on the pusher-shell interior, is most likely a
nonuniformity in pusher-ion density \( N_i \) indica-
tive of breakup of the pusher shell during target
irradiation.

In addition to the above observations, the STX
images provide some information about STE
transport in short-pulse, exploding-pushers, mi-
crosphere targets. The STX simulation of Fig. 3
includes neither STE transport inhibition\(^2\) nor en-
hanced azimuthal transport of STE's in the target
sheath.\(^3\) The good agreement between the images
of Figs. 2 and 3 indicate that under favorable ex-
perimental conditions (i.e., quasiuniform illumina-
tion—no hot spots) such STE transport mechan-
isms probably do not play an important role in
this class of laser fusion experiments.

We wish to acknowledge the assistance of our
Lawrence Livermore Laboratory colleagues
H. G. Ahlstrom, D. T. Atwood, Y. L. Pan,
G. Howe, D. Ciarlo, C. Dittmore, W. Herrman,
G. Stone, and the Shiva laser crew. This work
was performed under the auspices of the U. S.

\(^1\) J. F. Kephart, R. P. Godwin, and G. H. McCall,
Kornblum, and H. D. Shay, J. Appl. Phys. 46, 1973
(1975).

\(^2\) High-resolution imaging of the STX emission was
found to be beyond the capabilities of pinhole and graz-
ing incidence reflection techniques because of the low
flux levels and high energies involved.

\(^3\) Zone-plate coded imaging is a two-step imaging tech-
nique. In the first step the x-ray source casts a shadow-
graph through a Fresnel zone plate onto an appropriate
film. Image reconstruction (step two) is achieved opti-
cally with methods similar to holographic reconstruc-
tion. For further details see N. M. Ceglio, et al., J.
Appl. Phys. 46, 1965 (1977), and 48, 1566 (1977), and

A layer in this pack consists of a filter foil followed
by three single-sided x-ray films—types M, R, and FGP.
The foil material, thickness, and x-ray image energies
for the various layers are as follows: layer 1, Be, 150
\( \mu \)m, 4–7 keV; layer 2, Al, 250 \( \mu \)m, 10–20 keV; layer
3, Al, 1250 \( \mu \)m, 17–30 keV.

The image data format for all the figures is as fol-
loows: In the two-dimensional (2D) contour maps each
countour is a locus of constant x-ray emission intensity.
The incremental intensity change is constant between
successive contours. In the 2D plots the two opposing
ten-laser beam clusters are incident on the target from
top and bottom. In the 3D plots the beam clusters are
incident from the left and right, respectively. “Left”
on the 2D plots corresponds to “top” on the 2D plots.

\(^4\) LASnex is a 2D Lagrangian, magnetohydrodynamics
code with multigroup electron transport. For further
details see G. B. Zimmerman, Lawrence Livermore

\(^5\) In these short-pulse, large-ball experiments, peak
target compressions occur well after the peak of the
laser pulse.

\(^6\) S. Braginskii, Reviews of Plasma Physics (Consult-

\(^7\) J. R. Albritton et al., Phys. Rev. Lett. 59, 1536

Structural Stability of 495 Binary Compounds

Alex Zunger

Solar Energy Research Institute, Golden, Colorado 80401

(Received 18 June 1979)

With use of the characteristic turning-point radii of the first-principles nonlocal den-
sity-functional atomic pseudopotentials, a successful topological prediction of the crys-
tal structures of 495 binary \( AB \) compounds of transition and simple elements is ob-
tained.

There are about 500 stable, near-stoichio-
metric, ordered binary solids made of atoms be-
longing to the first five rows of the periodic ta-
ble whose crystal structure has been determined
experimentally. Most of these crystals appear in
their most stable form in one of about twenty
distinct spatial structures. In this Letter I show
that a simple nonempirical quantum mechanical
approach, based on the density-functional non-
local pseudopotential concept, explains this dis-
tribution diagrammatically with striking suc-
cess.

