that a similar conclusion [A. L. Jain and S. H. Koenig, Phys. Rev. **127**, 442 (1962)] has been advanced recently with regard to the number of electron ellipsoids in bismuth. This latter conclusion is based on a reevaluation of extensive bismuth data.

ACKNOWLEDGMENTS

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Rydberg Function as an Interatomic Potential for Metals

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The Rydberg potential has been applied to a number of body-centered and face-centered cubic metals. The parameters of the potential were calculated using the experimental values for the energy of vaporization, the lattice constant, and the compressibility. Results have been obtained for the elastic constants, for the equation-of-state curves, and for the volume dependence of the cohesive energy. They are compared with those obtained using the Morse potential, and with the experimental data. For the elastic constants, the results by the two potentials are not very different, but a significant improvement has been obtained in several cases for the equation of state and for the volume dependence of the cohesive energy.

INTRODUCTION

One approach to the theoretical study of the properties of solids is through the assumption of a central pairwise potential function. This approach was initiated in the early part of this century by the work of Mie[1] and Grüneisen[2,3] and has been developed in subsequent years by various people. Although in the case of molecules a large number of potential energy functions have been examined,[4] for solids very few have been investigated.

The conditions that a satisfactory interatomic potential in a crystal must satisfy have been discussed recently by Girifalco and Weizer.[5,6] Varshni[4] has discussed the requisite shape of a potential for the case of diatomic molecules and the same general requirements also apply here.

Born and his collaborators[7] have made a detailed analysis of the stability of crystal lattices and have given conditions that must be satisfied among the elastic constants. For cubic crystals these are: (1) All elastic constants are positive. (2) $C_{11}-C_{12}>0$.

For metals, two potential energy functions have been used by different workers:

1. The Mie-Grüneisen potential,

$$\phi(r)=-a/r^m+b/r^n;\quad (m<n), \tag{1}$$

where a, b, and m, and n are constants. This potential function has been used extensively[8] by workers in solid-state physics. Grüneisen employed this function in his early investigations on the theory of solids. Fürth,[9] and more recently Dayal and Sharma[10] and Cole,[11] have discussed the applicability of the function for a number of solids. Fürth finds that this equation gives reasonable results for the equation of state, but the values of m and n determined by him violate the stability conditions for body-centered cubic metals. The causes of this anomaly have been discussed by Girifalco and Weizer.

2. The Morse potential,[12]

$$\phi(r)=D[e^{-2\alpha(r-r_0)}-2e^{-\alpha(r-r_0)}], \tag{2}$$

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[2] E. Grüneisen, Ann. Physik **39**, 257 (1912).

[3] E. Grüneisen, in *Handbuch der Physik*, edited by H. Geiger and K. Scheel (Verlag Julius Springer, Berlin, 1926), Vol. 10, Part 1.

[4] Y. P. Varshni, Revs. Modern Phys. **29**, 664 (1957); **31**, 839 (1959).

[5] L. A. Girifalco and V. G. Weizer, Phys. Rev. **114**, 687 (1959).

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[8] For a partial list of references, see J. R. Partington, *An Advanced Treatise on Physical Chemistry* (Longmans, Green and Company, New York, 1952), Vol. III.

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originally suggested for molecules, has also been used for solids.[13–16,5,6] In Eq. (2), α and D are constants and r_0 is the equilibrium interatomic distance.

Recently, Girifalco and Weizer[5,6] have made an extensive examination of the applicability of the Morse potential to 16 face-centered and body-centered cubic metals. They have calculated the equation of state and the elastic constants. Their results reproduce the general pattern of the experimental data, but it seems that a closer agreement might be achieved with a different form of potential.

Rydberg[17] proposed the following function for diatomic molecules:

$$U=-D_e[1+b(r-r_e)]e^{-b(r-r_e)}, \tag{3}$$

where D_e is the dissociation energy, r_e is the equilibrium internuclear distance, and b is a constant. This potential has the shape appropriate for a potential function and has been found to give better results than the Morse potential for a good number of diatomic molecules.[4]

It is of interest therefore to compare the applicability of the Morse and Rydberg functions to cubic metals. To this end we have calculated the elastic constants C_{11} and C_{12}, the equation of state and the variation of cohesive energy with volume using the Rydberg potential, for the same 16 cubic metals which were treated by Girifalco and Weizer using the Morse potential.

