.1920                    | 0.0000       |
| 8.9842                    | 0.0000       |
| 9.8260                    | 0.0000       |
| 10.7188                   | 0.0000       |
| 11.6640                   | 0.0000       |
| 12.6633                   | 0.0000       |
| 13.7180                   | 0.0000       |
| 14.8297                   | 0.0000       |
| 16.0000                   | 0.0000       |
| 17.2302                   | 0.0000       |
| 18.5220                   | 0.0000       |
| 19.8767                   | 0.0000       |
| 21.2960                   | 0.0000       |
| 22.7812                   | 0.0000       |
| 24.3340                   | 0.0000       |
| 25.9558                   | 0.0000       |
| 27.6480                   | 0.0009       |
| 29.4122                   | 0.0171       |
| 31.2500                   | 0.0497       |
| 33.1628                   | 0.0822       |
| 35.1520                   | 0.1270       |
| 37.2192                   | 0.1784       |
| 39.3660                   | 0.2227       |
| 41.5938                   | 0.2703       |
| 43.9040                   | 0.3135       |
| 46.2982                   | 0.3475       |
| 48.7780                   | 0.3835       |
| 51.3448                   | 0.4188       |
| 54.0000                   | 0.4515       |
| 56.7452                   | 0.4743       |
| 59.5820                   | 0.5021       |
| 62.5118                   | 0.5274       |
| 65.5360                   | 0.5488       |
| 68.6562                   | 0.5751       |
| 71.8740                   | 0.5927       |
| 75.1908                   | 0.6137       |
| 78.6080                   | 0.6318       |
| 82.1272                   | 0.6503       |
| 85.7500                   | 0.6638       |
| 89.4778                   | 0.6783       |
| 93.3120                   | 0.6913       |
| 97.2542                   | 0.7011       |
| 101.3060                  | 0.7092       |
| 105.4688                  | 0.7202       |
| 109.7440                  | 0.7289       |
| 114.1332                  | 0.7364       |
| 118.6380                  | 0.7433       |
| 123.2598                  | 0.7509       |
| 128.0000                  | 0.7559       |
| 132.8602                  | 0.7608       |
| 137.8420                  | 0.7687       |
| 142.9468                  | 0.7736       |
| 148.1760                  | 0.7785       |

\[ \ldots \]

\[ \ldots \]
| $V_{\text{prim}}$ (Å$^3$) | CFR fraction |
|--------------------------|--------------|
| 3.9062                   | 0.0000       |
| 4.3940                   | 0.0000       |
| 4.9208                   | 0.0000       |
| 5.4880                   | 0.0000       |
| 6.0972                   | 0.0000       |
| 6.7500                   | 0.0000       |
| 7.4478                   | 0.0000       |
| 8.1920                   | 0.0000       |
| 8.9842                   | 0.0000       |
| 9.8260                   | 0.0000       |
| 10.7188                  | 0.0000       |
| 11.6640                  | 0.0000       |
| 12.6633                  | 0.0000       |
| 13.7180                  | 0.0000       |
| 14.8297                  | 0.0000       |
| 16.0000                  | 0.0000       |
| 17.2302                  | 0.0000       |
| 18.5220                  | 0.0000       |
| 19.8767                  | 0.0000       |
| 21.2960                  | 0.0000       |
| 22.7812                  | 0.0000       |
| 24.3340                  | 0.0000       |
| 25.9558                  | 0.0000       |
| 27.6480                  | 0.0000       |
| 29.4122                  | 0.0000       |
| 31.2500                  | 0.0000       |
| 33.1628                  | 0.0000       |
| 35.1520                  | 0.0000       |
| 37.2192                  | 0.0000       |
| 39.3660                  | 0.0000       |
| 41.5938                  | 0.0000       |
| 43.9040                  | 0.0000       |
| 46.2982                  | 0.0020       |
| 48.7780                  | 0.0093       |
| 51.3448                  | 0.0176       |
| 54.0000                  | 0.0208       |
| 56.7452                  | 0.0614       |
| 59.5820                  | 0.1012       |
| 62.5118                  | 0.1289       |
| 65.5360                  | 0.1677       |
| 68.6562                  | 0.1903       |
| 71.8740                  | 0.2162       |
| 75.1908                  | 0.2368       |
| 78.6080                  | 0.2632       |
| 82.1272                  | 0.2843       |
| 85.7500                  | 0.2955       |
| 89.4778                  | 0.3277       |
| 93.3120                  | 0.3444       |
| 97.2542                  | 0.3609       |
| 101.3060                 | 0.3779       |
| 105.4688                 | 0.3959       |
| 109.7440                 | 0.4042       |
| 114.1332                 | 0.4236       |
| 118.6380                 | 0.4381       |
| 123.2598                 | 0.4504       |
| 128.0000                 | 0.4648       |
| 132.8602                 | 0.4792       |
| 137.8420                 | 0.4887       |
| 142.9468                 | 0.5017       |

...
### S2. RAW DATA FOR GROUP 1 ELEMENTAL SOLIDS

TABLE S28: Raw data for Li as calculated with PBE in Castep.

| $V_{\text{prim}}$ (Å³) | CFR fraction |
|------------------------|--------------|
| 0.0000                 | 0.0000       |
| 4.6305                 | 0.0000       |
| 5.3240                 | 0.0000       |
| 6.0835                 | 0.0000       |
| 6.9120                 | 0.0000       |
| 7.8125                 | 0.0000       |
| 8.7880                 | 0.0000       |
| 9.8415                 | 0.0000       |
| 10.9760                | 0.0000       |
| 12.1945                | 0.0000       |
| 13.5000                | 0.0000       |
| 14.8955                | 0.0000       |
| 16.3840                | 0.0000       |
| 17.9685                | 0.0000       |
| 19.6520                | 0.0000       |
| 21.4375                | 0.0000       |
| 23.3280                | 0.0000       |
| 25.3265                | 0.0000       |
| 27.4360                | 0.0000       |
| 29.6595                | 0.0000       |
| 32.0000                | 0.0000       |
| 34.4605                | 0.0000       |
| 37.0440                | 0.0000       |
| 39.7535                | 0.0000       |
| 42.5920                | 0.0000       |
| 45.5625                | 0.0000       |
| 48.6680                | 0.0000       |
| 51.9115                | 0.0000       |
| 55.2960                | 0.0000       |
| 58.8245                | 0.0000       |
| 62.5000                | 0.0000       |
| 66.3255                | 0.0000       |
| 70.3040                | 0.0000       |
| 74.4385                | 0.0000       |
| 78.7320                | 0.0000       |
| 83.1875                | 0.0000       |
| 87.8080                | 0.0000       |
| 92.5965                | 0.0000       |
| 97.5560                | 0.0000       |
| 102.6900               | 0.0000       |
| 108.0000               | 0.0000       |
| 113.4900               | 0.0000       |
| 119.1640               | 0.0028       |
| 125.0230               | 0.0671       |
| 131.0720               | 0.1410       |
| 137.3120               | 0.2031       |
| 143.7480               | 0.2503       |
| 150.3820               | 0.2630       |
| 157.2160               | 0.2723       |
| 164.2550               | 0.3022       |
| 171.5000               | 0.3476       |
| 178.9550               | 0.4014       |
| 186.6240               | 0.4430       |
| 194.5080               | 0.4774       |
| 202.6120               | 0.5276       |
| 219.4880               | 0.5466       |

...
| $V_{\text{prin}}$ (Å$^3$) | CFR fraction |
|-------------------------|--------------|
|                         |              |
| 228.2670                | 0.5659       |
| 237.2760                | 0.5813       |
| 246.5200                | 0.5970       |
| 256.0000                | 0.6112       |
| 265.7200                | 0.6282       |
| 275.6840                | 0.6445       |
| 285.8940                | 0.6634       |
| 296.3520                | 0.6932       |
| 318.0280                | 0.7038       |
| 329.2510                | 0.7128       |
| 340.7360                | 0.7276       |
| 352.4850                | 0.7322       |
| 364.5000                | 0.7415       |
| 376.7850                | 0.7568       |
| 389.3440                | 0.7724       |
| 402.1790                | 0.7798       |
| 415.2920                | 0.7893       |
| 428.6880                | 0.8095       |
| 456.3360                | 0.8159       |
| 470.5960                | 0.8223       |
| 485.1500                | 0.8233       |
TABLE S29: Raw data for Na as calculated with PBE in Castep.

| $V_{\text{prim}}$ (Å$^3$) | CFR fraction |
|--------------------------|--------------|
| 4.0000                   | 0.0000       |
| 4.6305                   | 0.0000       |
| 5.3240                   | 0.0000       |
| 6.0835                   | 0.0000       |
| 6.9120                   | 0.0000       |
| 7.8125                   | 0.0000       |
| 8.7880                   | 0.0000       |
| 9.8415                   | 0.0000       |
| 10.9760                  | 0.0000       |
| 12.1945                  | 0.0000       |
| 13.5000                  | 0.0000       |
| 14.8955                  | 0.0000       |
| 16.3840                  | 0.0000       |
| 17.9685                  | 0.0000       |
| 19.6520                  | 0.0000       |
| 21.4375                  | 0.0000       |
| 23.3280                  | 0.0000       |
| 25.3265                  | 0.0000       |
| 27.4360                  | 0.0000       |
| 29.6595                  | 0.0000       |
| 32.0000                  | 0.0000       |
| 34.4605                  | 0.0000       |
| 37.0440                  | 0.0000       |
| 39.7535                  | 0.0000       |
| 42.5920                  | 0.0000       |
| 45.5625                  | 0.0000       |
| 48.6680                  | 0.0000       |
| 51.9115                  | 0.0000       |
| 55.2960                  | 0.0000       |
| 58.8245                  | 0.0000       |
| 62.5000                  | 0.0000       |
| 66.3255                  | 0.0000       |
| 70.3040                  | 0.0000       |
| 74.4385                  | 0.0000       |
| 78.7320                  | 0.0000       |
| 83.1875                  | 0.0000       |
| 87.5080                  | 0.0000       |
| 92.5965                  | 0.0000       |
| 97.5560                  | 0.0000       |
| 102.6900                 | 0.0000       |
| 108.0000                 | 0.0000       |
| 113.4900                 | 0.0000       |
| 119.1640                 | 0.0000       |
| 125.0230                 | 0.0000       |
| 131.0720                 | 0.0000       |
| 137.3120                 | 0.0000       |
| 143.7480                 | 0.0213       |
| 150.3820                 | 0.1049       |
| 157.2160                 | 0.1723       |
| 164.2550                 | 0.2389       |
| 171.5000                 | 0.2721       |
| 178.9550                 | 0.2818       |
| 186.6240                 | 0.2863       |
| 194.5080                 | 0.3044       |
| 202.6120                 | 0.3421       |
| 210.9380                 | 0.5110       |
| 256.0000                 | 0.5337       |
| 265.7200                 | 0.5655       |
| 285.8940                 | 0.6007       |
| ...                      | ...          |
| \(V_{\text{prim}} \text{ (Å}^3\) | CFR fraction |
|-----------------|-------------|
| :               | :           |
| 307.0620        | 0.6113      |
| 318.0280        | 0.6367      |
| 329.2510        | 0.6505      |
| 340.7360        | 0.6633      |
| 352.4850        | 0.6988      |
| 389.3440        | 0.7090      |
| 402.1790        | 0.7214      |
| 415.2920        | 0.7418      |
| 428.6880        | 0.7472      |
| 442.3680        | 0.7594      |
| 456.3360        | 0.7716      |
| 470.5960        | 0.7718      |
| 485.1500        | 0.7774      |
TABLE S30: Raw data for K as calculated with PBE in Castep.