Even before the pioneering studies of Gold-
schmidt, Pauling, and others\(^1\) it was known ther-
modynamically that the structure-dependent energy $\Delta E_s$ of most ordered solids is small compared to either the cohesive energy $\Delta E_c$ or the heat of formation. Measured heat-of-transformation data\(^2\) as well as quantum mechanical calculations of stable and hypothetical structures indicate that $\Delta E_s/\Delta E_c$ can be as small as $10^{-1} - 10^{-4}$. For the 50–60 nontransition-metal binary octet compounds, the problem of systematizing the five crystal structures (NaCl, CsCl, diamond, zincblende, and wurzite) has been solved through the use of optical dielectric\(^3\) as well as spectroscopic\(^4\) electronegativity concepts. These concepts\(^5\) display periodic trends diagrammatically when transferable elemental coordinates are used. Can the diagrammatic Pauling-esque approaches be extended to include intermetallic transition metal (TM) compounds as well?

While contemporary quantum mechanical models emphasize the decisive role of $d$ electrons in determining the cohesive properties of TM compounds (e.g., Ref. 6), semiclassical approaches\(^7,8\) have concentrated on more general phenomenological "factors" in an attempt to rationalize crystal structures. These include metallurgical and chemical constructs such as the atomic size, local geometry, coordination number, electronegativity, average electron count, and orbital promotion energy factors, where a certain subset of these ("strong factors") are frequently used to explain the occurrence of certain classes of compounds (usually 20–50 compounds at a time). The coexistence of many crystal structures in overlapping regions of these parameters, however, limits this approach to groups exhibiting narrow ranges of chemical properties. Nevertheless, these semiclassical approaches provide valuable insight into the problem because they point to the underlying importance of establishing elementary system-invariant energy scales (e.g., electronegativity, promotion energy) as well as length scales (e.g., covalent, metallic, or ionic radii).

In this Letter I show that the recently developed\(^9\) first-principles density-functional nonlocal atomic pseudopotentials can provide nonempirical angular-dependent internal energy and length scales. By using a dual coordinate system derived from these scales I am able to obtain a topological separation of the structures of binary compounds (including simple and TM atoms) with a surprising accuracy.

The resulting pseudopotentials as well as a recent variant of them\(^10\) have been shown to have a very small energy dependence\(^9\) and to yield in self-consistent electronic structure calculations remarkably precise results for orbital energies and charge densities for atoms,\(^9,10\) molecules,\(^11\) bulk semiconductors\(^12\) (e.g., for silicon deviation of 0.06 eV/state between the pseudopotential band structure and the full all-electron band structure, over a 20-eV range), and transition metals\(^13\) as well as cohesive properties of elemental simple\(^14\) and transition-metal\(^15\) solids. They differ from the empirical and semiempirical potentials used extensively in the literature (e.g., Cohen et al.\(^15\) and Appelbaum and Hamann\(^16\)) in the occurrence of "hard-core" $I$-dependent classical turning points $V_{\text{eff}}^\text{(l)}$ ($r_i^{0}=0$ reflecting the high-momentum-transfer components $q > 2k_F$ of the electron-core scattering).

I define the elementary nonlocal coordinates of an atom from the self-consistently screened pseudopotential $V_{\text{eff}}^\text{(l)}(r_i) = 0$, where $V_{\text{eff}}^\text{(l)}(r) = V_{\text{ps}}^\text{(l)}(r) + V_{\text{coh}}(\rho(r)) + V_{\text{ex}}(\rho(r)) + l(l + 1)/2r^2$. These radii differ from those of St. John and Bloch\(^7\) which were derived empirically from the spectra of single-electron ions for nontransition elements alone. I find that $r_i^{-1}$ scales linearly with the experimental multiplet-average atomic ionization energies $E_I$. As $r_i^{-1}$ reflects the scattering power of a screened atomic core to electrons with angular momentum $I$, the relation $r_i^{-1} = aE_i + b$ establishes $r_i^{-1}$ as an intrinsic angular-momentum-dependent energy scale (much like the Thomas-Gordy and Mulliken electronegativity scales, which are, however, isotropic).

I similarly find that $r_j$ scales linearly with the average position of the radial nodes in the "true" valence orbital $\psi_{nl}^\text{(l)}(r)^{10}$ suggesting that $r_j$ can also form an intrinsic length scale. The dual coordinates $\{r_i^{-1}, r_j\}$ hence satisfy the semiclassical ideas underlying successful structural indices\(^4,9,15\) as well as the dual electronegativity-size mismatch constructs of alloy heat of formation models.\(^17\) Similar characteristics of the empirically derived spectroscopic radii were first suggested by Bloch\(^16\) for the nontransition elements octet systems.