RYDBERG FUNCTION FOR SOLIDS

For solids, the Rydberg function may be written

$$\phi(r_{ij})=-D[1+b(r_{ij}-r_0)]e^{-b(r_{ij}-r_0)}, \tag{4}$$

where $\phi(r_{ij})$ represents the potential energy of two atoms i and j separated by a distance r_{ij}, b and D are constants, and r_0 is the equilibrium distance of approach of the two atoms.

The summation of Eq. (4) over a crystal consisting of N atoms is carried out by choosing one atom in the lattice to be the origin, adding together its interactions with all the other atoms in the crystal, and then multiplying by $N/2$. Thus, the total energy Φ is

$$\Phi=-\tfrac{1}{2}ND\textstyle\sum_j[1+b(r_j-r_0)]e^{-b(r_j-r_0)}, \tag{5}$$

where r_j is the distance from the origin to the jth atom. We shall find it convenient to express the results in terms of L and β defined by the equations

$$L=ND/2, \tag{6}$$

$$\beta=e^{br_0}. \tag{7}$$

Let us also write

$$r_j=[m_j^2+n_j^2+l_j^2]^{1/2}a=M_ja, \tag{8}$$

where m_ja, n_ja, l_ja are the position coordinates of any atom in the lattice and a is the lattice spacing. The volume per atom is then given by

$$V/N=ca^3, \tag{9}$$

where $c=4$ for body-centered crystals and $c=2$ for face-centered crystals.

The total energy is then

$$\Phi(a)=-L\beta\textstyle\sum_j[1+abM_j-br_0]e^{-abM_j}. \tag{10}$$

If a_0 is the equilibrium value of a at zero temperature, then

$$\Phi(a_0)=U_0(a_0), \tag{11}$$

where $U_0(a_0)$ is the energy of sublimation at zero temperature and pressure. We also have the relations

$$(d\Phi/da)_{a_0}=0, \tag{12}$$

and

$$1/K_{00}=V_{00}(d^2\Phi/dV^2)_{a_0}, \tag{13}$$

where V_{00} and K_{00} are volume and compressibility, respectively, at zero temperature and pressure. Using (9), the above equation may be written as

$$1/K_{00}=(1/9cNa_0)(d^2\Phi/da^2)_{a_0}. \tag{14}$$

Equations (9), (10), (11), and (14) may be combined to give

$$9V_{00}/K_{00}U_0(a_0)=-b^2a_0^2F(ba_0), \tag{15}$$

where

$$F(x)=\frac{-x(\sum_j M_je^{-M_jx})(\sum_j M_j^3e^{-M_jx})+x(\sum_j M_j^2e^{-M_jx})^2+(\sum_j M_je^{-M_jx})(\sum_j M_j^2e^{-M_jx})}{(\sum_j e^{-M_jx})(\sum_j M_je^{-M_jx})+x(\sum_j M_je^{-M_jx})^2-x(\sum_j e^{-M_jx})(\sum_j M_j^2e^{-M_jx})}. \tag{16}$$

The left-hand side of Eq. (15) contains experimentally known quantities, and the right-hand side is a function of ba_0 alone. The values of the ratio $V_{00}/K_{00}U_0(a_0)$ and those of $U_0(a_0)$ were taken from Fürth's paper.[9]

Equation (15) was solved for ba_0 by the Newton-Raphson method. The summations on the right-hand side were made, using an IBM 1620 computer, over a finite lattice for which m_j, n_j, and l_j each ranged from -10 to $+10$.

The other experimental data used here have been summarized in Table I together with the sources.[18–20]

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[19] *American Institute of Physics Handbook* (McGraw-Hill Book Company, New York, 1957).

[20] C. D. Hodgman, *Handbook of Chemistry and Physics* (Chemical Rubber Publishing Company, Cleveland, Ohio, 1961).

TABLE I. Data for metals.

