

Models and Paradoxes in Quantum Chemistry*

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Ten different, unexpected features of the classificatory results of quantum mechanics are discussed as well as the question whether the Schrödinger equation only applies to sufficiently small systems which can be indiscernibly reproduced and possess assembly properties.

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Since quantum mechanics is approximately 50 years old [1] it is an appropriate question to ask what it has done for chemistry. The primordial subject was the explanation of line spectra of monatomic entities much in the same way as classical celestial mechanics treated planetary systems in preference to the Pleiades, snow storms and flying birds. Before turning to whatever benefits chemists may have derived it is worthwhile to remember certain features of the understanding of atomic spectra obtained by quantum arguments. Disregarding the recognized correlation effects [2] the electron configurations containing an integral number of electrons in each nl -shell classify correctly the discrete energy levels, and in particular the symmetry types (J and parity; in the case of Russell-Saunders coupling, these levels can be grouped in terms with definite S and L) are predicted correctly. The coherent extension of this classification to molecules and polyatomic ions is the classification of *molecular orbitals* with definite symmetry type in the point-group determined by the equilibrium positions of the nuclei. Whereas molecular spectroscopy [3] previously characterized excited levels belonging to the lowest or higher M.O. configurations as a rather incomplete analogy to atomic line spectra, it has been possible [4, 5] to measure vertical (invariant internuclear distances) and adiabatic ionization energies of penultimate M.O. from photo-electron spectra obtained by bombarding gaseous molecules with 21.2 or 40.8 eV photons or solid samples [6, 7] with 1253.6 eV or 1486.6 eV X-rays originating in a magnesium or aluminium anti-cathode. At the same time as the classification of penultimate M.O. clearly has approached the theory of chemical bonding to the quantum description of atoms as a consequence of the progress of photo-electron spectrometry the last ten years, a multitude of unexpected problems and fascinating surprises has turned up.

Hartmann has at several occasions, and perhaps the most clearly in a *Geburtstagsschrift für Schwarzenbach* [8] pointed out that invented models are able to convey more understanding than attempts to approximate solution of the

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Schrödinger equation of the actual problem of interest. In text-books at lower levels, harmonic oscillators, electrons in boxes and hydrogen atoms are cherished because of the explicit solutions to be compared with Kepler ellipses in classical mechanics, but there is the further advantage of invented models of "Stellschrauben" parametrization. Most of the interesting models actually constitute continuous manifolds of related cases and, on the other side, Nature provides us with regular discrete manifolds such as isoelectronic series. Hartmann [8] also asks the question what one would learn from the total wave-function of a rose. We return below to this profound inquiry of the universality of quantum mechanics. Around the time Niels Bohr held his Nobel lecture in Stockholm, he stated that chemistry now has been reduced to properties of integers. But then he took the course in qualitative inorganic analysis of mixtures able to contain 55 elements (for which the writer later became instructor 1953 at the Chemistry Department A of the Technical University of Denmark) and he is not known to have repeated this platonism, whereas he frequently said that the discovery of quantum phenomena was an irreversible step like the recognition of $\sqrt{2}$ being irrational. However much discoveries in physics have irreversible features (for instance, we are not likely to revert from a heliocentric to a geocentric description of the Solar system) this comparison between natural sciences (always retaining surprising observations for tomorrow like the heavy stone put in the hat Berkeley was asked to give a kick) and mathematics is rather intriguing. In the following are discussed ten different aspects of recent problematic situations in quantum chemistry:

1. *Preponderant Configurations.* It is an empirical fact that the first 20–400 discrete levels of a given atom can be classified in electron configurations independently of the question of how well-defined the configuration is in the actual wave-function. It is worthwhile to emphasize that most excited levels correspond to the replacement of two, one or (in the case of partly filled shells) zero electron of the preponderant configuration of the groundstate. This concept can be generalized to inorganic chemistry, where the oxidation state in most compounds can be defined [9] from the number of electrons in the preponderant configuration of the groundstate. In particular, transition group compounds (and even metallic lanthanides) show a definite number of *d*-like or *f*-like electrons. It is one of the less recognized but lasting results [10] of "ligand field" theory that the absorption spectra clearly indicate this number. The electron transfer spectra [11] correspond to excited levels where the oxidation state of the central atom is decreased (or in the case of certain conjugated ligands increased) by one unit.

