A NEW MODEL OF METALLIC STRUCTURE AND BONDING

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This paper briefly describes the current physical model of metallic structure and bonding. An alternative model is introduced. It shows that the new model, which calculated internuclear distances of Group 1 and Group 2 crystalline binary salts to a remarkable degree of accuracy, can be applied to calculate metallic radii (equal to half the internuclear distances) of Group 1 and Group 2 metals precisely. A simple expression previously used to calculate lattice energies using the soft-sphere radii concept is applied to calculate enthalpies of formation of Group 1 and Group 2 metal ions and results compare well with observed values. A few of the limitations of the current models are described and properties of metals which can be accounted for by the soft-sphere model are discussed. The work functions of Group 1 and Group 2 metals are shown to be inverse functions of the soft sphere ionic radii.

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Introduction

Living organisms that made up much of nature are composed of molecules consisting of both metallic and non-metallic elements. An understanding of how atoms bond together is therefore very important in the study of natural science. However, even though over 70% of elements are metals, metallic bonding and structure are still not well understood. It is well known that most metals have hexagonal, cubic close packed or body centred cubic structures.[1] Paul Drude,[2] in 1900, was the first to propose the "free electron" model for electricity conduction in metals and metallic bonding. He suggested that in crystalline metals, positive ions were surrounded by an "electron gas". Estimates of the number of "free" electrons in a metal have also been described since the early twentieth century.[3] Other common accounts on electrical conductivity in metals include band theory,[4] which postulates the existence of energy bands in solids that influence the behaviour of electrons. In his book, *The Nature of the Chemical Bond*,[5] Pauling provided extensive details on the closest packing of spheres, metallic orbitals, metallic valencies, bond lengths, and bond numbers in metals. However, some other common properties of metals (see below) were not discussed in his book nor were they accounted for in the models of metallic structure and bonding.

In recent years, the theory of metallic bonding and structure has shown little advance on the "free electron" model. It is still very common to describe metallic structures and metallic bonding as "bonding which involves the delocalisation of electrons throughout the metal solid"[6] or as "metal cations in a sea of electrons".[7] It has been demonstrated[8] that the standard model used to describe metallic structure and properties is inadequate. The electrical conductivity of copper is higher than most metals. It has been reported that graphene, which is covalently bonded carbon, is a much better conductor of electricity than copper.[9] Surely, this must mean that electrons in a metallic bond are not "free" and better a model for describing metallic structure and bonding is needed.

Theory

We propose here an alternative "soft-sphere" model which considers that in a metallic crystal, the outermost electron(s) in each atom is/are not exactly "free" nor completely delocalised. The outermost electron(s) is/are separated/detached from the atom which forms a positive ion with one or more of the "detached" electron(s) behaving like negative ions. The detached outermost electron(s) can occupy certain equivalent positions that are at the midpoints between the nearest neighbours of the positive ions (similar to ionic crystals, where positive ions occupy positions between negative ions) but can move within these midpoint positions in a unit cell. For the remainder of this paper, these midpoint positions will be called "midpoint sites". Depending on the Group and metallic structure (hcp, fcp or bcc) the most likely number of outermost electrons detached from each individual metal atom range from 1 to a maximum of 5. The soft-sphere ionic radii of Group 1 and Group 2 metals (excluding beryllium) have already been determined very accurately[10] and in this work we use members of these 2 Groups to demonstrate the soft-sphere model of metallic structure and bonding.

Consider any crystal with a hexagonal (hcp) or cubic closed pack (fcp) structure of identical atoms, each atom has 12 co-ordination or 12 closest neighbours and in a body centred cubic (bcc) each atom has 8 closest neighbours. There are 2 atoms in each unit cell in a body centred cubic, 6 atoms in each unit in a hexagonal closed pack and 4 atoms per unit cell in a face centred cubic[11] crystal.