Metals	a (10^{-8} cm)	Temperature (°K)	Thermal expansion data from reference	a_0 (10^{-8} cm)	Debye (°K)
fcc					
Pb	2.4751[a]	298	19	2.4569	88[e]
Ag	2.0431[a]	298	19	2.0340	189[e]
Ni	1.7618[a]	291	20	1.7571	370[e]
Cu	1.8073[a]	293	19	1.8011	301[e]
Al	2.0248[a]	298	19	2.0142	400[f]
Ca	2.791[a]	291	19	2.7764	226[e]
Sr	3.0424[a]	298	[d]	3.0267	170[g]
bcc					
Mo	1.5734[a]	293	19	1.5716	380[e]
W	1.5825[a]	298	19	1.5808	286[e]
Cr	1.4423[a]	293	19	1.4409	485[e]
Fe	1.4332[a]	293	19	1.4295	420[e]
Ba	2.5096[a]	≃298	19	2.4989	113[e]
K	2.672[b]	293		2.6125[a] (5°K)	92.1[e]
Na	2.1453[a]	293		2.1125[a] (5°K)	172[e]
Cs	3.0801[c]	293	19	3.0225[a] (5°K)	42[e]
Rb	2.85[a]	293		2.7925[a] (5°K)	58[e]

[a] Taken from reference 18.
[b] Taken from reference 19, Table 2e-2.
[c] Calculated from the value at 263°K given in reference 18 using thermal expansion data.
[d] Estimated.
[e] Taken from reference 6.
[f] Reference 19, Table 4f-8.
[g] M. W. Holm, Atomic Energy Commission Report IDO-16399 (unpublished).

Except for the alkali metals, a values are usually available at the room temperature. The values for a_0 were calculated from these by using thermal expansion data. Occasionally, the thermal expansion data were not available over the whole temperature range. In these cases, extrapolations were made. The values of the parameters in the Rydberg potential are recorded in Table II.

TABLE II. Parameters in the Rydberg function.

Metals	ba_0	br_0	β	b (10^8 cm^{-1})	D (10^{-14} erg)
fcc					
Pb	4.2410	6.3523	573.9	1.7262	41.02
Ag	3.9960	6.0669	431.2	1.9646	57.76
Ni	3.6654	5.6984	298.5	2.0861	75.50
Cu	3.5954	5.6234	276.9	1.9962	61.78
Al	3.4509	5.4724	238.1	1.7133	49.07
Ca	3.2996	5.3201	204.4	1.0791	29.71
Sr	3.2996	5.3201	204.4	1.0902	27.70
bcc					
Mo	3.4130	6.4187	613.3	2.1717	139.1
W	3.2696	6.2069	496.1	2.0683	175.6
Cr	3.2203	6.1351	461.7	2.2349	76.08
Fe	2.9068	5.6952	297.4	2.0334	74.69
Ba	2.4504	5.1174	166.9	0.9806	26.36
K	1.9474	4.5988	99.35	0.7454	10.56
Na	1.9134	4.5692	96.45	0.9058	12.40
Cs	1.8961	4.5543	95.03	0.6273	8.747
Rb	1.8254	4.4960	89.64	0.6537	9.166

The following physical properties were calculated.

A. Elastic Constants C_{11} and C_{12}

Born[7] has shown that for cubic monatomic crystals, whose atoms are all at rest and interact with a central pairwise force, the elastic constants can be calculated according to the equations,

$$C_{11}=\frac{a^4}{2v}\sum_i m_j^4 D_j^2\phi(r_j), \tag{17}$$

and

$$C_{12}=C_{44}=\frac{a^4}{2v}\sum_i m_j^2 n_j^2 D_j^2\phi(r_j), \tag{18}$$

where the operator D_j is defined by

$$D_j=\frac{1}{r_j}\frac{d}{dr_j}, \tag{19}$$

and $v=V/N$ is the volume per atom. For the Rydberg potential, Eqs. (17) and (18) yield

$$C_{11}=\frac{D\beta ab}{2v}\sum_i\left[br_0\frac{m_j^4}{M_j^3}+b^2r_0a\frac{m_j^4}{M_j^2}-b^2a^2\frac{m_j^4}{M_j}\right]e^{-baM_j}, \tag{20}$$

$$C_{12}=\frac{D\beta ab}{2v}\sum_i\left[br_0\frac{m_j^2n_j^2}{M_j^3}\right.$$

$$\left.+b^2r_0a\frac{m_j^2n_j^2}{M_j^2}-b^2a^2\frac{m_j^2n_j^2}{M_j}\right]e^{-baM_j}. \tag{21}$$

The calculated values and some experimental values are shown in Table III.