| V_{prim} (Å³) | CFR fraction |
|---------------|--------------|
| 105.2869      | 0.0000       |
| 111.6025      | 0.0000       |
| 118.1657      | 0.0000       |
| 124.9814      | 0.0000       |
| 132.0541      | 0.0000       |
| 139.3888      | 0.0000       |
| 146.9902      | 0.0000       |
| 154.8631      | 0.0000       |
| 163.0121      | 0.0000       |
| 171.4422      | 0.0000       |
| 180.1579      | 0.0000       |
| 189.1642      | 0.0000       |
| 198.4658      | 0.0000       |
| 208.0673      | 0.0000       |
| 217.9736      | 0.0000       |
| 228.1895      | 0.0000       |
| 238.7197      | 0.0622       |
| 249.5690      | 0.1122       |
| 260.7421      | 0.1531       |
| 272.2438      | 0.1898       |
| 284.0789      | 0.2255       |
| 296.2521      | 0.2532       |
| 308.7682      | 0.2817       |
| 321.6319      | 0.3086       |
| 334.8480      | 0.3344       |
| 348.4213      | 0.3583       |
| 362.3565      | 0.3829       |
| 376.6585      | 0.4054       |
| 391.3318      | 0.4285       |
| 406.3815      | 0.4504       |
| 421.8120      | 0.4719       |
| 437.6284      | 0.4929       |
| 453.8352      | 0.5129       |
| 470.4373      | 0.5340       |
| 487.3995      | 0.5500       |
| 509.4292      | 0.6363       |
| 570.1397      | 0.6772       |
| 793.7138      | 0.6835       |
| 817.7633      | 0.6855       |
| 842.2970      | 0.7017       |
| 867.3149      | 0.7095       |
| 892.8169      | 0.7095       |
| 918.8189      | 0.7157       |
| 945.3290      | 0.7285       |
| 972.3311      | 0.7390       |
| 999.8492      | 0.7412       |
| 1027.8832     | 0.7569       |
| 1056.4331     | 0.7644       |
| 1085.5068     | 0.7710       |
| 1115.1123     | 0.7745       |
| 1145.2496     | 0.7922       |
| 1175.9187     | 0.8009       |
| 1207.1434     | 0.8080       |
| 1238.9078     | 0.8101       |
| 1271.2199     | 0.8179       |
| 1304.0956     | 0.8242       |
| 1337.5347     | 0.8292       |
| 1371.5374     | 0.8311       |
| 1406.1116     | 0.8431       |

...
| $V_{\text{prim}}$ (Å$^3$) | CFR fraction |
|---------------------------|--------------|
| 1441.2652                 | 0.8518       |
| 1476.9982                 | 0.8590       |
| 1513.3106                 | 0.8556       |
| 1550.2183                 | 0.8584       |
| 1587.7292                 | 0.8640       |
| 1625.8274                 | 0.8656       |
| 1664.5369                 | 0.8714       |
| 1703.8574                 | 0.8759       |
| 1743.7892                 | 0.8794       |
| 1784.3400                 | 0.8840       |
| 1825.5179                 | 0.8892       |
| 1867.3228                 | 0.8922       |
| 1909.7547                 | 0.8953       |
| 1952.8375                 | 0.8986       |
| 1996.5553                 | 0.9025       |
| 2040.9159                 | 0.9060       |
| 2085.9354                 | 0.9085       |
| 2131.6136                 | 0.9111       |
| 2177.9506                 | 0.9105       |
| 2224.9544                 | 0.9137       |
| 2272.6327                 | 0.9137       |
| 2320.9858                 | 0.9167       |
| 2370.0135                 | 0.9162       |
| 2419.7317                 | 0.9212       |
| 2470.1484                 | 0.9237       |
| 2521.2477                 | 0.9241       |
| 2573.0534                 | 0.9263       |
| 2625.5655                 | 0.9276       |
| 2678.7840                 | 0.9298       |
TABLE S31: Raw data for Rb as calculated with PBE in Castep.

| \( V_{\text{prim}} \) (\( \text{Å}^3 \)) | CFR fraction |
|----------------------------------------|--------------|
| 105.2869                               | 0.0000       |
| 111.6025                               | 0.0000       |
| 118.1657                               | 0.0000       |
| 124.9814                               | 0.0000       |
| 132.0541                               | 0.0000       |
| 139.3888                               | 0.0000       |
| 146.9902                               | 0.0000       |
| 154.8631                               | 0.0000       |
| 163.0121                               | 0.0000       |
| 171.4422                               | 0.0000       |
| 180.1579                               | 0.0000       |
| 189.1642                               | 0.0000       |
| 198.4658                               | 0.0000       |
| 208.0673                               | 0.0000       |
| 217.9736                               | 0.0000       |
| 228.1895                               | 0.0000       |
| 238.7197                               | 0.0000       |
| 249.5690                               | 0.0000       |
| 260.7421                               | 0.0000       |
| 272.2438                               | 0.0190       |
| 284.0789                               | 0.0771       |
| 296.2521                               | 0.1214       |
| 308.7682                               | 0.1607       |
| 321.6319                               | 0.1955       |
| 334.8480                               | 0.2260       |
| 348.4213                               | 0.2558       |
| 362.3565                               | 0.2826       |
| 376.6585                               | 0.3085       |
| 391.3318                               | 0.3340       |
| 406.3815                               | 0.3567       |
| 421.8120                               | 0.3803       |
| 437.6284                               | 0.4028       |
| 453.8352                               | 0.4248       |
| 470.4373                               | 0.4451       |
| 487.4395                               | 0.4667       |
| 504.8464                               | 0.4871       |
| 522.6629                               | 0.5082       |
| 540.8937                               | 0.5279       |
| 559.5436                               | 0.5479       |
| 578.6173                               | 0.5648       |
| 770.1397                               | 0.6180       |
| 867.3149                               | 0.6668       |
| 918.8189                               | 0.6710       |
| 1085.5068                              | 0.7181       |
| 1271.2199                              | 0.7692       |
| 1304.0956                              | 0.7931       |
| 1337.5347                              | 0.8011       |
| 1371.5374                              | 0.7976       |
| 1441.2652                              | 0.8094       |
| 1513.3106                              | 0.8266       |
| 1550.2183                              | 0.8274       |
| 1587.7292                              | 0.8403       |
| 1625.8274                              | 0.8441       |
| 1664.5369                              | 0.8540       |
| 1703.8574                              | 0.8503       |
| 1743.7892                              | 0.8553       |
| 1784.3400                              | 0.8577       |
| 1825.5179                              | 0.8718       |
| 1867.3228                              | 0.8665       |
| ...                                    | ...          |
\[ V_{\text{prim}} (\AA^3) \quad \text{CFR fraction} \]

| \( V_{\text{prim}} (\AA^3) \) | CFR fraction |
|----------------------------------|--------------|
| 1909.7547                        | 0.8716       |
| 1952.8375                        | 0.8726       |
| 1996.5553                        | 0.8774       |
| 2040.9159                        | 0.8877       |
| 2085.9354                        | 0.8876       |
| 2131.6136                        | 0.8924       |
| 2177.9506                        | 0.8958       |
| 2224.9544                        | 0.8977       |
| 2272.6327                        | 0.9008       |
| 2320.9858                        | 0.8986       |
| 2370.0135                        | 0.9055       |
| 2419.7317                        | 0.9082       |
| 2470.1484                        | 0.9084       |
| 2521.2477                        | 0.9108       |
| 2573.0534                        | 0.9103       |
| 2625.5655                        | 0.9140       |
| 2678.7840                        | 0.9137       |
TABLE S32: Raw data for Cs as calculated with PBE in Castep.