I use the definition of structural indices for an AB compound suggested previously\(^4,18\) $R_{\text{st}}^{\text{AB}} = |r_A^A - r_A^B| - (r_A^A + r_A^B)$ and $R_{\text{st}}^{\text{AB}} = (|r_A^A + r_A^B| - (r_A^A + r_A^B))$, to construct structure-separation diagrams for the most stable phase of 109 octet $A^{8}B^{8}$ (Fig. 1(a)) and 351 nonoctet $A^{P}B^{8-P}$, $3 < P \leq 6$ (Fig. 1(b)), binary compounds. I arbitrarily define structural domains by calculating the smallest number of straight lines in the $R_{\text{st}}^{\text{AB}}$
plane which minimize the number of violations and enclose minimal areas [Figs. 1(a) and 1(b)]. I am unable to separate the closely related NiAs(B8) - MnP(B31) and the CsCl(B2) - CuAu(L10) pairs [Fig. 1(b)] which show extremely small heats of transformation (often 1 kcal/mol) for compounds appearing in both forms. Also I find that the seventeen B20 (FeSi) compounds, the ten B19 (AuCd) compounds, and the eight B27 (FeB) compounds [omitted from Fig. 1(b)] for clarity] overlap precisely with the B8, B2, and B33 domains, respectively. These pairs of crystal structures are crystallographically very closely interrelated and often coexist in narrow regions of thermodynamic parameters. Otherwise, the number of violations in the present structure prediction is less than 1%. A better resolution of the TM-TM compounds belonging to the B19 - B2 - L10 structures may require a specialized scale involving d electrons more directly.

The success of the nonlocal atomic coordinates r₁ in predicting crystal structures is nontrivial: Using the Miedema heat-of-formation coordinates ΔρAB and ΔRAB or combinations of electronegativity differences ΔXAB and various metallic or ionic radii ΔRAB, or the Mooser-Pearson coordinates, I find a poor overall structural separation over the present data base. It therefore seems that the radii of the screened first-principles nonlocal DF pseudopotential provide the best structural coordinates to data for the full data base of binary crystal structures available.

From the nonoctet compounds, two groups of systematic errors can be identified: The six B2 compounds appearing in the present plots in the B8-B31-B20 domain (CoAl, CoBe, CoGa, FeAl, FeGa, and NiAl) tend to support local magnetic moments; magnetic moments are indeed known to alter significantly periodic trends in AE. The seven B33 compounds appearing in the B2 domain (AgCa, HfPt, NiHf, NiLa, NiZr, PtLa, and RhLa) are all characterized as inverted (i.e., a/c < 1) B33 structures, more akin to the cubic B2 lattice. The remaining violations (the experimentally reported structure and the presently assigned domain are denoted, respectively) are TiB (B1,B33), InP (B8,B2), PtB (B8,B1), RhBi (B8,B2), NaPb (I64,B2-B32), OsSi (B2,B8,-B20-B31), PdBe (B2,B33), CoPt (L10,B8), and TiAl (L10,B8).

Interestingly, the present structural separation scheme predicts unusual electronic properties even within given structural groups: Whereas almost all of the 150 known bcc-like B2 + L10 nonoctet binary compounds, are metals, the CsAu and RbAu systems appearing as an isolated subgroup in Fig. 1(b) were shown to be semiconductors!

The success of the present dual coordinates in separating the crystal structures of many TM-TM and TM-non-TM compounds presents a striking result: The fact that the d electrons enter the nonlocal coordinates r₁ only indirectly (via screening) suggests that the structural part ΔE₀ of the cohesive energies of compounds may be determined by s-p coordinates (which, in turn, are found to be approximately linearly dependent on the d coordinate). This points to the possibility that while the localized d electrons determine central-cell effects (such as octahedral and Jahn-Teller ligand-field stabilization, chemical trends in Mössbauer nuclear isomer shifts), and the regularities in the structure-insensitive cohesive energy ΔE₀, the longer-range s-p wave functions are responsible for the stabilization of a certain complex space-group arrangement over another. While the resonant tight-binding (TB) models have explained the periodic trends in the structure-insensitive part ΔE₀ of both elemental and alloyed TM systems by considering changes in the rectangular distributions of the one-electron d energy levels alone, it may be that in binary AB compounds with large differences in the constituent d-band energies, Eₐ(A) ≠ Eₐ(B), the s-p contribution to the structural energy ΔEₐ is indeed dominant. If ΔEₐ is derived primarily from Brillouin-zone-induced changes in the gaps E₂ and these gaps can be divided into two groups E₂ ≠ E₂ ≠ Eₐ(A) - Eₐ(B). E₂ is then expected to scale with Rg much like the heteropolar gaps. It is remarkable indeed, however, that these complex weak interactions, often masked in elaborate calculations by errors in the strong Coulombic interactions, can be regularized by transferable s-p coordinates.