TABLE III. Elastic constants and the Grüneisen constant γ. All elastic constants are in units of 10^{11} dynes/cm^2.

	C_{11} (theoret)	C_{11} (expt)	C_{12} (theoret)	C_{12} (expt)	γ
fcc					
Pb	5.6082	6.7 (0°K)[a]	3.8000	5.7 (0°K)[a]	2.518
Ag	12.808	13.15 (0°K)[b]	8.8725	9.73 (0°K)[b]	2.334
Ni	23.256	26.12 (0°K)[b]	16.606	15.07 (0°K)[b]	2.232
Cu	17.269	17.62 (0°K)[b]	12.410	12.49 (0°K)[b]	2.063
Al	9.3574	12.30 (0°K)[b]	6.8140	7.08 (0°K)[b]	2.682
Ca	2.0615		1.5220		1.266
Sr	1.4836		1.0953		1.047
bcc					
Mo	29.315	46.0[a]	28.379	17.9[a]	2.056
W	35.056	50.1[a]	33.265	19.8[a]	2.018
Cr	19.809		18.670		1.041
Fe	18.393	23.7[a]	16.618	14.1[a]	2.209
Ba	1.0873		0.9281		0.721
K	0.3407	0.459 (77°K)[a]	0.2755	0.372 (77°K)[a]	1.232
Na	0.7514	0.945 (90°K)[a]	0.6057	0.779 (90°K)[a]	1.137
Cs	0.1803		0.1451		0.818
Rb	0.2360		0.1886		0.924

[a] J. deLaunay, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1956), Vol. 2.
[b] G. A. Alers and J. R. Neighbours, Revs. Modern Phys. **31**, 675 (1959).

B. Equation of State

According to the Debye model, the Helmholtz free-energy A is given by the relation

$$A=\Phi+3NkT\ln(1-e^{-\theta/T})-NkTD(\theta/T), \qquad (22)$$

where k is the Boltzmann constant, T is the absolute temperature, θ is the Debye temperature and