| \( V_{\text{prim}} \) (\( \text{Å}^3 \)) | CFR fraction |
|----------------------|-------------|
| 105.2869             | 0.0000      |
| 111.6025             | 0.0000      |
| 118.1657             | 0.0000      |
| 124.9814             | 0.0000      |
| 132.0541             | 0.0000      |
| 139.3888             | 0.0000      |
| 146.9902             | 0.0000      |
| 154.8631             | 0.0000      |
| 163.0121             | 0.0000      |
| 171.4422             | 0.0000      |
| 180.1579             | 0.0000      |
| 189.1642             | 0.0000      |
| 198.4658             | 0.0000      |
| 208.0673             | 0.0000      |
| 217.9736             | 0.0000      |
| 228.1895             | 0.0000      |
| 238.7197             | 0.0000      |
| 249.5690             | 0.0000      |
| 260.7421             | 0.0000      |
| 272.2438             | 0.0000      |
| 284.0789             | 0.0000      |
| 296.2521             | 0.0000      |
| 308.7682             | 0.0000      |
| 321.6319             | 0.0000      |
| 334.8480             | 0.0008      |
| 348.4213             | 0.0133      |
| 362.3565             | 0.0649      |
| 376.6585             | 0.1087      |
| 391.3318             | 0.1452      |
| 421.8120             | 0.2068      |
| 437.6284             | 0.2343      |
| 453.8352             | 0.2509      |
| 470.4373             | 0.2852      |
| 487.4395             | 0.3100      |
| 504.8464             | 0.3316      |
| 522.6629             | 0.3536      |
| 540.9037             | 0.3743      |
| 559.5436             | 0.3944      |
| 578.6173             | 0.4154      |
| 598.1197             | 0.4353      |
| 618.0554             | 0.4558      |
| 638.4292             | 0.4744      |
| 659.2460             | 0.4849      |
| 680.5104             | 0.5138      |
| 702.2272             | 0.5325      |
| 747.0371             | 0.5648      |
| 770.1397             | 0.5818      |
| 793.7138             | 0.5968      |
| 817.7633             | 0.6089      |
| 842.2970             | 0.6208      |
### S3. RAW DATA FOR GROUP 2 ELEMENTAL SOLIDS

**TABLE S3:** Raw data for Be as calculated with PBE in Castep.

| \( V_{\text{prim}} \) (Å³) | CFR fraction |
|----------------------------|--------------|
| 4.0000                     | 0.0000       |
| 4.6305                     | 0.0000       |
| 5.3240                     | 0.0000       |
| 6.0835                     | 0.0000       |
| 6.9120                     | 0.0000       |
| 7.8125                     | 0.0000       |
| 8.7880                     | 0.0000       |
| 9.8415                     | 0.0000       |
| 10.9760                    | 0.0000       |
| 12.1945                    | 0.0000       |
| 13.5000                    | 0.0000       |
| 14.8955                    | 0.0000       |
| 16.3840                    | 0.0000       |
| 17.9685                    | 0.0000       |
| 19.6520                    | 0.0000       |
| 21.4375                    | 0.0000       |
| 23.3280                    | 0.0000       |
| 25.3265                    | 0.0000       |
| 27.4360                    | 0.0000       |
| 29.6595                    | 0.0000       |
| 32.0000                    | 0.0000       |
| 34.4605                    | 0.0000       |
| 37.0440                    | 0.0000       |
| 39.7535                    | 0.0000       |
| 42.5920                    | 0.0000       |
| 45.5625                    | 0.0000       |
| 48.6680                    | 0.0860       |
| 51.9115                    | 0.2329       |
| 55.2960                    | 0.2980       |
| 58.8245                    | 0.3521       |
| 62.5000                    | 0.4029       |
| 66.3255                    | 0.4482       |
| 70.3040                    | 0.4933       |
| 74.4385                    | 0.5267       |
| 78.7320                    | 0.5535       |
| 83.1875                    | 0.5755       |
| 87.8080                    | 0.5978       |
| 92.5965                    | 0.6185       |
| 97.5560                    | 0.6427       |
| 102.6900                   | 0.6675       |
| 108.0000                   | 0.6886       |
| 113.4900                   | 0.7072       |
| 119.1640                   | 0.7267       |
| 125.0230                   | 0.7430       |
| 131.0720                   | 0.7564       |
| 137.3120                   | 0.7698       |
| 143.7480                   | 0.7806       |
| 150.3820                   | 0.7926       |
| 157.2160                   | 0.8026       |
| 164.2550                   | 0.8122       |
| 171.5000                   | 0.8210       |
| 178.9550                   | 0.8288       |
| 186.6240                   | 0.8356       |
| 194.5080                   | 0.8433       |
| 202.6120                   | 0.8497       |
| 210.9380                   | 0.8560       |
| 219.4880                   | 0.8542       |

| \( V_{\text{prim}} \) (Å³) | CFR fraction |
|----------------------------|--------------|
| 228.2670                   | 0.8597       |
| 237.2760                   | 0.8663       |
| 246.5200                   | 0.8721       |
| 256.0000                   | 0.8779       |
| 265.7200                   | 0.8827       |
| 275.6840                   | 0.8883       |
| 285.8940                   | 0.8930       |
| 296.3520                   | 0.8977       |
| 307.0620                   | 0.9024       |
| 318.0280                   | 0.9066       |
| 329.2510                   | 0.9108       |
| 340.7360                   | 0.9149       |
| 352.4850                   | 0.9192       |
| 364.5000                   | 0.9227       |
| 376.7850                   | 0.9265       |
| 389.3440                   | 0.9295       |
TABLE S34: Raw data for Mg as calculated with PBE in Castep.

| $V_{\text{prim}}$ (Å$^3$) | CFR fraction |
|-----------------------------|--------------|
| 4.0000                      | 0.0000       |
| 4.6305                      | 0.0000       |
| 5.3240                      | 0.0000       |
| 6.0835                      | 0.0000       |
| 6.9120                      | 0.0000       |
| 7.8125                      | 0.0000       |
| 8.7880                      | 0.0000       |
| 9.8415                      | 0.0000       |
| 10.9760                     | 0.0000       |
| 12.1945                     | 0.0000       |
| 13.5000                     | 0.0000       |
| 14.8955                     | 0.0000       |
| 16.3840                     | 0.0000       |
| 17.9685                     | 0.0000       |
| 19.6520                     | 0.0000       |
| 21.4375                     | 0.0000       |
| 23.3280                     | 0.0000       |
| 25.3265                     | 0.0000       |
| 27.4360                     | 0.0000       |
| 29.6595                     | 0.0000       |
| 32.0000                     | 0.0000       |
| 34.4605                     | 0.0000       |
| 37.0440                     | 0.0000       |
| 39.7535                     | 0.0000       |
| 42.5920                     | 0.0000       |
| 45.5625                     | 0.0000       |
| 48.6680                     | 0.0000       |
| 51.9115                     | 0.0000       |
| 55.2960                     | 0.0000       |
| 58.8245                     | 0.0000       |
| 62.5000                     | 0.0000       |
| 66.3255                     | 0.0000       |
| 70.3040                     | 0.0000       |
| 74.4385                     | 0.0000       |
| 78.7320                     | 0.0333       |
| 83.1875                     | 0.0645       |
| 87.5080                     | 0.1366       |
| 92.9665                     | 0.2134       |
| 97.5560                     | 0.2733       |
| 102.6900                    | 0.3216       |
| 108.0000                    | 0.3615       |
| 113.4900                    | 0.3928       |
| 119.1640                    | 0.4301       |
| 125.0230                    | 0.4440       |
| 131.0720                    | 0.4952       |
| 137.3120                    | 0.5106       |
| 143.7480                    | 0.5413       |
| 150.3820                    | 0.5516       |
| 157.2160                    | 0.5772       |
| 164.2550                    | 0.5913       |
| 171.5000                    | 0.6013       |
| 178.9550                    | 0.6086       |
| 186.6240                    | 0.6263       |
| 194.5080                    | 0.6325       |
| 202.6120                    | 0.6466       |
| 210.9380                    | 0.6612       |
| 219.4880                    | 0.6639       |
| 228.2670                    | 0.6674       |
| 237.2760                    | 0.6679       |

$V_{\text{prim}}$ (Å$^3$)  | CFR fraction |
|-----------------------------|--------------|
| 246.5200                    | 0.6812       |
| 256.0000                    | 0.6933       |
| 265.7200                    | 0.7053       |
| 275.6840                    | 0.7097       |
| 285.8940                    | 0.7221       |
| 296.3520                    | 0.7239       |
| 307.0620                    | 0.7369       |
| 318.0280                    | 0.7439       |
| 329.2510                    | 0.7500       |
| 340.7360                    | 0.7523       |
| 352.4850                    | 0.7574       |
| 364.5000                    | 0.7656       |
| 376.7850                    | 0.7703       |
| 389.3440                    | 0.7772       |
| 402.1790                    | 0.7812       |
| 415.2920                    | 0.7853       |
| 428.6880                    | 0.7904       |
| 442.3680                    | 0.7996       |
| 456.3360                    | 0.8035       |
| 470.5960                    | 0.8085       |
| 485.1500                    | 0.8142       |
TABLE S35: Raw data for Ca as calculated with PBE in Castep.

| $V_{\text{prim}}$ (Å$^3$) | CFR fraction |
|--------------------------|--------------|
| 4.0000                   | 0.0000       |
| 4.6305                   | 0.0000       |
| 5.3240                   | 0.0000       |
| 6.0835                   | 0.0000       |
| 6.9120                   | 0.0000       |
| 7.8125                   | 0.0000       |
| 8.7880                   | 0.0000       |
| 9.8415                   | 0.0000       |
| 10.9760                  | 0.0000       |
| 12.1945                  | 0.0000       |
| 13.5000                  | 0.0000       |
| 14.8955                  | 0.0000       |
| 16.3840                  | 0.0000       |
| 17.9685                  | 0.0000       |
| 19.6520                  | 0.0000       |
| 21.4375                  | 0.0000       |
| 23.3280                  | 0.0000       |
| 25.3265                  | 0.0000       |
| 27.4360                  | 0.0000       |
| 29.6595                  | 0.0000       |
| 32.0000                  | 0.0000       |
| 34.4605                  | 0.0000       |
| 37.0440                  | 0.0000       |
| 39.7535                  | 0.0000       |
| 42.5920                  | 0.0000       |
| 45.5625                  | 0.0000       |
| 48.6680                  | 0.0000       |
| 51.9115                  | 0.0000       |
| 55.2960                  | 0.0000       |
| 58.8245                  | 0.0000       |
| 62.5000                  | 0.0000       |
| 66.3255                  | 0.0000       |
| 70.3040                  | 0.0000       |
| 74.4385                  | 0.0000       |
| 78.7320                  | 0.0000       |
| 83.1875                  | 0.0000       |
| 87.5080                  | 0.0000       |
| 92.5965                  | 0.0000       |
| 97.5560                  | 0.0000       |
| 102.6900                 | 0.0000       |
| 108.0000                 | 0.0000       |
| 113.4900                 | 0.0000       |
| 119.1640                 | 0.0000       |
| 125.0230                 | 0.0000       |
| 131.0720                 | 0.0000       |
| 137.3120                 | 0.0000       |
| 143.7480                 | 0.0000       |
| 150.3820                 | 0.0000       |
| 157.2160                 | 0.0683       |
| 164.2550                 | 0.1777       |
| 171.5000                 | 0.2383       |
| 178.9550                 | 0.2798       |
| 186.6240                 | 0.3148       |
| 194.5080                 | 0.3443       |
| 202.6120                 | 0.3760       |
| 210.9380                 | 0.4071       |
| 219.4880                 | 0.4406       |
| 228.2670                 | 0.4711       |
| 237.2760                 | 0.5008       |
| ...                      | ...          |
| ...                      | ...          |
| 246.5200                 | 0.5208       |
| 256.0000                 | 0.5415       |
| 265.7200                 | 0.5598       |
| 275.6840                 | 0.5734       |
| 285.8940                 | 0.5889       |
| 296.3520                 | 0.6042       |
| 307.0620                 | 0.6192       |
| 318.0280                 | 0.6352       |
| 329.2510                 | 0.6467       |
| 340.7360                 | 0.6605       |
| 352.4850                 | 0.6721       |
| 364.5000                 | 0.6840       |
| 376.7850                 | 0.6948       |
| 389.3440                 | 0.7056       |
| 402.1790                 | 0.7151       |
| 415.2920                 | 0.7242       |
| 428.6880                 | 0.7330       |
| 442.3680                 | 0.7432       |
| 456.3360                 | 0.7499       |
| 470.5960                 | 0.7589       |
| 485.1500                 | 0.7653       |
TABLE S36: Raw data for Sr as calculated with PBE in Castep.