A complete account of the present work will be presented elsewhere. A table of all orbital radii used here is available from the author on request.

I am grateful to J. C. Phillips for comments and discussions. I am indebted to W. Andreoni
FIG. 1. Structural separation maps for $AB$ compounds. The coordinates are $R_{\text{c}}^{AB} = |r_p^A - r_s^A| + |r_p^B - r_s^B|$ and $R_{\text{e}}^{AB} = |(r_p^A + r_s^A) - (r_p^B + r_s^B)|$. The radii $r_s$ and $r_p$ mark the crossing points of the screened nonlocal atomic pseudopotentials. (a) Octet, (b) nonoctet.

and A. Baldereschi for indicating to me a numerical error in my previous Li $r_s$ radius.

6J. Friedel, in *The Physics of Metals*, edited by J. M.
Sublimation Rate of Cobalt near Its Curie Temperature

B. C. Sales, J. E. Turner, and M. B. Maple

Department of Physics and Institute for Pure and Applied Physical Sciences, University of California, San Diego, La Jolla, California 92033

(Received 21 May 1979)

Measurements are reported of the sublimation rate of cobalt in the vicinity of its Curie temperature \( T_C = 1400 \text{ K} \) which show a relatively large change (~0.8 eV/atom) in the apparent activation energy for sublimation near \( T_C \). The results can be accounted for in terms of a simple model that incorporates into the sublimation process the temperature dependence of the magnetic contribution to the binding energy of a cobalt surface atom.

The effects of magnetic and crystallographic phase transitions of metallic substrates on their oxidation kinetics have received increasing attention in recent years. In particular, we report that there is a relatively large change in the oxidation rate of nickel in the vicinity of its Curie temperature \( T_C = 631 \text{ K} \). The change in oxidation rate was manifested as a decrease of about 1 eV/atom in the apparent activation energy for oxidation as the nickel was heated above \( T_C \). The typical oxide layers formed in this experiment were about 100 Å thick. For such thin oxide layers an important and possibly rate-limiting step in the oxidation process is the transfer of nickel ions through the nickel-nickel-oxide interface. The transfer of nickel ions into nickel oxide is analogous to the sublimation of nickel atoms into vacuum and suggests that the sublimation rate of a ferromagnetic solid might also change upon passing through its Curie temperature. However, the ferromagnetic transition of nickel, and iron as well, occurs at too low a temperature to generate a measurable sublimation current. Fortunately, such an experiment is feasible for cobalt because of its relatively high Curie temperature \( T_C = 1400 \text{ K} \). Measurements of the sublimation rate of cobalt in the vicinity of its Curie temperature are reported in this Letter.

A schematic diagram of the experimental arrangement is shown in Fig. 1. During the sublimation-rate measurements the pressure in the ultrahigh-vacuum chamber was kept below \( 3 \times 10^{-9} \) Torr. A 0.0254-cm-thick cobalt foil was heated resistively by use of a high-current power supply. The cobalt atoms that sublimed from a small region near the center of the foil were deposited on a quartz-crystal thickness monitor held near room temperature. By monitoring of the change in the resonant frequency of the quartz crystal as a function of time, the rate of cobalt deposition and hence the sublimation rate could be determined. The temperature of the cobalt foil was measured and controlled to within ±0.5 K using a Pt/Pt-13%Rh thermocouple spotwelded to the back of the foil. The Pt/Pt-13%Rh thermocouple was shielded from the harmful Co metal vapor by a 0.00254-cm-thick tantalum foil. In order to provide an additional check of the temperature, a resistance anomaly associated with the magnetic transition of cobalt was measured.