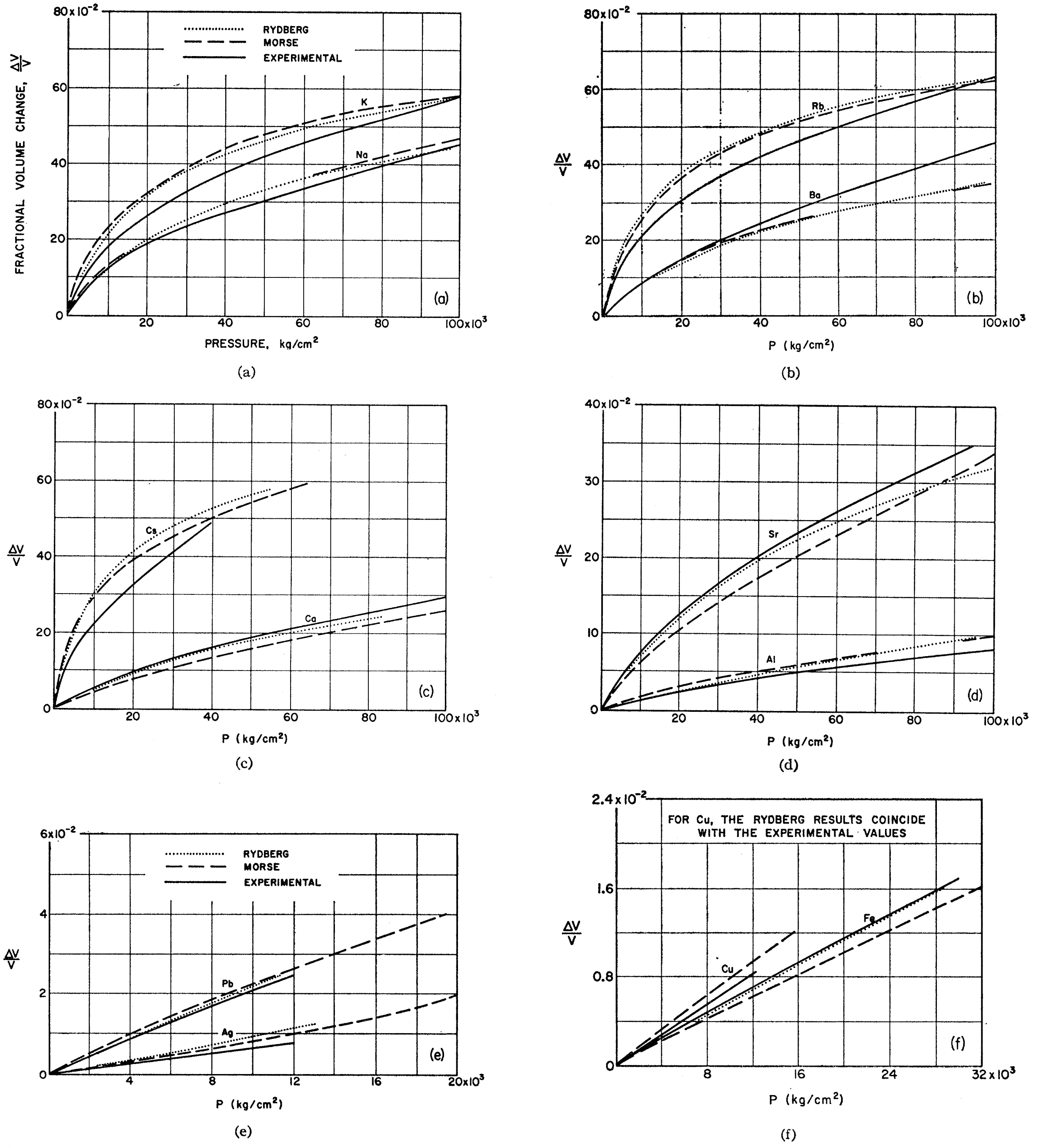
$$D\left(\frac{\theta}{T}\right)=3\left(\frac{T}{\theta}\right)^3\int_0^{\theta/T}\frac{x^3}{e^x-1}dx. \qquad (23)$$

Then the pressure P is

$$P=-\left(\frac{\partial A}{\partial V}\right)_T=\frac{1}{3Nca^2}\frac{d\Phi}{da}+\frac{3\gamma RT}{V}D\left(\frac{\theta}{T}\right), \qquad (24)$$

where γ is Grüneisen's constant.

Equation (24) was solved for γ, giving P the value of one atmosphere and V the value Nca^3, where a is the lattice parameter determined experimentally at temperature T. The values of a and T together with their

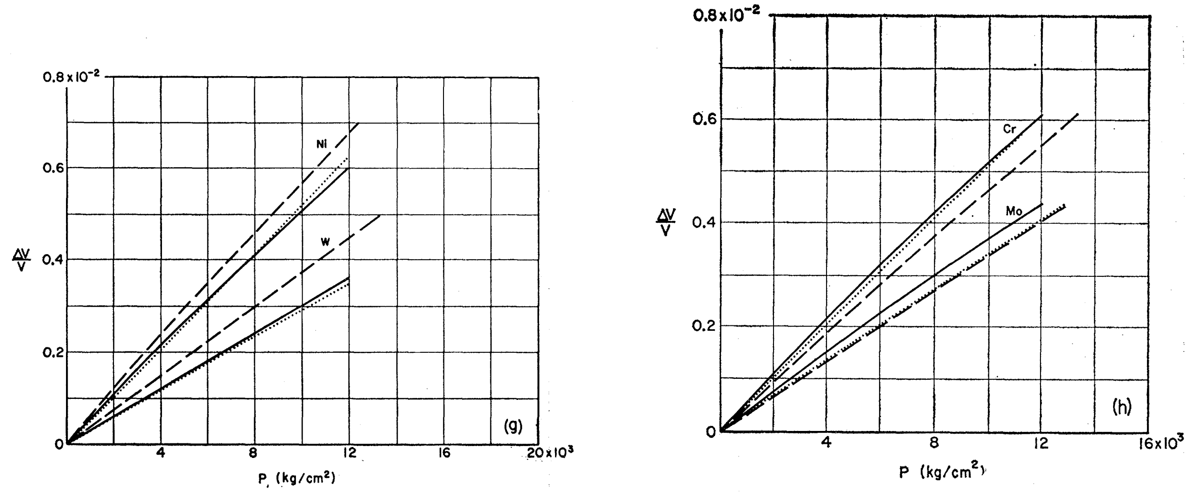


(a) (b)

(c) (d)

(e) (f)

Caption is on opposite page.