| $V_{\text{prim}}$ (Å$^3$) | CFR fraction |
|---------------------------|--------------|
| 4.0000                    | 0.0000       |
| 4.6305                    | 0.0000       |
| 5.3240                    | 0.0000       |
| 6.0835                    | 0.0000       |
| 6.9120                    | 0.0000       |
| 7.8125                    | 0.0000       |
| 8.7880                    | 0.0000       |
| 9.8415                    | 0.0000       |
| 10.9760                   | 0.0000       |
| 12.1945                   | 0.0000       |
| 13.5000                   | 0.0000       |
| 14.8955                   | 0.0000       |
| 16.3840                   | 0.0000       |
| 17.9685                   | 0.0000       |
| 19.6520                   | 0.0000       |
| 21.4375                   | 0.0000       |
| 23.3280                   | 0.0000       |
| 25.3265                   | 0.0000       |
| 27.4360                   | 0.0000       |
| 29.6595                   | 0.0000       |
| 32.0000                   | 0.0000       |
| 34.4605                   | 0.0000       |
| 37.0440                   | 0.0000       |
| 39.7535                   | 0.0000       |
| 42.5920                   | 0.0000       |
| 45.5625                   | 0.0000       |
| 48.6680                   | 0.0000       |
| 51.9115                   | 0.0000       |
| 55.2960                   | 0.0000       |
| 58.8245                   | 0.0000       |
| 62.5000                   | 0.0000       |
| 66.3255                   | 0.0000       |
| 70.3040                   | 0.0000       |
| 74.4385                   | 0.0000       |
| 78.7320                   | 0.0000       |
| 83.1875                   | 0.0000       |
| 87.5080                   | 0.0000       |
| 92.5965                   | 0.0000       |
| 97.5560                   | 0.0000       |
| 102.6900                  | 0.0000       |
| 108.0000                  | 0.0000       |
| 113.4900                  | 0.0000       |
| 119.1640                  | 0.0000       |
| 125.0230                  | 0.0000       |
| 131.0720                  | 0.0000       |
| 137.3120                  | 0.0011       |
| 143.7480                  | 0.0000       |
| 150.3820                  | 0.0018       |
| 157.2160                  | 0.0048       |
| 164.2550                  | 0.0066       |
| 171.5000                  | 0.0000       |
| 178.9550                  | 0.0062       |
| 186.6240                  | 0.0206       |
| 194.5080                  | 0.0808       |
| 202.6120                  | 0.1777       |
| 210.9380                  | 0.2240       |
| 219.4880                  | 0.2693       |
| 228.2670                  | 0.3002       |
| 237.2760                  | 0.3264       |
| 246.5200                  | 0.3608       |
| 256.0000                  | 0.3901       |
| 265.7200                  | 0.4192       |
| 275.6840                  | 0.4482       |
| 285.8940                  | 0.4711       |
| 296.3520                  | 0.4913       |
| 307.0620                  | 0.5125       |
| 318.0280                  | 0.5294       |
| 329.2510                  | 0.5507       |
| 340.7360                  | 0.5620       |
| 352.4850                  | 0.5799       |
| 364.5000                  | 0.5946       |
| 376.7850                  | 0.6059       |
| 389.3440                  | 0.6144       |
| 402.1790                  | 0.6305       |
| 415.2920                  | 0.6404       |
| 428.6880                  | 0.6506       |
| 442.3680                  | 0.6616       |
| 456.3360                  | 0.6712       |
| 470.5960                  | 0.6820       |
| 485.1500                  | 0.6944       |
TABLE S37: Raw data for Ba as calculated with PBE in Castep.

| \( V'_{\text{prim}} (\text{Å}^3) \) | CFR fraction |
|---------------------------------|-------------|
| 4.0000  | 0.0000  |
| 4.6305  | 0.0000  |
| 5.3240  | 0.0000  |
| 6.0835  | 0.0000  |
| 6.9120  | 0.0000  |
| 7.8125  | 0.0000  |
| 8.7880  | 0.0000  |
| 9.8415  | 0.0000  |
| 10.9760 | 0.0000  |
| 12.1945 | 0.0000  |
| 13.5000 | 0.0000  |
| 14.8955 | 0.0000  |
| 16.3840 | 0.0000  |
| 17.9685 | 0.0000  |
| 19.6520 | 0.0000  |
| 21.4375 | 0.0000  |
| 23.3280 | 0.0000  |
| 25.3265 | 0.0000  |
| 27.4360 | 0.0000  |
| 29.6595 | 0.0000  |
| 32.0000 | 0.0000  |
| 34.4605 | 0.0000  |
| 37.0440 | 0.0000  |
| 39.7535 | 0.0000  |
| 42.5920 | 0.0000  |
| 45.5625 | 0.0000  |
| 48.6680 | 0.0000  |
| 51.9115 | 0.0000  |
| 55.2960 | 0.0000  |
| 58.8245 | 0.0000  |
| 62.5000 | 0.0000  |
| 66.3255 | 0.0000  |
| 70.3040 | 0.0000  |
| 74.4385 | 0.0000  |
| 78.7320 | 0.0000  |
| 83.1875 | 0.0000  |
| 87.5080 | 0.0000  |
| 92.9065 | 0.0000  |
| 97.5560 | 0.0000  |
| 102.6900| 0.0000 |
| 108.0000| 0.0000 |
| 113.4900| 0.0000 |
| 119.1640| 0.0000 |
| 125.0230| 0.0000 |
| 131.0720| 0.0000 |
| 137.3120| 0.0000 |
| 143.7480| 0.0000 |
| 150.3820| 0.0000 |
| 157.2160| 0.0000 |
| 164.2550| 0.0000 |
| 171.5000| 0.0000 |
| 178.9550| 0.0000 |
| 186.6240| 0.0003 |
| 194.5080| 0.0000 |
| 202.6120| 0.0000 |
| 210.9380| 0.0039 |
| 219.4880| 0.0078 |
| 228.2670| 0.0108 |
| 237.2760| 0.0446 |

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| \( V'_{\text{prim}} (\text{Å}^3) \) | CFR fraction |
|---------------------------------|-------------|
| 246.5200 | 0.1016 |
| 256.0000 | 0.1691 |
| 265.7200 | 0.2206 |
| 275.6840 | 0.2636 |
| 285.8940 | 0.2989 |
| 296.3520 | 0.3304 |
| 307.0620 | 0.3591 |
| 318.0280 | 0.3852 |
| 329.2510 | 0.4142 |
| 340.7360 | 0.4424 |
| 352.4850 | 0.4714 |
| 364.5000 | 0.4992 |
| 376.7850 | 0.5192 |
| 389.3440 | 0.5400 |
| 402.1790 | 0.5546 |
| 415.2920 | 0.5706 |
| 428.6880 | 0.5844 |
| 442.3680 | 0.5989 |
| 456.3360 | 0.6108 |
| 470.5960 | 0.6220 |
| 485.1500 | 0.6377 |
**TABLE S38:** Raw data for Ra as calculated with PBE in Castep.

