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Stereochemistry of Ionic Solids J. Stereochemistry of B-Subgroup Ions. Structure and Bonding in Organometallic Compounds. Reactivity and Reaction Pathways of Organometallic Compounds. Applications of Organometallic Reagents in Research and Technology. Present Trends in Organometallic Research. Sulfur Oxidation States Containing Fluorine. Perfluoroalkanesulfonic Acids. Sulfur Imides Derived from Sulfur Dioxide via the Sulfur Trioxide Route. Sulfur Imides Derived from Sulfur Trioxide. Amides of Oxyacids of Sulfur VI. Survey of the Actinide Element Halides. Properties of Halides Elemental Sulfur IV. The Sulfur Hydrides and Halides VI. Sulfur Chains Terminated by Cyano Groups. Comments on Structural Evidence. Sulfur Chains Terminated by Sulfonate Groups: Reactions of the Boranes and Their Derivatives I Mass Spectrometry in Nuclear Chemistry H. Neutron Capture Cross Sections. The Structure of Compounds with the Spinel Arrangement. Concluding Remarks on Jahn-Teller Distortions. Stereochemistry of B-subgroup Ions. Introduction Once upon a time it was believed that the structures of most binary solid compounds of the metals, other than alloys and related substances, could be described adequately in terms of an ionic model in which metal ions and anions are treated as charged, more or less compressible, spheres. This model leads to a classical electrostatic theory of bonding and is relatively well understood. Naturally, modern developments in valency theory have tended to emphasize those situations in which a simple ionic theory is inadequate and so have occasionally led to the multiplication of hypotheses to explain observations which could just as well be interpreted in terms of an electrostatic theory. This has tended to obscure the fact that the ionic model still forms a satisfactory background to a great part of the theory of the stereochemistry of metal compounds. In this review we shall emphasize the ionic viewpoint, but not because we are unaware of the importance of covalent bonding. We believe that only after the irrelevance of much that has been presented as qualitative stereochemical evidence for covalency has been appreciated, does the importance of quantitative studies, for example of unpaired electron distributions by paramagnetic resonance experiments, become clear. However, it must be appreciated that factors important in determining the structures of ionic solids also decide the stereochemistry of discrete molecules and complex ions. We shall not, therefore, hesitate to discuss the stereochemistry of isolated molecules or complex ions in solution insofar as they are relevant to the general problem of the stereochemistry of metal ions. Ionic Coordination Polyhedra 76, We first examine the implications of a very much oversimplified ionic model of charged, incompressible, nonpolarizable spheres. This places a lower limit to the ratio of the radius of the central ion to that of the surrounding ions for each type of coordination polyhedron Table I. It follows from 4 that an ideal MX₂ complex should be linear, for this theory. In a similar way the repulsion between X ions in an MX₃ complex is minimized if the molecule is planar and the X ions are at the vertices of an equilateral triangle. There are two other cases in which the preferred configuration depends only on symmetry, namely MX₄ and MX₆ complexes which should adopt regular tetrahedral and regular octahedral structures respectively. These symmetry determined arrangements are illustrated in Fig. Symmetry determined coordination polyhedra: In all other important cases the optimum configuration is not determined by symmetry alone. Although it is possible to arrange eight equivalent X anions at the vertices of a cube about a central M ion as in Fig. Possible arrangements for 8-coordination: The only configurations of MX₅, MX₆? Some of the configurations which lead to relatively favorable electrostatic energies are illustrated in Fig. So far we have made use of condition 4 to determine the ideal configuration for each type of MX_n complex. Now we must determine for each coordination number the values of the ratios of the radii r_M and r_X of the M

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and X ions which are consistent with 2 and 3. Clearly a linear MX₂ arrangement but not a nonlinear one satisfies 3 for any value of the radius ratio. Thus according to 2 a very small A ion cannot form an MX_n complex with n large X ion, but must instead form an MX₂ complex. Proceeding in this way we can derive the ranges of the radius ratio consistent with each type of coordination as shown in Table I. Thus 3 leads us to expect that the octahedron will be preferred to the trigonal pyramid. An exactly similar argument shows that the octahedron will also be preferred to a square prism. Thus, except in isolated molecules or ions, we should not expect to find MX_n arrangements. This discussion shows that the widespread occurrence of regular tetrahedrally and octahedrally coordinated metal ions calls for no special interpretation in terms of bond hybridization, and so forth. It is rather the other four or six coordinated structures which require explanation: The arguments from electrostatic theory apply formally only to substances made up from monoatomic ions. However, there is reason to believe that they are qualitatively correct in a much less restricted context. Thus, although it cannot be proved generally and rigorously, it is very plausible that regular octahedral and tetrahedral coordination are the preferred arrangements for the disposition of most dipolar molecules or polyatomic anions around a central positive ion.

Simple Ionic Compounds The compounds which according to almost any theory should be the most nearly ionic in character are the alkali halides. It is, therefore, particularly interesting to compare their structures with those predicted by our simple theory. In the NaCl structure each cation is surrounded by a regular octahedron of anions and each anion by a regular octahedron of cations, and the structure conforms to our deductions about stable coordination polyhedra. On the other hand, the coordination about 6 is not confined to the alkali halides. A study of a number of other compounds which are generally considered to be among the most completely ionic shows that cubic coordination is not confined to the alkali halides. It occurs, for example, in the fluorite structure in which many ionic MX₂ compounds crystallize. This underlines a point of great importance to the understanding of ionic crystal structures, namely, that the requirement that a structure can be extended indefinitely in space imposes severe restrictions on the types of coordination which are possible. Cubic coordination can be extended indefinitely, but it is not possible to form an extended lattice. Thus a cubic structure is stable for a crystal as a whole, although it does not provide individual ions with their optimum environments; the stability of individual coordination polyhedra is sacrificed in the interest of long-range order. To proceed further with our comparison of theory with experiment we require values of the radii of the ions concerned. The values recommended by Pauling are given in Table II and a discussion of their derivation in the Appendix. A comparison of the limiting radius-ratio values given in Table I with the radius ratios of alkali halides in Table II shows that the observed structures are not always as predicted. According to the simple theory, the rocksalt structure should be stable only within the range 0.414 to 0.732. On the other hand, the CsCl structure is confined under ordinary conditions to CsCl, CsBr, CsI, and the ammonium halides where it is stabilized by a favorable hydrogen bonding arrangement. Since the Madelung constant for this structure is somewhat greater than for the 6-coordinated structure. It is striking that halides and sulfides of metal ions with d⁶ and d¹⁰ shells have a tendency to crystallize in the sphalerite structure; for example, the cuprous halides, AgI, HgS, MnS, CdS, and ZnS. The last three also occur in the wurtzite modification, as do the oxides of Zn and Be. Here again, the simple ionic theory fails to account for the facts for 1 the radius ratios of some of these compounds are compatible with a 6-coordinated structure, and 2 interatomic distances calculated from the usual ionic radii decreased by 5% to 8%. However a satisfactory and detailed interpretation of these structures is still wanting. Tetrahedrally coordinated MX₂ structures; a sphalerite, b wurtzite.

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Chapter 7 : H.J. Emeleus (Author of Advances in Inorganic Chemistry and Radiochemistry, Volume 27)

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