At room temperature, all Group 1 metals have a bcc structure and each atom has 1 outermost electron. In the soft-sphere model of metallic structure/bonding, each atom has a single outermost electron which it loses to form a unipositive (1+) ion. Each positive ion has 8 nearest identical neighbours of positive ions. Hence there are 8 equivalent sites that are midpoint between the internuclear distance of a positive ion and its 8 neighbouring positive ions. The outermost electron which is detached from the atom can occupy and move around any one of these 8 midpoint sites in a unit cell at any *one* time. Since there are 2 positive ions in each unit cell, there are only 2 detached electrons in each cell and, therefore, at any *one* time only 2 of the 8 (or a

Table 1. Soft-sphere ionic radii and atomic/metallic radii of Group 1 and Group 2 metals

Element	Soft-sphere radii Å	Cell constants		Internuclear separation Å	Atomic radii Å
(A)	(B)	a (C)	c (D)	(E)	(F)
Li	1.094	3.509		3.039	1.520
Na	1.497	4.291		3.716	1.858
K	1.971	5.321		4.608	2.304
Rb	2.160	5.700		4.937	2.469
Cs	2.368	6.176		5.348	2.674
Be	0.750	2.286	3.584	2.256	1.128
Mg	1.282	3.209	5.211	3.203	1.602
Ca	1.657	5.580		3.946	1.973
Sr	1.861	6.086		4.303	2.152
Ba	2.084	5.023		4.350	2.175

quarter of) midpoint sites are occupied and the rest are vacant and under certain conditions can be occupied by other detached electrons moving in from other unit cells.

Group 2 metals present a slightly more complicated problem in that beryllium and magnesium have hcp structures at room temperature whereas calcium and strontium possess a ccp structure and barium has a bcc structure respectively.[12] In any Group 2 metal crystal, each metal atom loses its 2 outermost electrons to form a dipositive (2+) ion. In a barium crystal (with a bcc structure), each dipositive ion has 8 identical nearest neighbours of positive ions. Since there are 2 positive ions in each unit cell, there are 4 detached electrons in a cell. Hence, at any *one* time there are 4 electrons occupying the 8 equivalent midpoint sites between the positive ions, which mean that only half of the 8 sites are occupied. In calcium and strontium (with fcp structures), there are 4 positive ions and 8 detached electrons (with each atom losing 2 electrons) occupying 24 equivalent midpoint sites per unit cell. Thus, at any *one* time, only a third of the sites are occupied. Similarly for beryllium and magnesium (hcp), with 6 positive ions and 12 detached electrons per cell, at any *one* time only a third of the 36 sites are occupied.

The atomic/metallic radius of a metal atom (which is half the internuclear distance between the nearest neighbours) or distance between the centres of the positive ion and the detached outermost electron, just as in the case of an ionic crystal, can be calculated from the relationship:[10]

$$S(\text{calc})^k = [M]^k + [e]^k$$

where S is the radius of the metal atom, $[M]$ is the ionic radius of the positive ion, $[e]$ is the "radius" of the midpoint site containing the detached electron and the exponent k is defined in previous work.[10]

Column B of Table 1 shows the soft-sphere ionic radii determined by previous work,[10] the observed unit cell constants[13] quoted to appropriate number of decimal places (after taking account of the size of experimental uncertainties) are shown in columns C and D. Internuclear separations (between nearest neighbours) calculated from unit cell constants of Group 1and Group 2 metals are listed

in Column E. The distances between the centres of the positive ions and space occupied by the detached electron (or the metallic radii of the atoms) are given in Column F of the table. All figures are given in Angstrom (Å) units, where 1 Å equals 10^{-10} m. Since the soft-sphere radius of beryllium has not been determined previously, for this work we have extrapolated from the known soft-sphere radii of Group 2 metals to obtain an estimate of 0.75±0.02 Å.