(g) (h)

FIG. 1. (a) to (h) Equation-of-state curves for various metals. Where Rydberg and Morse results overlap (e.g., Na, Ba, Al), only one curve has been shown.

sources are given in Table I, and the values of γ so determined are given in Table III.

The predictions by the Rydberg potential for the equations of state for the various metals at the temperatures T above are shown in Figs. 1(a) to 1(h), together with the experimental data of Bridgman[21] and the Morse potential curves, as given by Girifalco and Weizer.[6]

C. Volume Dependence of the Cohesive Energy

Recently, Benedek[22] has deduced the volume dependence of Φ from the shock wave data obtained by Walsh *et al.*[23] He has presented results for six metals, including four in our list. The theoretical dependence of Φ on volume for these metals was calculated from Eq. (10), again at the temperatures indicated in Table I. Corresponding results were derived for the Morse potential, using the parameters determined by Girifalco and Weizer.[24] The experimental data and the Rydberg and Morse predictions are presented in Figs. 2(a) to 2(d). The value of Φ_0 (value of Φ at one atmosphere pressure and temperature given in Table I), for the theoretical curves refers to the "theoretical" value Φ_0 for each potential.

DISCUSSION

Elastic Constants

It will be noticed that the theoretical values of the elastic constants satisfy Born's requirements. Experimental values of C_{11} and C_{12} at low or zero temperature are available only for four metals in the list; the theoretical values are for zero temperature. The results obtained here are not very different from those obtained by the Morse potential.[5,6] In general the results are reasonable, but for Mo, W, K, and Na the errors are rather large.

Equation of State

The Rydberg predictions are considerably better than the Morse predictions for Cu, Fe, Ni, W, and Cr, slightly better for K, Na, Ca, Sr, Al, and Pb, and worse than the Morse predictions for Rb, Cs, and Ag. For Ba and Mo the predictions differ from the experimental results by about the same amount.

For the alkali metals, neither the Rydberg nor the Morse[5,6] nor the Mie-Grüneisen (see Figs. 6 and 7 of Fürth[9]) potential gives satisfactory results, but it is interesting that for such metals all three potentials show the same general behavior. At low pressures the calculated values of $\Delta V/V$ are less than the observed ones, and at high pressures they are greater.

Variation of Cohesive Energy with Pressure

The Rydberg results are better than the Morse ones for Al and Ni, worse for Ag, and about the same as the Morse results for Cu. The comparison of theory with experiment may not be meaningful in the case of Al since the experiments were done using 24ST aluminum which is an alloy of 4.5% Cu, 0.6% Mn, and 1.5% Mg.

The case of Cu is interesting. The equation-of-state curve for this metal is fitted much more closely by the Rydberg curve than by the Morse curve. The pressure dependence of the cohesive energy indicates, at high pressures, a preference for the Morse version. Now the (Φ,V) curve depends directly upon Φ whereas the (P,V)