2. *Relevant Symmetry.* The best one-electron functions in a polyatomic system are not always the eigen-functions of the Hamiltonian of the full point-group, but the preponderant configuration corresponds to another point-group [10]. This result has been recognized for many years in hydrogen molecules at large internuclear distance, and one should not apply M.O. theory in a naive way to complexes containing two or more partly filled shells. Actually, antiferromagnetic coupling of dimeric chromium (III) [12] and copper (II) [13] complexes can be described by non-diagonal elements of the two-electron operator between diagonal elements corresponding to definite oxidation states of each of the two

central atoms, and in the case of Kamimura-stable half-filled sub-shells [10] the exchange formula of Heisenberg is obtained.

The L.C.A.O. model has been so popular that (with the exception of crystals with plane waves orthogonalized on core orbitals) little attempt has been made to solve the Schrödinger equation directly with an approximate one-electron operator. However, the $X\alpha$ method [14, 15] has recently produced results in excellent agreement with photo-electron spectra though the choice of the radii for the atomic constituents may seem slightly arbitrary. Hartmann [16] counted the angular nodes in the eigen-functions of a central field representing a hexahalide complex. Exactly this class [10, 11] shows red-shifts of electron transfer bands at high pressure and by solution in organic solvents compared with water, corresponding to strong effects of repulsion between the loosest bound orbitals (g -like having four angular nodes) of adjacent ligands. By the same token, the M.O. formed mainly from fluorine $2s$ orbitals are separated several eV in CF_4 and SF_6 according to the number of angular nodes [17]. Nevertheless, the persistence of atomic orbitals in compounds [13, 18] is demonstrated to a remarkable extent by X-ray and photo-electron spectra of inner shells, and the arguments of hybridization theory [19, 20] are counteracted by the clear-cut separation of $2s$ -like M.O. in carbon, nitrogen and oxygen compounds with ionization energies between 22 and 39 eV [4].

3. *Watson Effect.* One of the most important difficulties for performing meaningful calculations on many-electron systems is the evaluation of interelectronic repulsion [2]. The choice of M.O. configurations belonging to irrelevant symmetries produce more correlation effects than needed. In monoatomic isoelectronic series [13, 21] a striking regularity is that the energy differences between the terms belonging to one partly filled shell are decreased relative to Hartree-Fock functions (first studied in a systematic fashion by Watson) by an amount almost independent of the ionic charge. This Watson effect should not be confused with the effect of mixing of $2s^2 2p^q$ and $2p^{q+2}$ increasing as a linear function of the ionic charge and, in a way, expressing beginning irrelevance in the general central field $U(r)$ compared with the more special Coulomb potential. However, the explanation by configuration interaction with invariant principal quantum number might be debated since $2s 2p^q$ show comparable deviations from Hartree-Fock results [9].

4. *Virial Artifacts.* Several authors [22–25] have pointed out that the conventional Slater-Condon-Shortley explanation of term distances as differing interelectronic repulsion, though asymptotically valid for high ionic charges, is incorrect for neutral atoms and moderately positive ions insofar the lowest term has higher interelectronic repulsion overcompensated by increased nuclear attraction because of slightly contracted radial functions (also of electrons not occupying the partly filled shells). A closer analysis shows that this paradox is of the same category as the effect of the virial theorem pointed out by Löwdin [26] that when a crystal such as LiF is formed from gaseous ions, the potential energy decreases *twice* as much as the Madelung energy, but is half compensated by increased kinetic energy. A related problem is whether one can argue legitimately that covalent bonding is equivalent to decreased local contributions to the kinetic energy [10, 27]. Again, a minor adaptation of all the orbitals guarantee

that the virial theorem is valid for the molecule at equilibrium internuclear distances. It is not possible in large molecules and crystals to discuss nuclear attraction and interelectronic repulsion separately. These quantities diverge (as contrasted to a combined Hartree and Madelung potential) and are proportional to the fifth power of the linear dimension of samples having the same shape and chemical composition, whereas the total energy goes as the third power [10].