| $V_{\text{prim}}$ (Å³) | CFR fraction |
|-------------------------|--------------|
| 4.0000                  | 0.0000       |
| 4.6305                  | 0.0000       |
| 5.3240                  | 0.0000       |
| 6.0835                  | 0.0000       |
| 6.9120                  | 0.0000       |
| 7.8125                  | 0.0000       |
| 8.7880                  | 0.0000       |
| 9.8415                  | 0.0000       |
| 10.9760                 | 0.0000       |
| 12.1945                 | 0.0000       |
| 13.5000                 | 0.0000       |
| 14.8955                 | 0.0000       |
| 16.3840                 | 0.0000       |
| 17.9685                 | 0.0000       |
| 19.6520                 | 0.0000       |
| 21.4375                 | 0.0000       |
| 23.3280                 | 0.0000       |
| 25.3265                 | 0.0000       |
| 27.4360                 | 0.0000       |
| 29.6595                 | 0.0000       |
| 32.0000                 | 0.0000       |
| 34.4605                 | 0.0000       |
| 37.0440                 | 0.0000       |
| 39.7535                 | 0.0000       |
| 42.5920                 | 0.0000       |
| 45.5625                 | 0.0000       |
| 48.6680                 | 0.0000       |
| 51.9115                 | 0.0000       |
| 55.2960                 | 0.0000       |
| 58.8245                 | 0.0000       |
| 62.5000                 | 0.0000       |
| 66.3255                 | 0.0000       |
| 70.3040                 | 0.0000       |
| 74.4385                 | 0.0000       |
| 78.7320                 | 0.0000       |
| 83.1875                 | 0.0000       |
| 87.5080                 | 0.0000       |
| 92.9065                 | 0.0000       |
| 97.5560                 | 0.0000       |
| 102.6900                | 0.0000       |
| 108.0000                | 0.0000       |
| 113.4900                | 0.0000       |
| 119.1640                | 0.0000       |
| 125.9230                | 0.0000       |
| 131.0720                | 0.0000       |
| 137.3120                | 0.0000       |
| 143.7480                | 0.0000       |
| 150.3820                | 0.0000       |
| 157.2160                | 0.0000       |
| 164.2550                | 0.0000       |
| 171.5000                | 0.0000       |
| 178.9550                | 0.0082       |
| 186.6240                | 0.0082       |
| 194.5080                | 0.0083       |
| 202.6120                | 0.0079       |
| 210.9380                | 0.0079       |
| 219.4880                | 0.0146       |
| 228.2670                | 0.0126       |
| 237.2760                | 0.0200       |
| 246.5200                | 0.0444       |
| 256.0000                | 0.1211       |
| 265.7200                | 0.1902       |
| 275.6840                | 0.2390       |
| 285.8940                | 0.2735       |
| 296.3520                | 0.3009       |
| 307.0620                | 0.3213       |
| 318.0280                | 0.3456       |
| 329.2510                | 0.3732       |
| 340.7360                | 0.3998       |
| 352.4850                | 0.4278       |
| 364.5000                | 0.4507       |
| 376.7850                | 0.4713       |
| 389.3440                | 0.4923       |
| 402.1790                | 0.5110       |
| 415.2920                | 0.5273       |
| 428.6880                | 0.5430       |
| 442.3680                | 0.5553       |
| 456.3360                | 0.5716       |
| 470.5960                | 0.5816       |
| 485.1500                | 0.5942       |
S4. RAW DATA FOR GROUP 14 ELEMENTAL SOLIDS

TABLE S39: Raw data for C as calculated with PBE in VASP.

| $V_{prim}$ (Å$^3$) | CFR fraction | Band gap from DOS (eV) |
|---------------------|--------------|------------------------|
| 11.3748             | 0.0000       | 4.1395                 |
| 13.3957             | 0.0000       | 3.7061                 |
| 15.6427             | 0.0000       | 3.4592                 |
| 18.1279             | 0.0000       | 3.0474                 |
| 20.8634             | 0.0212       | 2.3060                 |
| 23.8610             | 0.1116       | 0.5534                 |
| 27.1328             | 0.1794       | 0.0000                 |
| 30.6909             | 0.2407       | 0.0000                 |
| 34.5471             | 0.3036       | 0.0000                 |
| 38.7135             | 0.3615       | 0.0000                 |
| 43.2022             | 0.4113       | 0.0000                 |
| 48.0250             | 0.4548       | 0.0000                 |
| 53.1940             | 0.4946       | 0.0000                 |
| 58.7213             | 0.5302       | 0.0000                 |
| 64.6187             | 0.5637       | 0.0000                 |
| 70.8983             | 0.5943       | 0.0000                 |
| 77.5722             | 0.6228       | 0.0000                 |
| 84.6522             | 0.6492       | 0.0000                 |
| 92.1505             | 0.6747       | 0.0000                 |
| 100.0789            | 0.6986       | 0.0000                 |
| 108.4495            | 0.7214       | 0.0000                 |
| 117.2744            | 0.7436       | 0.0000                 |
| 126.5654            | 0.7654       | 0.0000                 |
| 136.3346            | 0.7839       | 0.0000                 |
| 146.5941            | 0.7981       | 0.0000                 |
| 157.3557            | 0.8104       | 0.0000                 |
| 168.6315            | 0.8217       | 0.0000                 |
| 180.4336            | 0.8321       | 0.0000                 |
| 192.7738            | 0.8421       | 0.0000                 |
| 205.6642            | 0.8509       | 0.0000                 |
| 219.1169            | 0.8593       | 0.0000                 |
| 233.1437            | 0.8669       | 0.0000                 |
| 247.7567            | 0.8743       | 0.0000                 |
TABLE S40: Raw data for Si as calculated with PBE in VASP.

| $V_{prim}$ (Å$^3$) | CFR fraction | Band gap from DOS (eV) |
|---------------------|--------------|------------------------|
| 40.9168             | 0.0000       | 0.5507                 |
| 45.5711             | 0.0000       | 0.7547                 |
| 48.7857             | 0.0064       | 0.8074                 |
| 50.5655             | 0.0242       | 0.7107                 |
| 55.9121             | 0.0849       | 0.0000                 |
| 61.6230             | 0.1457       | 0.0000                 |
| 67.7100             | 0.1885       | 0.0000                 |
| 74.1852             | 0.2351       | 0.0000                 |
| 81.0607             | 0.2779       | 0.0000                 |
| 88.3483             | 0.3186       | 0.0000                 |
| 96.0601             | 0.3554       | 0.0000                 |
| 104.2082            | 0.3890       | 0.0000                 |
| 112.8044            | 0.4199       | 0.0000                 |
| 121.8609            | 0.4493       | 0.0000                 |
| 131.3895            | 0.4764       | 0.0000                 |
| 141.4023            | 0.5022       | 0.0000                 |
| 151.9114            | 0.5264       | 0.0000                 |
| 162.9286            | 0.5491       | 0.0000                 |
| 174.4660            | 0.5709       | 0.0000                 |
| 186.5357            | 0.5916       | 0.0000                 |
| 199.1495            | 0.6114       | 0.0000                 |
| 212.3195            | 0.6304       | 0.0000                 |
| 226.0578            | 0.6488       | 0.0000                 |
| 240.3762            | 0.6661       | 0.0000                 |
| 255.2868            | 0.6834       | 0.0000                 |
TABLE S41: Raw data for C as calculated with LSDA in VASP.

| \( V'_\text{ion} \) (\( \text{Å}^3 \)) | CFR fraction | Band gap from DOS (eV) |
|---------------------------------|--------------|------------------------|
| 10.9967                        | 0.0000       | 4.0588                 |
| 12.9738                        | 0.0000       | 3.7563                 |
| 15.1746                        | 0.0000       | 3.3818                 |
| 17.6112                        | 0.0000       | 3.0743                 |
| 20.2957                        | 0.0000       | 2.5228                 |
| 23.2399                        | 0.0097       | 1.0881                 |
| 26.4560                        | 0.0827       | 0.0000                 |
| 29.9558                        | 0.1643       | 0.0000                 |
| 33.7514                        | 0.2401       | 0.0000                 |
| 37.8549                        | 0.3093       | 0.0000                 |
| 42.2781                        | 0.3676       | 0.0000                 |
| 47.0331                        | 0.4171       | 0.0000                 |
| 52.1320                        | 0.4621       | 0.0000                 |
| 57.5866                        | 0.5012       | 0.0000                 |
| 63.4090                        | 0.5374       | 0.0000                 |
| 69.6113                        | 0.5703       | 0.0000                 |
| 76.2053                        | 0.6005       | 0.0000                 |
| 83.2031                        | 0.6294       | 0.0000                 |
| 90.6168                        | 0.6555       | 0.0000                 |
| 98.4582                        | 0.6803       | 0.0000                 |
| 106.7394                       | 0.7041       | 0.0000                 |
| 115.4725                       | 0.7258       | 0.0000                 |
| 124.6693                       | 0.7471       | 0.0000                 |
| 134.3419                       | 0.7665       | 0.0000                 |
| 144.5024                       | 0.7837       | 0.0000                 |
| 155.1626                       | 0.7981       | 0.0000                 |
| 166.3347                       | 0.8106       | 0.0000                 |
| 178.0305                       | 0.8224       | 0.0000                 |
| 190.2621                       | 0.8327       | 0.0000                 |
| 203.0416                       | 0.8422       | 0.0000                 |
| 216.3808                       | 0.8512       | 0.0000                 |
| 230.2918                       | 0.8591       | 0.0000                 |
| 244.7867                       | 0.8666       | 0.0000                 |
TABLE S42: Raw data for Si as calculated with LSDA in VASP.

| $V_{prim}$ (Å$^3$) | CFR fraction | Band gap from DOS (eV) |
|---------------------|--------------|------------------------|
| 39.3660             | 0.0000       | 0.3872                 |
| 43.9040             | 0.0000       | 0.6069                 |
| 48.7780             | 0.0000       | 0.6498                 |
| 54.0000             | 0.0105       | 0.1719                 |
| 59.5820             | 0.0497       | 0.0000                 |
| 65.5360             | 0.1094       | 0.0000                 |
| 71.8740             | 0.1685       | 0.0000                 |
| 78.6080             | 0.2202       | 0.0000                 |
| 85.7500             | 0.2672       | 0.0000                 |
| 93.3120             | 0.3090       | 0.0000                 |
| 101.3060            | 0.3468       | 0.0000                 |
| 109.7440            | 0.3819       | 0.0000                 |
| 118.6380            | 0.4135       | 0.0000                 |
| 128.0000            | 0.4430       | 0.0000                 |
| 137.8420            | 0.4716       | 0.0000                 |
| 148.1760            | 0.4970       | 0.0000                 |
| 159.0140            | 0.5221       | 0.0000                 |
| 170.3680            | 0.5455       | 0.0000                 |
| 182.2500            | 0.5678       | 0.0000                 |
| 194.6720            | 0.5892       | 0.0000                 |
| 207.6460            | 0.6091       | 0.0000                 |
| 221.1840            | 0.6285       | 0.0000                 |
| 235.2980            | 0.6472       | 0.0000                 |
| 250.0000            | 0.6648       | 0.0000                 |
TABLE S43: Raw data for C as calculated with PBE in Castep.