We have made an empirical assumption that (a) e, the "radius" of the midpoint sites containing the separated/detached electron(s) and (b) the exponent k, are different for Group 1 and Group 2 metals because of the difference in charge on the positive ion, the value of k may also be influenced by the number of nearest neighbours. However, in order to limit the number of values of k in the calculations we have used k equals 1.5 for Group 1 and k equals 1.3 for Group 2. All Group 1 metals have 1 outermost electron and all have bcc structure, therefore we assume that the value of e is the same for all 5 elements is the same. By trial and error, an estimated value of 0.8 is obtained to give the best results. For Group 2 metals, beryllium, magnesium, calcium and strontium have 12 co-ordination and we assume that e is the same for all 4 elements and has the value of 0.565. Barium has 8 co-ordination and the value of e is 0.22.

Results and Discussion

The observed metallic radii (or half the internuclear distance) of Group 1 and Group 2 metals are shown in Column B and the calculated radii in Column C of Table 2, respectively. As shown in Column D of the table, differences between the observed and calculated are in all cases less than 0.01 Å. This is very good agreement since the experimental uncertainties of the appropriate cell constants can be as great as 0.01 Å (as in the case of calcium). The root mean square (r.m.s.) deviation δ is given at the top of Table 2.

The lattice energy of a compound is the energy change when 1 mole of the compound at one atmospheric pressure is converted into gaseous positive and negative ions which are separated from each other at infinity. Lattice energies

can be calculated using the Born-Haber cycle or from equations such as the Born-Lande/Born-Mayer equations or the improved Kapustinskii equation. The enthalpy of formation of a positive univalent Group 1 or divalent Group 2 metal ion is the energy change when 1 mole of the crystalline metal is converted into one mole of the gaseous positive metal ion separated from 1 (in the case of Group 1) and or 2 (in the case of Group 2) moles of electrons. In the case of a metal solid, the detached electron is treated as be equivalent to the "negative ion" bonded to positive ion. Under this assumption, the energy change when a crystalline metal is converted into 1 mole of the gaseous positive ion and the respective number of moles of "electrons" can then be regarded as equivalent to the lattice energy of an ionic crystal. The enthalpies of formation of gaseous metal atoms for Group 1 and Group 2 metals are shown in Column B of Table 3. The first and (for Group 2 only) second ionisation energies of the metals are listed in Columns C and D respectively. Ionisation energies are converted from eV (electron volts) to kJ mol^{-1} by the relationship of 1 eV = 96.49 kJ mol^{-1}. The enthalpies of formation of the appropriate gaseous ions are shown in Column E of the Table. All ionisation energies, enthalpies of formation, work functions, bond dissociation energies and enthalpies of formation that are utilised in this work are quoted from the *CRC Handbook of Chemistry and Physics*.[14]

Table 2. Comparison of observed and soft-sphere atomic/metallic radii of Groups 1 Group 2 metals

Element	Observed Å	Soft-sphere calculated, Å	Obs – Calc Å
(A)	(B)	(C)	(D)
Li	1.520	1.512	0.008
Na	1.858	1.865	–0.007
K	2.304	2.298	0.006
Rb	2.469	2.474	–0.005
Cs	2.674	2.669	0.005
Be	1.128	1.124	0.004
Mg	1.602	1.610	–0.008
Ca	1.973	1.964	0.009
Sr	2.152	2.158	–0.006
Ba	2.175	2.175	0.000

^{a}The root mean square (r.m.s.) deviation is δ=0.00629.

We have previously developed a simple expression to calculate lattice energies of Group 1 and Group 2 salts.[10] The results produced agreed well with lattice energies calculated from the Born-Haber cycle. We reproduce the expression here as follows:

$$E_L = \frac{R \frac{H_0}{M} M^{k-1}}{\left(X^{k-1.3333}\right)\left(0.5^{0.33333}\right)\left(\sum Q_i^2\right)}$$

where

E_L is the lattice energy,

R is the Rydberg constant for infinite mass converted to kJ mol^{-1},

H_o is the classical Bohr radius,

M is the size of the cation,

X is the size of the anion and

Q_i is the charge on the ions.