5. *High Ionization Energies of the Partly Filled Shells.* It is not possible here to discuss the complicated question whether the angular overlap model [10, 28–32] is adequately described by local contributions to the kinetic energy in the anti-bonding *d*- and *f*-like orbitals. However, an important experimental argument from photo-electron spectra is that the partly filled shell has *higher* ionization energy *I* than the loosest bound M.O. delocalized on the ligands in many *d*-group compounds [33], *f*-group salts [6, 34] and higher *I* than the conduction electrons in metallic lanthanides [35, 36]. This is in obvious disagreement with covalent models of the type proposed by Hückel and by Wolfsberg and Helmholz, where the M.O. (L.C.A.O.) are the eigen-vectors of determinants having non-diagonal elements of an effective one-electron operator between definite orbitals of adjacent atoms. Nevertheless, recent calculations on iron (III) [37], nickel (II) [38] and copper (II) [39] containing molecules have confirmed this situation, and actually, two of the five *3d*-like electrons of gaseous iron tris (hexafluoro-acetylacetonate) represent anti-bonding electrons having higher *I* than their bonding counterparts of the same symmetry types. Several chemical arguments [40, 41] can be based on these facts but, obviously, the determination of L.C.A.O. eigen-vectors is not as simple as believed previously in the cases of partly filled shells with so low average radii that the electron affinity is much lower than *I*. It is possible to discuss the phenomenological baricenter polynomial [9] explaining the characteristic features of the five transition groups [42] including the standard oxidation potentials of the aqua ions if the treatment of hydration energy differences [43] is applied. By the same token, the spin-pairing energy considered as definite multiples [13] of the Slater or Racah parameters of interelectronic repulsion describes closely the observed spectroscopic and chemical properties of the transition groups, in particular the lanthanides [44] and the *5f* group [45].

6. *Continua of Penultimate Ionization.* It has frequently been stated by quantum chemists that the *I* values of penultimate M.O. and of inner shells do not have the same physical significance as the lowest *I* constituting the dividing limit between the discrete energy levels and the over-all dense continuum. Though X-ray and photo-electron spectra may be discussed in terms of suddenly changing transition probabilities in the immediate vicinity of singularities in the continuum, the chemical shifts of *I* values for inner shells [6] show clear-cut intra- and inter-atomic relaxation effects [46]. The former case is the difference between the Hartree-Fock eigen-value (assuming “frozen” orbitals according to Koopmans) and *I* of the inner shell of the gaseous atom. Empirically, this difference is close to 0.8 eV times the square-root of *I* in eV. The inter-atomic relaxation effects can be quite substantial (3–10 eV) and their variation from a less to a more polarizable compound contributes strongly to the chemical shift. Ori-

ginally, I was connected with the oxidation state and later with fractional atomic charge much in analogy to the nephelauxetic effect (the decrease of the parameters of interelectronic repulsion in the compound [9] relative to the corresponding gaseous ion) but there is no doubt that strong deviations from the behaviour described by Koopmans occur.

7. *Continua of Translational Energy.* The first factor of the Born-Oppenheimer product of a translational, a rotational, a vibrational and an electronic wave-function (among which monatomic entities lack the second and the third factors) is usually forgotten. Nevertheless, it produces a continuum starting immediately with the groundstate of any system not heavily confined in a small volume, and the absence of perceptible coupling with spectroscopic levels finally is of the same kind as the phenomenological singularities in the continuum. Analogous difficulties occur in the presence of electric potential differences above 25 V. Since all known atoms and molecules have their lowest I below 25 eV, any distance between two such electric potentials remove, technically speaking, the discrete character of any energy level. The same is true in the presence of any radioactive nucleus, again not compatible with strictly stationary states.

8. *Questions of Time-scale.* When selection rules and coincidence of normal frequencies (degenerate for group-theoretical reasons) are observed in vibrational (infra-red and Raman) spectra, they refer to the time-average point-group of the nuclear positions. The instantaneous picture obtained according to the Franck-Condon principle by optical or photo-electron spectra frequently show a much lower symmetry, whereas other experimental techniques such as Mössbauer and nuclear and electron magnetic resonance have intermediate time-scales between the time of passage of one wave-length of exciting radiation and the average content of a unit cell obtained by diffraction of X-rays or neutrons. The comprehensible discrepancies between the structure of Jahn-Teller unstable [e.g. copper (II)] and Gillespie-unstable [e.g. tellurium (IV), xenon (VI) and lead (II)] systems are quite instructive [10]. Another situation which cannot be an eigenfunction of a time-independent Hamiltonian is an optically active enantiomer. Actually, the set of internuclear distances suffices to determine the electronic wave-function without reference to the point-group, but it does not distinguish between enantiomers.