| $V_{\text{prim}}$ (Å$^3$) | CFR fraction |
|----------------------------|--------------|
| 3.9062                     | 0.0000       |
| 4.3940                     | 0.0000       |
| 4.9208                     | 0.0000       |
| 5.4880                     | 0.0000       |
| 6.0972                     | 0.0000       |
| 6.7500                     | 0.0000       |
| 7.4478                     | 0.0000       |
| 8.1920                     | 0.0000       |
| 8.9842                     | 0.0000       |
| 9.8260                     | 0.0000       |
| 10.7188                    | 0.0000       |
| 11.6640                    | 0.0000       |
| 12.6633                    | 0.0000       |
| 13.7180                    | 0.0000       |
| 14.8297                    | 0.0000       |
| 16.0000                    | 0.0000       |
| 17.2302                    | 0.0000       |
| 18.5220                    | 0.0000       |
| 19.8767                    | 0.0000       |
| 21.2960                    | 0.0015       |
| 22.7812                    | 0.0270       |
| 24.3340                    | 0.0986       |
| 25.9558                    | 0.1420       |
| 27.6480                    | 0.1723       |
| 29.4122                    | 0.2062       |
| 31.2500                    | 0.2465       |
| 33.1628                    | 0.2781       |
| 35.1520                    | 0.3125       |
| 37.2192                    | 0.3388       |
| 39.3660                    | 0.3700       |
| 41.5938                    | 0.3923       |
| 43.9040                    | 0.4157       |
| 46.2982                    | 0.4402       |
| 48.7780                    | 0.4602       |
| 51.3448                    | 0.4800       |
| 54.0000                    | 0.4969       |
| 56.7452                    | 0.5169       |
| 59.5820                    | 0.5373       |
| 62.5118                    | 0.5536       |
| 65.5360                    | 0.5724       |
| 68.5662                    | 0.5900       |
| 71.5740                    | 0.6077       |
| 75.1908                    | 0.6258       |
| 78.6080                    | 0.6447       |
| 82.1272                    | 0.6581       |
| 85.7500                    | 0.6731       |
| 89.4778                    | 0.6911       |
| 93.3120                    | 0.7033       |
| 97.2542                    | 0.7137       |
| 101.3060                   | 0.7253       |
| 105.4688                   | 0.7362       |
| 109.7440                   | 0.7474       |
| 114.1332                   | 0.7576       |
| 118.6380                   | 0.7664       |
| 123.2598                   | 0.7776       |
| 128.0000                   | 0.7847       |
| 132.8602                   | 0.7922       |
| 137.8420                   | 0.8000       |
| 142.9468                   | 0.8063       |

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TABLE S44: Raw data for Si as calculated with PBE in Castep.

| \( V_{\text{prim}} \) (\( \text{Å}^3 \)) | CFR fraction |
|----------------------------------------|-------------|
| 3.9062                                 | 0.0000      |
| 4.3940                                 | 0.0000      |
| 4.9208                                 | 0.0000      |
| 5.4880                                 | 0.0000      |
| 6.0972                                 | 0.0000      |
| 6.7500                                 | 0.0000      |
| 7.4478                                 | 0.0000      |
| 8.1920                                 | 0.0000      |
| 8.9842                                 | 0.0000      |
| 9.8260                                 | 0.0000      |
| 10.7188                                | 0.0000      |
| 11.6640                                | 0.0000      |
| 12.6633                                | 0.0000      |
| 13.7180                                | 0.0000      |
| 14.8297                                | 0.0000      |
| 16.0000                                | 0.0000      |
| 17.2302                                | 0.0000      |
| 18.5220                                | 0.0000      |
| 19.8767                                | 0.0000      |
| 21.2960                                | 0.0000      |
| 22.7812                                | 0.0000      |
| 24.3340                                | 0.0000      |
| 25.9558                                | 0.0000      |
| 27.6480                                | 0.0000      |
| 29.4122                                | 0.0000      |
| 31.2500                                | 0.0000      |
| 33.1628                                | 0.0000      |
| 35.1520                                | 0.0000      |
| 37.2192                                | 0.0000      |
| 39.3660                                | 0.0000      |
| 41.5938                                | 0.0000      |
| 43.9040                                | 0.0005      |
| 46.2982                                | 0.0020      |
| 48.7780                                | 0.0093      |
| 51.3448                                | 0.0176      |
| 54.0000                                | 0.0208      |
| 56.7452                                | 0.0614      |
| 59.5820                                | 0.1012      |
| 62.5118                                | 0.1289      |
| 65.5360                                | 0.1677      |
| 68.6562                                | 0.1903      |
| 71.8740                                | 0.2162      |
| 75.1908                                | 0.2368      |
| 78.6080                                | 0.2632      |
| 82.1272                                | 0.2843      |
| 85.7500                                | 0.2955      |
| 89.4778                                | 0.3277      |
| 93.3120                                | 0.3444      |
| 97.2542                                | 0.3609      |
| 101.3060                               | 0.3779      |
| 105.4688                               | 0.3959      |
| 109.7440                               | 0.4042      |
| 114.1332                               | 0.4236      |
| 118.6380                               | 0.4381      |
| 123.2598                               | 0.4504      |
| 128.0000                               | 0.4648      |
| 132.8602                               | 0.4792      |
| 137.8420                               | 0.4887      |
| 142.9468                               | 0.5017      |

\( V_{\text{prim}} \) (\( \text{Å}^3 \)) | CFR fraction
--- | ---
148.1760 | 0.5154
153.5312 | 0.5292
159.0140 | 0.5456
164.6258 | 0.5569
170.3680 | 0.5639
176.2422 | 0.5773
182.2500 | 0.5851
188.3928 | 0.5964
194.6720 | 0.6056
201.0900 | 0.6121
207.6460 | 0.6224
214.3440 | 0.6357
221.1840 | 0.6455
228.1680 | 0.6540
235.2980 | 0.6659
242.5740 | 0.6758
TABLE S45: Raw data for Ge as calculated with PBE in Castep.

| $V_{\text{atom}}$ (Å$^3$) | CFR fraction |
|---------------------------|--------------|
| 3.9062                    | 0.0000       |
| 4.3940                    | 0.0000       |
| 4.9207                    | 0.0000       |
| 5.4880                    | 0.0000       |
| 6.0972                    | 0.0000       |
| 6.7500                    | 0.0000       |
| 7.4478                    | 0.0000       |
| 8.1920                    | 0.0000       |
| 8.9842                    | 0.0000       |
| 9.8260                    | 0.0000       |
| 10.7188                   | 0.0000       |
| 11.6640                   | 0.0000       |
| 12.6633                   | 0.0000       |
| 13.7180                   | 0.0000       |
| 14.8297                   | 0.0000       |
| 16.0000                   | 0.0000       |
| 17.2302                   | 0.0000       |
| 18.5220                   | 0.0000       |
| 19.8767                   | 0.0000       |
| 21.2960                   | 0.0000       |
| 22.7812                   | 0.0000       |
| 24.3340                   | 0.0000       |
| 25.9558                   | 0.0000       |
| 27.6480                   | 0.0000       |
| 29.4123                   | 0.0000       |
| 31.2500                   | 0.0000       |
| 33.1627                   | 0.0000       |
| 35.1520                   | 0.0000       |
| 37.2192                   | 0.0000       |
| 39.3660                   | 0.0000       |
| 41.5938                   | 0.0000       |
| 43.9040                   | 0.0000       |
| 46.2983                   | 0.0000       |
| 48.7780                   | 0.0000       |
| 51.3448                   | 0.0008       |
| 54.0000                   | 0.0065       |
| 56.7452                   | 0.0284       |
| 59.5820                   | 0.0616       |
| 62.5117                   | 0.1063       |
| 65.5360                   | 0.1401       |
| 68.6562                   | 0.1633       |
| 71.8740                   | 0.1882       |
| 75.1908                   | 0.2109       |
| 78.6080                   | 0.2322       |
| 82.1273                   | 0.2523       |
| 85.7500                   | 0.2727       |
| 89.4777                   | 0.2919       |
| 93.3120                   | 0.3104       |
| 97.2542                   | 0.3287       |
| 101.3060                  | 0.3456       |
| 105.4690                  | 0.3628       |
| 109.7440                  | 0.3789       |
| 114.1330                  | 0.3934       |
| 118.6380                  | 0.4087       |
| 123.2600                  | 0.4239       |
| 128.0000                  | 0.4382       |
**TABLE S46:** Raw data for Sn as calculated with PBE in Castep.