Hence, for sodium chloride $\sum Q_i^2 = 1 + 1 = 2$ and for calcium chloride it is $4 + 1 + 1 = 6$ etc. $\sum Q_i^2$ is the sum of all the squares of the charges on the ions, since the higher the charge on the ions the more electrons need to be removed from the overlap region and the more energy is required to separate them. R is the amount of energy needed to remove an electron from a species the size of a hydrogen atom, (H_o/M) provides a ratio of the distance of the electron from the nucleus, since the greater the size the less is the energy needed to remove the electron. $(M^{k-1})/(X^{k-1.3333})$ gives an approximation of the overlap, this is multiplied by a factor which is approximated to ($\frac{1}{2}$ $^{0.33333}$) because the electron is not removed to infinity away from both ions (but rather removed from the overlap region).

We have applied the same expression given above with only two very minor differences to calculate the enthalpies of formation of Group 1 and Group 2 metal ions. X, rather than being the size of an anion, is the "radius" of the space occupied by the electrons detached from the metal atom. In the above expression, $\sum Q_i^2$ is the sum of the squares of the charges on the positive ions and the detached outermost electron(s). For Group 1 metals, Q_i^2 of the positive ion is 1 since each ion has a +1 charge. However, Q_i^2 of the outermost electron is only 0.25 since at any *one* time only a quarter of the available midpoint sites is occupied. Hence, $\sum Q_i^2$ is equal to 1 + 0.25 (1.25) for all the Group 1 metals. For elements of Group 2, Q_i^2 for the positive ions in all cases is 4 (since they are all dipositive). However, there are 2 different values of $\sum Q_i^2$. For the first 4 elements, only a third of the sites are occupied by electrons. Hence, Q_i^2 for the electrons is $(1^2 + 1^2)\cdot0.3333$ and $\sum Q_i^2$ is approximately 4.7. As for barium, since half of the available midpoint sites are occupied at any *one* time, Q_i^2 for the electrons is $(1^2 + 1^2)\cdot0.5$. Hence $\sum Q_i^2$ has a value of 4 plus 1 which equals 5. The soft-sphere calculated values are shown in Column B of Table 4. The conventionally produced results are shown in Column C and the absolute percentage differences are shown in Column D of the Table. Differences between observed and soft-sphere calculated are less than 10% for all cases.

Metals are malleable and ductile because not all available midpoint sites are occupied by electrons. When a metal is twisted or bent, electrons can move from 1 site to another within a unit cell, allowing the shape of the metal to change without any bonds being broken. It has been shown that when sodium is under very high pressure the resistance of the metal increases drastically and acts more like an insulator[15] than a conductor of electricity.

This behaviour cannot be easily explained by the standard model. However, this can be accounted for by the soft-sphere model. When a metal is under high pressure, the shape and size of the unit cell changes. This reduces the volume of the site(s) occupied by the detached electron(s) and if the volume is reduced sufficiently it can no longer be occupied. When the shape of the unit cell changes, the distance between some of the midpoint sites may increase to such an extent, the detached electrons occupying those sites may no longer be able to move from one site to another.

Table 3. Enthalpies of formation of Group 1 and Group 2 metal ions

Element	Enthalpies of formation of M(g) kJ mol^{-1}	Ionisation energies		Enthalpies of formation M$^+$(Group 1)/M^{2+}(Group 2) kJ mol^{-1}
		1st eV (kJ mol^{-1})	2nd, eV (kJ mol^{-1})	
(A)	(B)	(C)	(D)	(E)
Li	159.3	5.392 (520.25)		679.55
Na	107.5	5.139 (495.87)		603.37
K	89.0	4.341 (418.83)		507.83
Rb	80.9	4.177 (403.05)		483.95
Cs	76.5	3.894 (375.72)		452.22
Be	324.0	9.323 (899.55)	18.211 (1757.19)	2980.74
Mg	147.1	7.646 (737.78)	15.035 (1450.75)	2335.64
Ca	177.8	6.113 (589.86)	11.872 (1145.50)	1913.16
Sr	164.4	5.695 (549.50)	11.030 (1064.29)	1778.19
Ba	180.0	5.212 (502.87)	10.004 (965.27)	1648.14