9. *Quantum Mechanics Pretends to be Valid for Other Systems than Electrons.* The short-term specific interest of quantum chemists is concentrated on the $3N$ spatial variables needed for N electrons, and the nuclei are forming a skeleton of points. However, the objects for quantum mechanics form a series of Chinese boxes in the sense that the De Broglie wave-length (h/mv) applies also to systems heavier than electrons. One of the most precise techniques for evaluating the charge distribution of nuclei [47] is the study of energy levels of a negative muon (207 times heavier than electrons) added to an atom, corresponding to the Schrödinger equation for one muon in a non-Coulombic central field without any complications of orthogonality and indiscernibility among the electrons. Since optical gratings do not occur in practice with characteristic distances below 10^{-8} cm, particles heavier than neutrons are not usually diffracted. However, Estermann and Stern [48] demonstrated interference effects by the reflection of beams of He and H₂ on crystalline surfaces of LiF and NaCl. The experiment

with hydrogen molecules is particularly important because m equal to 2 units of atomic weight shows that the diffracted entity is not separated in nuclei. There is not really something completely specific about elementary particles. The invariant mass in chemical analysis and the possibility of recovering the individual atoms from a compound are expressions of decreases in the ninth or tenth decimal place of the rest mass by formation of compounds from atoms, whereas the formation of nuclei from protons and neutrons induces changes in the third decimal. It cannot be said without qualification that an α -particle *consists* of two protons and two neutrons. In one way, it is a higher-level system than a proton, but it is also simpler by being a boson with zero spin.

10. *Assembly Properties and Repeated Small Entities.* Weisskopf [49] pointed out a tendency in Nature to repeat similar systems. However, this tendency becomes less pronounced for large systems; Galaxies are certainly not identical. Also, the genetic code is not sufficient to make animals of the same species similar to more than a given extent anymore than the printer's plate is sufficient to ensure that all stamps of a given emission are strictly identical. The situation is entirely different in quantum mechanics. Disregarding certain details of the Born-Oppenheimer factorization, all helium atoms and other monoatomic systems, and even all small molecules such as methane, titanium tetrachloride and anthracene are *really* identical. They can be reproduced indefinitely, and the solution of the Schrödinger equation has the whole class as reference of comparison. It is true that the regularities of natural sciences consist to a great extent to very few of the 2^n classes formed in bivalent (Aristotelian) logic by n properties [50] having members. However, in classical physics it became a suspicion that two systems cannot be entirely identical (when counting spatial relations to other things, identity is logically excluded) whereas quantum mechanics has brought absolute indiscernibility. It is worthwhile to realize why it is so impossible that electrons carry sub-microscopic identity cards. The concept of number of independent (mutually orthogonal) states has been magnificently confirmed by the energy levels containing $(2J + 1)$ states in spherical symmetry. In the case of one partly filled shell, the number of states is obtained by the rules of indiscernible permutations. One may get an idea of how absolute identity in small systems develop into qualitative similarity in large systems by remarking that the single state of a closed shell is the neutral element (in the sense of "one" in multiplication) of group-theoretical vector coupling. Hence, the Periodic Table and the similarity of properties in isologous series (such as OCN^- , SCN^- , and SeCN^- to be distinguished from isoelectronic species such as NCN^{-2} , N_3^- , CNO^- , and OCO) are based on such neutral elements [10]. The non-diagonal elements of the two-electron operator might readily have produced a perfect confusion, but the unexpected regularities discussed above as points 1, 2, 3, and 6 show that in the domain of discrete energy levels, it is legitimate to count the number of states (which is invariant even for a given symmetry type under second- and higher-order perturbations).

It might be argued that the statistical conclusions of quantum mechanics are physically meaningful for systems which can be repeated in large assemblies. This attitude is more difficult to defend in the case of quantum numbers such as symmetry types. If there were a time when no mendelevium atom existed,