| \( V_{\text{prim}} (\text{Å}^3) \) | CFR fraction |
|----------------------------------|--------------|
| 3.9062                          | 0.0000       |
| 4.3940                          | 0.0000       |
| 4.9207                          | 0.0000       |
| 5.4880                          | 0.0000       |
| 6.0972                          | 0.0000       |
| 6.7500                          | 0.0000       |
| 7.4478                          | 0.0000       |
| 8.1920                          | 0.0000       |
| 8.9842                          | 0.0000       |
| 9.8260                          | 0.0000       |
| 10.7188                         | 0.0000       |
| 11.6640                         | 0.0000       |
| 12.6633                         | 0.0000       |
| 13.7180                         | 0.0000       |
| 14.8297                         | 0.0000       |
| 16.0000                         | 0.0000       |
| 17.2302                         | 0.0000       |
| 18.5220                         | 0.0000       |
| 19.8767                         | 0.0000       |
| 21.2960                         | 0.0000       |
| 22.7812                         | 0.0000       |
| 24.3340                         | 0.0000       |
| 25.9558                         | 0.0000       |
| 27.6480                         | 0.0000       |
| 29.4123                         | 0.0000       |
| 31.2500                         | 0.0000       |
| 33.1627                         | 0.0000       |
| 35.1520                         | 0.0000       |
| 37.2192                         | 0.0000       |
| 39.3660                         | 0.0000       |
| 41.5938                         | 0.0000       |
| 43.9040                         | 0.0000       |
| 46.2983                         | 0.0000       |
| 48.7780                         | 0.0000       |
| 51.3448                         | 0.0000       |
| 54.0000                         | 0.0000       |
| 56.7452                         | 0.0000       |
| 59.5820                         | 0.0000       |
| 62.5117                         | 0.0000       |
| 65.3360                         | 0.0001       |
| 68.6562                         | 0.0027       |
| 71.9740                         | 0.0136       |
| 75.1908                         | 0.0349       |
| 78.6080                         | 0.0655       |
| 82.1273                         | 0.1074       |
| 85.7500                         | 0.1375       |
| 89.4777                         | 0.1621       |
| 93.3120                         | 0.1850       |
| 97.2542                         | 0.2056       |
| 101.3060                        | 0.2252       |
| 105.4690                        | 0.2434       |
| 109.7440                        | 0.2641       |
| 114.1330                        | 0.2831       |
| 118.6380                        | 0.3009       |
| 123.2600                        | 0.3178       |
| 128.0000                        | 0.3343       |
| 132.8600                        | 0.3485       |
| 137.8420                        | 0.3651       |
| 142.9470                        | 0.3798       |
| ...                             | ...          |
| $V_{\text{prim}}$ (Å$^3$) | CFR fraction | $V_{\text{prim}}$ (Å$^3$) | CFR fraction |
|--------------------------|--------------|--------------------------|--------------|
| 3.9062                   | 0.0000       | 148.1760                 | 0.3584       |
| 4.3940                   | 0.0000       | 153.5310                 | 0.3726       |
| 4.9207                   | 0.0000       | 159.0140                 | 0.3866       |
| 5.4880                   | 0.0000       | 164.6260                 | 0.3977       |
| 6.0972                   | 0.0000       | 170.3680                 | 0.4124       |
| 6.7500                   | 0.0000       | 176.2420                 | 0.4230       |
| 7.4478                   | 0.0000       | 182.2500                 | 0.4369       |
| 8.1920                   | 0.0000       | 188.3930                 | 0.4481       |
| 8.9842                   | 0.0000       | 194.6720                 | 0.4612       |
| 9.8260                   | 0.0000       | 201.0890                 | 0.4706       |
| 10.7188                  | 0.0000       | 207.6460                 | 0.4838       |
| 11.6640                  | 0.0000       | 214.3440                 | 0.4911       |
| 12.6633                  | 0.0000       | 221.1840                 | 0.5033       |
| 13.7180                  | 0.0000       | 228.1680                 | 0.5128       |
| 14.8297                  | 0.0000       | 235.2980                 | 0.5225       |
| 16.0000                  | 0.0000       | 242.5750                 | 0.5328       |
| 17.2302                  | 0.0000       |                          |              |
| 18.5220                  | 0.0000       |                          |              |
| 19.8767                  | 0.0000       |                          |              |
| 21.2960                  | 0.0000       |                          |              |
| 22.7812                  | 0.0000       |                          |              |
| 24.3340                  | 0.0000       |                          |              |
| 25.9558                  | 0.0000       |                          |              |
| 27.6480                  | 0.0000       |                          |              |
| 29.4123                  | 0.0000       |                          |              |
| 31.2500                  | 0.0000       |                          |              |
| 33.1627                  | 0.0000       |                          |              |
| 35.1520                  | 0.0000       |                          |              |
| 37.2192                  | 0.0000       |                          |              |
| 39.3660                  | 0.0000       |                          |              |
| 41.5938                  | 0.0000       |                          |              |
| 43.9040                  | 0.0000       |                          |              |
| 46.2983                  | 0.0000       |                          |              |
| 48.7780                  | 0.0000       |                          |              |
| 51.3448                  | 0.0000       |                          |              |
| 54.0000                  | 0.0000       |                          |              |
| 56.7452                  | 0.0000       |                          |              |
| 59.5820                  | 0.0000       |                          |              |
| 62.5117                  | 0.0000       |                          |              |
| 65.5360                  | 0.0000       |                          |              |
| 68.6562                  | 0.0000       |                          |              |
| 71.8740                  | 0.0000       |                          |              |
| 75.1908                  | 0.0023       |                          |              |
| 78.6080                  | 0.0177       |                          |              |
| 82.1273                  | 0.0418       |                          |              |
| 85.7500                  | 0.0711       |                          |              |
| 89.4777                  | 0.1127       |                          |              |
| 93.3120                  | 0.1429       |                          |              |
| 97.2542                  | 0.1641       |                          |              |
| 101.3060                 | 0.1877       |                          |              |
| 105.4690                 | 0.2061       |                          |              |
| 109.7440                 | 0.2239       |                          |              |
| 114.1330                 | 0.2426       |                          |              |
| 118.6380                 | 0.2628       |                          |              |
| 123.2600                 | 0.2771       |                          |              |
| 128.0000                 | 0.2971       |                          |              |
| 132.8600                 | 0.3106       |                          |              |
| 137.8420                 | 0.3295       |                          |              |
| 142.9470                 | 0.3429       |                          |              |
|                          |              |                          |              |
|                          |              |                          |              |
S5. RAW DATA FOR GROUP 18 ELEMENTAL SOLIDS

TABLE S48: Raw data for Ne as calculated with PBE in VASP.

| $V_{\text{prim}}$ (Å³) | CFR fraction | Band gap from DOS (eV) |
|-------------------------|--------------|------------------------|
| 5.7873                  | 0.0933       | 18.5758                |
| 6.0972                  | 0.1847       | 17.9035                |
| 6.4181                  | 0.3122       | 17.5081                |
| 6.7500                  | 0.3719       | 16.9491                |
| 8.1920                  | 0.5404       | 14.9407                |
| 9.8260                  | 0.6535       | 13.8288                |
| 11.6640                 | 0.7240       | 12.9718                |
| 13.7180                 | 0.7763       | 12.4658                |
| 16.0000                 | 0.8088       | 12.1625                |
| 18.5220                 | 0.8356       | 11.7600                |
| 21.2960                 | 0.8570       | 11.5588                |
| 24.3340                 | 0.8745       | 11.4974                |
| 27.6480                 | 0.8899       | 11.2233                |
| 31.2500                 | 0.9024       | 11.2762                |
| 35.1520                 | 0.9131       | 11.2431                |
| 39.3660                 | 0.9227       | 11.3382                |
| 43.9040                 | 0.9306       | 11.3177                |
| 48.7780                 | 0.9373       | 11.3249                |
| 54.0000                 | 0.9436       | 11.3459                |
TABLE S49: Raw data for Ne as calculated with LSDA in VASP.

| V\text{prim} (\ang) | CFR fraction | Band gap from DOS (eV) |
|---------------------|--------------|------------------------|
| 5.7873              | 0.0281       | 18.3436                |
| 6.0972              | 0.0576       | 17.6710                |
| 6.4181              | 0.1407       | 17.0007                |
| 6.7500              | 0.2294       | 16.4522                |
| 8.1920              | 0.5164       | 14.4664                |
| 9.8260              | 0.6455       | 13.4261                |
| 11.6640             | 0.7178       | 12.4733                |
| 13.7180             | 0.7691       | 11.9869                |
| 16.0000             | 0.8060       | 11.6004                |
| 18.5220             | 0.8354       | 11.3754                |
| 21.2960             | 0.8570       | 11.3931                |
| 24.3340             | 0.8745       | 11.3476                |
| 27.6480             | 0.8899       | 11.2333                |
| 31.2500             | 0.9024       | 11.3534                |
| 35.1520             | 0.9131       | 11.3287                |
| 39.3660             | 0.9227       | 11.4308                |
| 43.9040             | 0.9306       | 11.5933                |
| 48.7780             | 0.9373       | 11.6459                |
| 54.0000             | 0.9436       | 11.7969                |
TABLE S50: Raw data for He as calculated with PBE in Castep.

| \( V_{\text{prim}} \) (Å³) | CFR fraction |
|---------------------------|--------------|
| 0.2500                    | 0.0000       |
| 0.3327                    | 0.0000       |
| 0.4320                    | 0.0000       |
| 0.5493                    | 0.0000       |
| 0.6860                    | 0.0000       |
| 0.8438                    | 0.0000       |
| 1.0240                    | 0.0000       |
| 1.2283                    | 0.0000       |
| 1.4580                    | 0.0000       |
| 1.7147                    | 0.0000       |
| 2.0000                    | 0.0000       |
| 2.3152                    | 0.0000       |
| 2.6620                    | 0.0365       |
| 3.0417                    | 0.2798       |
| 3.4560                    | 0.4887       |
| 3.9062                    | 0.6073       |
| 4.3940                    | 0.6560       |
| 4.9207                    | 0.7319       |
| 5.4880                    | 0.7805       |
| 6.0972                    | 0.8058       |
| 6.7500                    | 0.8310       |
| 7.4478                    | 0.8523       |
| 8.1920                    | 0.8745       |
| 8.9842                    | 0.8871       |
| 9.8260                    | 0.8966       |
| 10.7188                   | 0.9104       |
| 11.6640                   | 0.9145       |
| 12.6633                   | 0.9207       |
| 13.7180                   | 0.9315       |
| 14.8297                   | 0.9356       |
| 16.0000                   | 0.9407       |
| 17.2302                   | 0.9441       |
| 18.5220                   | 0.9501       |
| 19.8767                   | 0.9534       |
| 21.2960                   | 0.9556       |
| 22.7812                   | 0.9585       |
| 24.3340                   | 0.9615       |
| 25.9558                   | 0.9636       |
| 27.6480                   | 0.9665       |
| 29.4123                   | 0.9694       |
| 31.2500                   | 0.9711       |
| 33.1627                   | 0.9728       |
| 35.1520                   | 0.9733       |
| 37.2192                   | 0.9760       |
| 39.3660                   | 0.9764       |
| 41.5938                   | 0.9773       |
| 43.9040                   | 0.9797       |
| 46.2983                   | 0.9809       |
| 48.7780                   | 0.9812       |
| 51.3448                   | 0.9824       |
| 54.0000                   | 0.9829       |
| 56.7452                   | 0.9834       |
| 59.5820                   | 0.9843       |
| 62.5117                   | 0.9852       |
| 65.5360                   | 0.9863       |
| 68.6562                   | 0.9863       |
| 71.8740                   | 0.9871       |
| 75.1908                   | 0.9878       |
| 78.6080                   | 0.9885       |
| 82.1273                   | 0.9887       |
| 85.7500                   | 0.9891       |
| 89.4777                   | 0.9898       |
| 93.3120                   | 0.9901       |
| 97.2542                   | 0.9904       |
| 101.3060                  | 0.9909       |
| 105.4690                  | 0.9914       |
| 109.7440                  | 0.9920       |
| 114.1330                  | 0.9921       |
| 118.6380                  | 0.9921       |
| 123.2600                  | 0.9926       |
| 128.0000                  | 0.9928       |
TABLE S51: Raw data for Ne as calculated with PBE in Castep.