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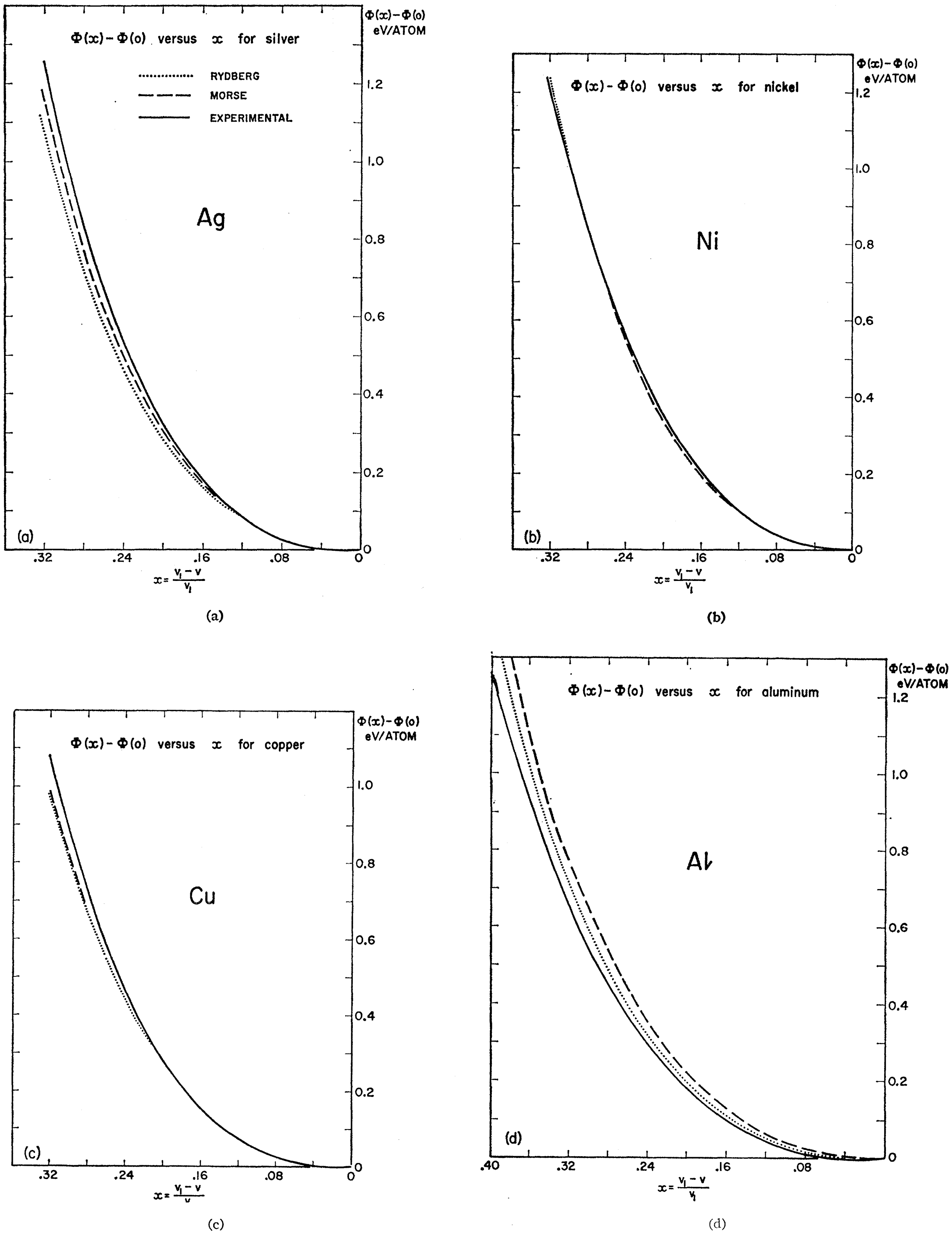
[21] P. W. Bridgman, Proc. Am. Acad. Arts Sci. **58**, 165 (1923); **60**, 305 (1925); **60**, 385 (1925); **74**, 11 (1940); **74**, 425 (1942).

[22] G. B. Benedek, Phys. Rev. **114**, 467 (1959).

[23] J. M. Walsh, M. H. Rice, R. G. McQueen and F. L. Yarger, Phys. Rev. **108**, 196 (1957).

[24] We are indebted to Dr. Weizer for kindly supplying us with a list of the values of a_{300} used by them.



(a) (b)

(c) (d)

FIG. 2. (a) to (d) The change in the cohesive energy as a function of the volume decrement $x=(v_1-v)/v_1$. v_1 is the volume at $P=1$ atm and T = temperature given in Table I for Rydberg results, 300°K for Morse results and 293°K for experimental values. (For all practical purposes these differences of temperatures may be disregarded.) $\Phi(0)$ is the cohesive energy at $v=v_1$.

curve depends upon $d\Phi/dV$; in other words the (P,V) relation is a more sensitive test for correct potential shape. Thus, for copper also, the Rydberg potential more closely approximates the interatomic interaction.