it is an interesting question whether it had the groundstate ${}^2F_{7/2}$. The writer is almost convinced that quantum mechanics cannot be applied to so large molecules (including animals) that they cannot be reproduced in the sense of indiscernibility. This restriction would remove quite a lot of dilemmas. It is known that several of the founders of quantum mechanics such as De Broglie [51, 52], Einstein and Schrödinger with time acquired a rather sceptical attitude toward the Copenhagen interpretation, and for our purpose, it is particularly important to analyze the Gedankenexperiment of Schrödinger [53] where a cat in company with a very weakly radioactive sample and a Geiger counter which would trigger off a hammer breaking an ampulla containing hydrocyanic acid is kept inside a black box for a certain time, corresponding to a known probability of the Geiger counter being activated. Besides the question of the total wave-function consisting of a linear combination of living and dead states until someone opens the box (unless the cat is considered a qualified observer) a much more impressive corollary is that *determinism is gone*. It is not easy to know whether similar spectacular consequences of quantum choices occur in nature, but they certainly occur in the laboratory. It is regrettable that Bohm [54] as a prelude to a defence [55] of the local contribution to the kinetic energy [9, 10] being a “quantum potential” of unspecified origin, where the Brownian motion of a sub-quantum medium changes the causally determined trajectory of the hard core of the double solution [51, 52] in a pretty short time, went as far as to suggest that a radioactive nucleus has an inner dynamics predestining it to explode at a definite future time. Besides the question why the “life insurance statistics” is exactly an exponential decay (the only one compatible with a constant probability of decay per small unit of time) the nuclei show a definite number of states in the sense of partly filled shells discussed above, and this number is 1 in the case of even numbers of protons and neutrons “contained” in the nucleus. It is highly significant that the half-life of a given isotope (or specifically nuclear isomer) does not depend on the mode of production, and that it is immediately established; there are not a few unstable card-house collapsing at once. Using the analogy of the Arrhenius equation, it is astonishing that the activation energy is always the same for a given isotope, and that the “temperature” is everywhere identical.

The psychological motivation behind this discussion may be a distinction developing this century between physicists considering the conservation of energy as more important than causality and those regarding causality as more sacred and a necessary condition for rationality. Thus, the studies of radioactivity by Becquerel and the Curies and the postulate of β particles and neutrinos dividing the energy were used for radical changes of opinion regarding the constitution of matter by people defending invariant energy, whereas Einstein and De Broglie believe in causality. It is true that fluctuations compatible with Heisenberg’s uncertainty principle allow the energy to vary in very short time intervals. However, fluctuations in Dirac fields become slightly intellectually uneconomical when each new elementary particle needs its own Dirac field. Arguments can be given that the Universe is a vacuo fluctuation [56] with zero total energy and at the same time a closed “black hole”. Quite independently of possible difficulties for detailed energy conservation, the fate of Schrödinger’s cat is, in the writer’s

opinion, an even more serious stroke against universal determinism than Michelson's experiment was against absolute space. Obviously, one can *always* invent alternative models and contraptions. The analogy to the Lorentz-Fitzgerald contraction (as one-valued function of the absolute velocity) is here the suggestion of Everett and Wheeler [57] that each quantum choice splits the Universe in mutually distinct worlds without possibilities of communications. This rather desperate attempt seems uneconomical, because the quantum choice is not of the simple kind as the political party of the president of U.S.A., but infinite and diffuse in all directions. Thus, a spoonful of a thorium compound is a continuous, violent source of Everett-Wheeler splitting. Each second, $2 \cdot 10^{-18}$ among the thorium nuclei emit an α particle. But this is not their only choice; the α particle can be emitted in *any* (how many different?) direction. The writer is far more sympathetic to the discussion by Ballentine [58] and to the suggestion that quantum mechanics is only applicable to reproducible small systems with assembly properties. If this suggestion is not adopted, what to do with Schrödinger's cat?

The ancient Romans would have asked whether quantum mechanics *works*, and the Greeks whether it is *valid*. It is perhaps characteristic that most present-day criticism is directed toward the continuum (including the De Broglie waves of translational motion with positive energy) and the time-dependent processes (such as racemization of enantiomers and radioactivity) whereas the discrete levels (however discrete they are under close scrutiny) are well understood, but in part due to apparent accidents of successful description based on symmetry arguments, both in spherical and in lower point-groups. When one wants a formula for the world, one may either (like Newton, Maxwell and Einstein) hope for a few pregnant equations, where the material parameters by insertion *in principle* would predict any event, including the outcome of a football match tomorrow; or one may modestly ask for equations compatible with any world. Whereas the principle of identity ($A = A$) is a quite good candidate for the second use, it does not seem plausible that the Schrödinger equation has the former qualification. Already the transfinite cardinality of the number of continuum states is a problem. In practice, the classification of energy levels of transition group compounds and of photo-electron spectra has followed the same development as atomic spectra did [59] and chemical kinetics and minor differences in free energy seem beyond reach. Seen from the point of view of the chemist [60] quantum mechanics has mainly provided models and allowed extrapolated correlations in classes of compounds.

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