Table 4. Comparison of observed and calculated enthalpies of formation of M$^+$(Group 1)/M^{2+}(Group 2)

Element	Observed kJ mol^{-1}	Soft-sphere calcd., kJ mol^{-1}	Abs. difference, %
(A)	(B)	(C)	(D)
Li	679.55	683.56	0.6
Na	603.37	584.35	3.2
K	507.83	509.26	0.3
Rb	483.95	486.47	0.5
Cs	452.22	464.61	2.7
Be	2980.74	3108.22	4.3
Mg	2335.64	2135.67	8.5
Ca	1913.16	1784.55	6.7
Sr	1778.19	1645.25	7.5
Ba	1648.14	1569.24	4.8

Hence, if there are no vacant sites which can facilitate electron movement or if the electrons cannot move between sites in neighbouring unit cells the metal becomes a virtual insulator. Assuming that in a metal the positive ions are truly surrounded by "a sea of electrons", then it may be reasonable to assume that the work function of any metal is not a significant value since electrons from the "electron sea" can move freely to the surface of the metal. However, the work functions of some metals are much higher than some bond dissociation energies and ionisation energies. For example, the work function of beryllium is 480.5 kJ mol^{-1} and the bond dissociation energy of C-H is 338.4 kJ mol^{-1}, that of Cl-Cl is just 242.4 kJ mol^{-1} and that of Ca-Ca is only 16.5 kJ mol^{-1}. The first ionisation energy of caesium is 3.8939 eV (or 375.6 kJ mol^{-1}).

In the soft-sphere model, the separated/detached outermost electrons can only occupy midpoint sites between 2 positive ions inside a unit cell, which means that there is almost no outermost electron on the surface of the metal. Hence, energy has to be expended to draw those electrons onto the surface of the metal. The work function is an inverse function of the soft-sphere radius since the further the outermost electron(s) is/are from the nucleus of the positive metal ion the less energy is required to draw it/them to the surface. The work functions of the Group 1 and Group 2 metals can be approximated by the following simple expression:

$$W = \frac{C}{R^{0.5}}$$

where

W is the work function,

R is the soft-sphere ionic radius and

the constant C is 3.08 for Group 1 and 3.85 for Group 2 metals.

Column B of Table 5 lists the soft-sphere radii, Column C shows the work functions (in eV) calculated by the above formula, Column D shows the observed work functions in eV. The absolute percentage differences are shown in Column E of Table 5. With the exception of beryllium, all values agree to better than 90%.

The detached electrons can move around different mid-point sites inside a unit cell. As discussed above, only some of those sites are occupied at any *one* time. Since all unit cells in a metal are identical and there are vacant sites which facilitate electron movement, a very small potential difference between the ends of a metal strip or wire can generate an electron flow along the wire (i.e. an electric current). When a metal wire with an electric current flowing through it is placed in a transverse magnetic field, a potential difference is developed across the wire at right angles to both the field and the length of the wire.[16] This is known as the Hall effect.[17] It happens because the magnetic field at right angles to the length of the wire creates a force acting on each electron and since, within every unit cell, there are many vacant sites between the positive ions for the electrons to occupy, detached electrons can move across at right angles to the length of the wire and congregate into more sites on one side of the wire than the other, thus developing a potential difference across the cross section of the wire and at right angles to the field and length of the wire.

Enthalpies of fusion of most halides, oxides and other binary compounds can be many times higher than that of the metals. For example, Column B of Table 6 lists the enthalpies of fusion of Group 1 and Group 2 metals, Columns C and D show the enthalpies of the chlorides and oxides of those metals respectively.