| $V_{\text{prim}}$ (Å$^3$) | CFR fraction |
|---------------------------|--------------|
| 3.9062                    | 0.0000       |
| 4.3940                    | 0.0000       |
| 4.9207                    | 0.0000       |
| 5.4880                    | 0.0359       |
| 6.0972                    | 0.1041       |
| 6.7500                    | 0.2904       |
| 7.4478                    | 0.4156       |
| 8.1920                    | 0.4829       |
| 8.9842                    | 0.5534       |
| 9.8260                    | 0.6158       |
| 10.7188                   | 0.6594       |
| 11.6640                   | 0.6909       |
| 12.6633                   | 0.7259       |
| 13.7180                   | 0.7556       |
| 14.8297                   | 0.7745       |
| 16.0000                   | 0.7939       |
| 17.2302                   | 0.8111       |
| 18.5220                   | 0.8253       |
| 19.8767                   | 0.8388       |
| 21.2960                   | 0.8494       |
| 22.7812                   | 0.8594       |
| 24.3340                   | 0.8702       |
| 25.9558                   | 0.8772       |
| 27.6480                   | 0.8868       |
| 29.4123                   | 0.8949       |
| 31.2500                   | 0.9007       |
| 33.1627                   | 0.9049       |
| 35.1520                   | 0.9111       |
| 37.2192                   | 0.9157       |
| 39.3660                   | 0.9197       |
| 41.5938                   | 0.9249       |
| 43.9040                   | 0.9280       |
| 46.2983                   | 0.9307       |
| 48.7780                   | 0.9364       |
| 51.3448                   | 0.9396       |
| 54.0000                   | 0.9422       |
| 56.7452                   | 0.9436       |
| 59.5820                   | 0.9470       |
| 62.5117                   | 0.9497       |
| 65.5360                   | 0.9525       |
| 68.6562                   | 0.9535       |
| 71.8740                   | 0.9565       |
| 75.1908                   | 0.9583       |
| 78.6080                   | 0.9599       |
| 82.1273                   | 0.9621       |
| 85.7500                   | 0.9634       |
| 89.4777                   | 0.9650       |
| 93.3120                   | 0.9661       |
| 97.2542                   | 0.9682       |
| 101.3060                  | 0.9694       |
| 105.4690                  | 0.9704       |
| 109.7440                  | 0.9725       |
| 118.6380                  | 0.9737       |
| 123.2600                  | 0.9746       |
| 128.0000                  | 0.9755       |
| 132.8600                  | 0.9762       |
| 137.8420                  | 0.9773       |
| 142.9470                  | 0.9785       |
| 148.1760                  | 0.9787       |

$V_{\text{prim}}$ (Å$^3$) | CFR fraction
---|---
153.5310 | 0.9795
159.0140 | 0.9802
164.6260 | 0.9812
TABLE S52: Raw data for Ar as calculated with PBE in Castep.

| $V_{pima}$ (Å³) | CFR fraction |
|------------------|--------------|
| 3.9062           | 0.0000       |
| 4.3940           | 0.0000       |
| 4.9207           | 0.0000       |
| 5.4880           | 0.0000       |
| 6.0972           | 0.0000       |
| 6.7500           | 0.0000       |
| 7.4478           | 0.0000       |
| 8.1920           | 0.0000       |
| 8.9842           | 0.0000       |
| 9.8260           | 0.0000       |
| 10.7188          | 0.0000       |
| 11.6640          | 0.0000       |
| 12.6633          | 0.0000       |
| 13.7180          | 0.0004       |
| 14.8297          | 0.0346       |
| 16.0000          | 0.0745       |
| 17.2302          | 0.1886       |
| 18.5220          | 0.3148       |
| 19.8767          | 0.4153       |
| 21.2960          | 0.4775       |
| 22.7812          | 0.5201       |
| 24.3340          | 0.5577       |
| 25.9558          | 0.6075       |
| 27.6480          | 0.6405       |
| 29.4123          | 0.6620       |
| 31.2500          | 0.6832       |
| 33.1627          | 0.7079       |
| 35.1520          | 0.7326       |
| 37.2192          | 0.7475       |
| 39.3660          | 0.7626       |
| 41.5938          | 0.7791       |
| 43.9040          | 0.7936       |
| 46.2983          | 0.8051       |
| 48.7780          | 0.8159       |
| 51.3448          | 0.8249       |
| 54.0000          | 0.8374       |
| 56.7452          | 0.8412       |
| 59.5820          | 0.8505       |
| 62.5117          | 0.8583       |
| 65.5360          | 0.8652       |
| 68.6562          | 0.8705       |
| 71.8740          | 0.8771       |
| 75.1908          | 0.8816       |
| 78.6080          | 0.8874       |
| 82.1273          | 0.8935       |
| 85.7500          | 0.8955       |
| 89.4777          | 0.9016       |
| 93.3120          | 0.9068       |
| 97.2542          | 0.9090       |
| 101.3060         | 0.9127       |
| 105.4690         | 0.9175       |
| 109.7440         | 0.9195       |
| 114.1330         | 0.9244       |
| 118.6380         | 0.9252       |
| 123.2600         | 0.9288       |
| 128.0000         | 0.9309       |
TABLE S53: Raw data for Kr as calculated with PBE in Castep.

| $V_{prim}$ (Å$^3$) | CFR fraction |
|---------------------|--------------|
| 3.9062              | 0.0000       |
| 4.3940              | 0.0000       |
| 4.9207              | 0.0000       |
| 5.4880              | 0.0000       |
| 6.0972              | 0.0000       |
| 6.7500              | 0.0000       |
| 7.4478              | 0.0000       |
| 8.1920              | 0.0000       |
| 8.9842              | 0.0000       |
| 9.8260              | 0.0000       |
| 10.7188             | 0.0000       |
| 11.6640             | 0.0000       |
| 12.6633             | 0.0000       |
| 13.7180             | 0.0000       |
| 14.8297             | 0.0000       |
| 16.0000             | 0.0000       |
| 17.2302             | 0.0000       |
| 18.5220             | 0.0000       |
| 19.8767             | 0.0140       |
| 21.2960             | 0.0396       |
| 22.7812             | 0.0852       |
| 24.3340             | 0.1839       |
| 25.9558             | 0.2970       |
| 27.6480             | 0.3921       |
| 29.4123             | 0.4530       |
| 31.2500             | 0.5050       |
| 33.1627             | 0.5314       |
| 35.1520             | 0.5740       |
| 37.2192             | 0.6097       |
| 39.3660             | 0.6343       |
| 41.5938             | 0.6668       |
| 43.9040             | 0.6832       |
| 46.2983             | 0.7079       |
| 48.7780             | 0.7229       |
| 51.3448             | 0.7394       |
| 54.0000             | 0.7556       |
| 56.7452             | 0.7661       |
| 59.5820             | 0.7796       |
| 62.5117             | 0.7901       |
| 65.5360             | 0.8006       |
| 68.6562             | 0.8087       |
| 71.8740             | 0.8187       |
| 75.1908             | 0.8276       |
| 78.6080             | 0.8347       |
| 82.1273             | 0.8416       |
| 85.7500             | 0.8500       |
| 89.4777             | 0.8562       |
| 93.3120             | 0.8617       |
| 97.2542             | 0.8646       |
| 101.3060            | 0.8756       |
| 105.4690            | 0.8772       |
| 109.7440            | 0.8828       |
| 114.1330            | 0.8892       |
| 118.6380            | 0.8915       |
| 123.2600            | 0.8964       |
| 128.0000            | 0.9006       |
TABLE S54: Raw data for Xe as calculated with PBE in Castep.

| $V_{\text{cell}}$ (Å$^3$) | CFR fraction |
|---------------------------|--------------|
| 4.3940                    | 0.0000       |
| 4.9207                    | 0.0000       |
| 5.4880                    | 0.0000       |
| 6.0972                    | 0.0000       |
| 6.7500                    | 0.0000       |
| 7.4478                    | 0.0000       |
| 8.1920                    | 0.0000       |
| 8.9842                    | 0.0000       |
| 9.8260                    | 0.0000       |
| 10.7188                   | 0.0000       |
| 11.6640                   | 0.0000       |
| 12.6633                   | 0.0000       |
| 13.7180                   | 0.0000       |
| 14.8297                   | 0.0000       |
| 16.0000                   | 0.0000       |
| 17.2302                   | 0.0000       |
| 18.5220                   | 0.0000       |
| 19.8767                   | 0.0000       |
| 21.2960                   | 0.0000       |
| 22.7812                   | 0.0000       |
| 24.3340                   | 0.0000       |
| 25.9558                   | 0.0003       |
| 27.6480                   | 0.0003       |
| 29.4123                   | 0.0177       |
| 31.2500                   | 0.0200       |
| 33.1627                   | 0.0796       |
| 35.1520                   | 0.1097       |
| 37.2192                   | 0.2275       |
| 39.3660                   | 0.3067       |
| 41.5938                   | 0.3767       |
| 43.9040                   | 0.4407       |
| 46.2983                   | 0.4856       |
| 48.7780                   | 0.5124       |
| 51.3448                   | 0.5530       |
| 54.0000                   | 0.5850       |
| 56.7452                   | 0.6147       |
| 59.5820                   | 0.6271       |
| 62.5117                   | 0.6556       |
| 65.5360                   | 0.6769       |
| 68.6562                   | 0.6884       |
| 71.8740                   | 0.7074       |
| 75.1908                   | 0.7250       |
| 78.6080                   | 0.7426       |
| 82.1273                   | 0.7550       |
| 85.7500                   | 0.7599       |
| 89.4777                   | 0.7708       |
| 93.3120                   | 0.7832       |
| 97.2542                   | 0.7924       |
| 101.3060                  | 0.7986       |
| 105.4690                  | 0.8110       |
| 109.7440                  | 0.8192       |
| 114.1330                  | 0.8245       |
| 118.6380                  | 0.8341       |
| 123.2600                  | 0.8389       |
| 128.0000                  | 0.8427       |