Choice of Grüneisen's Constant

The value of the Grüneisen constant γ affects the (Φ,V) curves of Benedek quite strongly. In fact, the discrepancies between Benedek's results and the theoretical results could all be removed by judicious adjustment of γ by at most 20% from the values taken here. It is difficult to say what is the correct value for γ and our values, having been obtained by a different method, differ slightly from Benedek's. The replacement of our values for γ by those of Benedek would not, however, always improve the agreement.

Girifalco and Weizer have found $\gamma \approx 0$ for Mo, W, and Cu, and a very small value for γ for Fe. The method by which γ is determined in this work gives a result which is rather sensitive to the thermal expansion of the lattice constant, $(a_{300}-a_0)$. The truncation error in the experimental data which they have used for a_0 and a_{300} is of the same order as $(a_{300}-a_0)$ for these metals,[25] so that the expansion property was "lost" and γ turned out to be nearly zero.

ACKNOWLEDGMENTS

The authors are most grateful to Dr. T. Y. Wu for his kind interest in the work. Thanks are also due to Dr. S. D. Baxter for making available the facilities of the Computation Center.

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[25] We may here recall the approximate rule that the total relative thermal expansion from 0°K to the melting point of the metal, $\Delta l/l_0 \approx 0.027$. Mo, W, Cu, and Fe are all high-melting-point elements.

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Electronic Structures of Manganese-Substituted Iron-Cobalt Alloys

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The substitution of manganese in the body-centered cubic iron-cobalt alloys has been shown to either decrease or increase the mean saturation moment of the ternary alloys. On the basis of a modified band model, this varying effect of Mn may be ascribed to the occupancy of the localized *d* states of the Mn atoms. An increase in the moment is observed when Mn is added to alloys with the atom ratio of Co:Fe$\geqslant$1 in which the localized *d* states are just half filled according to Hund's rule. At lower ratios, however, the localized *d* states become first more than half empty and eventually nonexistent. The rates at which the substitution of Mn decreases the mean moment of the latter alloys change accordingly. The proposed electronic structures are consistent with observations that the increase of residual resistivities of Fe-Co alloys caused by 1 at.% Mn also varies sensitively with the Co:Fe ratio of the host alloy.

A RECENT study has shown that the substitution of manganese in the equiatomic iron-cobalt alloy leads to an increase of the mean saturation moment of the ternary alloy.[1] Furthermore, this effect was found to be unique to Mn because all other transition elements reduced the mean amount. A search to find similar behavior by substitution of such elements as Cr, which resembles Mn magnetically in the metallic state, and Rh, Ir, and Pt, which exhibit similar increasing effects on the saturation moment of Fe, has proved to be futile. To attain this unique effect, Mn must fulfill two conditions: Each Mn atom must "carry" a larger moment than the average moment of the parent Fe-Co alloy and its atomic spin must be aligned in a direction parallel to that of the Fe and Co atoms.

An interesting aspect of these findings is related to the fact that the binary alloy of Fe and Co containing 35 at.% Co has hitherto displayed the highest saturation magnetization among all ferromagnetic materials at room temperature. Should the substitution of Mn in alloys close to this composition continue to increase the moment, a new maximum value of saturation magnetization would be reached. It has not been possible to make a trustworthy prediction, however, because of numerous conflicting factors. At first glance, such an effect seemed quite feasible since the magnetic properties of Mn-substituted FeCo alloys were found to be strikingly similar to those of the dilute Ni alloys, despite the difference in their crystal structures. Accordingly, Fe-Co alloys with an atom ratio of Co:Fe$>$35:65, which lie on the right side of the peak of the familiar Pauling-Slater curves[2] might provide an electronic structure of the right type for an increase in the moment to occur when Mn is incorporated. On the other hand, a deduction of this sort could be invalidated

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[1] C. W. Chen, Phil. Mag. (to be published).

[2] R. M. Bozorth, *Ferromagnetism* (D. Van Nostrand Company, Inc., Princeton, New Jersey, 1951), 1st ed., p. 441.