Table 5. Observed and Soft-sphere calculated work functions of Group 1 and Group 2 metals

Element	Soft-sphere radii Å	Soft-sphere calculated, eV	Observed work functions, eV	Abs. difference %
(A)	(B)	(C)	(D)	(E)
Li	1.094	2.94	2.93	0.5
Na	1.497	2.52	2.36	6.7
K	1.971	2.19	2.29	4.2
Rb	2.160	2.10	2.26	7.3
Cs	2.368	2.00	1.95	2.6
Be	0.750	4.45	4.98	10.7
Mg	1.282	3.40	3.66	7.1
Ca	1.657	2.99	2.87	4.2
Sr	1.861	2.82	2.59	8.9
Ba	2.084	2.67	2.52	5.8

Table 6. Enthalpies of fusion of Group 1 and Group 2 metals and some halides

Element	Metal kJ mol⁻¹	Chloride kJ mol⁻¹	Oxide kJ mol⁻¹	Nr of times difference between chloride/metal	oxide/metal
(A)	(B)	(C)	(D)	(E)	(F)
Li	3.00	19.80	35.60	6.6	11.9
Na	2.60	28.16	47.70	10.8	18.3
K	2.34	26.28	27.00	11.2	11.5
Rb	2.19	24.40	20.00	11.1	9.1
Cs	2.09	20.40	20.00	9.8	9.6
Be	7.90	8.66	86.00	1.1	10.9
Mg	8.48	43.10	77.00	5.1	9.1
Ca	8.54	28.05	80.00	3.3	9.4
Sr	7.43	16.22	81.00	2.2	10.9
Ba	7.12	15.85	46.00	2.2	6.5

The number of times that the enthalpies of the chlorides and oxides are greater than that of the metals (enthalpy of chloride/oxide divided by enthalpy of respective metal) are listed in Columns E and F respectively. These figures clearly show that, with the exception of beryllium, the enthalpies of fusion of the chlorides and oxides are at least twice or up to 18 times the enthalpy of the respective metal.

This cannot be easily accounted for by the standard model even if it is assumed that the bond formed between the metal and chlorine/oxygen is much stronger than the bonding in the metal itself. However, as we have already shown above, in every unit cell in a metal crystal, each positive ion is surrounded by only 2 (in Group 1) or 4 (in Group 2) detached electrons in the midpoint sites between the positive ions. Whereas, in the halide or oxide crystals, every positive ion/negative ion (depending on the structure of the crystal) is always surrounded by more than twice number of oppositely charged ions. So for example, in sodium chloride each sodium ion is surrounded by 6 chloride ions and vice versa. This means that, for every mole of metal, fewer bonds are made when a metal solidifies, but many more bonds are made when a metal halide or oxide solidifies. Beryllium chloride is an exception because its structure is different from other chlorides and oxides (beryllium chloride possesses a chain structure[18] whereas most other halides and oxides of Groups 1 and 2 metals have typical crystal structures such as the NaCl, wurtzite, rutile or fluorite structures).

It has been suggested by some authors that the number of free (or loosely attached) electrons in a metal lies in the range of 1 to 3.5 per atom.[19] Calculations have also shown that only a very small percentage of electrons in a metal are "free".[20] This can easily be accounted for by the soft-sphere model. There are only a limited number of sites that the "free" electrons can occupy. For example, in a body centred cubic, there are only 8 available midpoint sites for the detached electrons and since there are 2 atoms per unit cell the maximum number of electrons that can be detached from each atom is 4. Hence, it is not surprising that the number of "free" electrons per atom is limited to a very low number.

Conclusion

In contrast to the standard "electron sea" model, we are able to show that, for Group 1 and Group 2 metals, the atomic/metallic radii and enthalpy of formation of the positive ions calculated with the soft-sphere model and soft-sphere radii give very good agreement with experimental values. We also showed that the work functions of Group 1 and Group 2 metals are inverse functions of the soft-sphere radii. Additionally, the soft-sphere model can (in a qualitatively manner) account for the differences between the enthalpies of fusion of metals and their halides and oxides. The changes in resistance of a metal under pressure can also be interpreted by the soft-sphere model.

There is strong evidence that the soft-sphere model is a more realistic representation of the structure and bonding of metals than the "electron sea" model.

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