

# How Electronic Energy Levels Get Modified in Potentially Bound Forms (Atoms, Molecules, Crystals)

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**Abstract**—Electron in free state is not at all similar to that in bound system. A bound system is nothing but a system with modified potentials. The nature of potential effects on the solution of Schrodinger's equation. Hence the distribution of energy levels is modified along with. A free electron movement gives rise to continuous spectra but this is not the case always. In case of atoms the nature of barrier potential changes as one increases number of electrons from one (Hydrogen atom), two (Helium atom), three, four (Alkali atoms) and so on. In case of molecules the overlap of atomic orbitals and their symmetry have substantial effects on the energy spectra. Lastly, whenever one deals with lattice in case of crystalline structure the periodicity as well as defects in some of the concerns influence the energy spectra. Whenever the average potential due to core ions and neighbouring electrons are replaced by the periodic potentials which are quite obvious in case of crystalline structure, an array of allowed and forbidden bands of energy are availed. The allowed bands consist of series of sharp energy levels the numbers and spacing of which again are functions of nature of the potential energy barrier.

**Keywords**—quasi-free electron, binding energy, energy levels, energy bands, energy spectra)

## I. INTRODUCTION

Free electron motion is an abstract idealization in the real field. In the classical domain one speaks of a continuum. There is no story of discretization. The dispersion relation is continuous in form. Even if in case of quantal treatment where there is no potential barrier one finds Schrodinger's equation resembling with simple harmonic type. So, the solution is obvious to be of sinusoidal form or linear combination of such forms. The value of wave-vector varies continuously from 0

to  $\infty$ . Thus, the dispersion relation takes the form-  $E = \frac{\hbar^2 k^2}{2m_e}$  - Plank's constant,  $k$ -Wave-vector,  $m_e$  - Mass of electron]. Whenever a potential well or potential barrier is envisaged which has finite length. So, boundness of potential causes discretization of energy. The aforementioned equation

turns into  $E = \frac{n^2 \pi^2 \hbar^2}{2m_e L^2}$  on application of the proper boundary conditions [ $n=1,2,3,\dots,\infty$  (Principal quantum

number,  $m_e$  - mass of electron,  $L$ - length of well/barrier]. So, energy is an explicit function of  $n$  and  $L$ . the functional dependence shows that although having occurrence of discrete character the bound state- levels do not maintain uniform distance from each other. As the quantum number  $n$  increases, the energy levels get more and more separated. In case of hydrogen and Alkali atoms the same energy equation

acquires the form  $E = \frac{1}{n^2}$  and  $E = \frac{1}{(n-\mu)^2}$

respectively, where  $\mu$  is quantum defect [1]. In these

bound states the energy levels get closer with increase of principal quantum number resulting in a nearly continuum at higher frequency. This is quite obviously obtained through "Bohr's Correspondence Principal" [2] which is eminently known as a merger between classical and quantal theory.

Again, if one looks at simple harmonic potential i.e.  $V = \frac{1}{2} k x^2$  the energy levels are found to be equidistant. In case of vibronic diatomic molecule along its axis the same spectra is observed but in real life some sorts of anharmonicity happens to exist which refrains the energy levels from being equidistant. The levels get closer and closer as vibrational quantum number increases. The emission and absorption lines merge to a continuum finally. It is conclusive that whenever the lines are equidistant the bonding remains stronger than when anharmonicity appears to make the levels closer. Now, let's turn at rotational molecules. For low to moderate energies, the energy levels are determined by mere kinetic energy which falls in the category of rigid rotor [3] but above a certain energy this model fails. Then the centrifugal distortion potential enters along with the mere kinetic energy. This results in shift of the lines toward lower frequency with increase in rotational quantum number in power of three [4]. So, the emission and absorption lines are also no more equidistant. They come closer with increase of rotational number. i.e. the bond weakens and the levels are less stable for molecules to sustain.

The solid-state structure is most complicated. It is classified into two sections- one is amorphous and the other is crystalline. We will confine ourselves to crystal which consists of three salient properties running holding their hands together. They are- lattice, basis and periodicity [5]. Basis compromises with atoms, molecules, ions. Different types of potentials, usually combination of short range and long-range parts [6] actually remains effective among the bases pairwise. The lattice structure happens to be in equilibrium state for some distance among the atoms pairwise. The distribution of electron clouds obviously has an effect over formation of sustainable bonds along with ion cores. The lattice constants and parameters determine the dimension of potential well and barrier as mentioned earlier. Those are generally of atomic dimensions. The difference lies in the nature of potential. For constancy of barrier potential, the simple harmonic solution is obtained in case of  $E > V$  [ $E$ - Total energy of electron,  $V$ - Height of Barrier]. The barrier potential is most likely to be of periodic nature for the lattice property. The difference here occurs in the nature of wavefunction and energy-wave-vector relationship. The dispersion relation is not only discontinuous but also complicated. This property is quite pronounced at the edge of the material. It may be understood that as a fact that

sharing of outer shell electrons among the ion cores splits the original energy level into several levels which gives rise to band structure. More detailing gives that a band structure rather than distinct sharp edges relying on the potential as evident from "Bloch's theorem" and "Kronig Penny model" [7]. In case of periodic potential several bands of energies are found along with band gap. So, all the domain are not accessible for the electrons. The orders of magnitude of band gap are decider of whether a material will be metal or insulator or semiconductor.

## II. THEORY

This section will be divided into three separate subsections. The first one will be dedicated to atoms, second one will be dedicated to molecules and the third one will be dedicated to lattice.

### A. Atomic Binding

First of all the atomic spectra of Hydrogen atom is considered. Out of Bohr's theory of quantization of angular momentum one finds radius of Bohr's orbit to be  $r_n = n^2 a_n$  [n-Principal quantum no.,  $a_n$ - Bohr's radius]. Putting it into energy equation one obtains energy of electron in the n-th orbit to be  $E_n = -\frac{13.6}{n^2}$  electron-volt. According to this semiclassical theory all the energy levels with different azimuthal numbers (l) have same energy. This concept had existed until the appearance of "Vector Atomic Model" [8]. In the year of 1925 G.E. Uhlenbeck and S.A. Goudsmit introduced the quantal concept spin. The circular motion of the electron causes orbital angular momentum. This angular momentum interacts with spin angular momentum which gives rise to L-S. coupling. This very perturbation removes l-degeneracy. The modified energy acquires the form-

$$\Delta E = [j(j+1) - l(l+1) - s(s+1)] \quad (1)$$

l- angular momentum quantum number

s- spin angular quantum number

j= l+s- total angular quantum number

s=1/2 holds for electron. That's why there is a doubling of each of the spectral lines which is said to be fine structure.

The next simplest structure is di-electronic Helium atom. It is tackled with three different ways.

The first one is independent electron model. Here, The total Hamiltonian discards the static correlation [9] term pertaining to the interaction between the electrons. It follows the total energy is the sum of two semiclassical terms as in Hydrogen atom which reads as-

$$E_n = -\frac{Z^2}{2} \left( \frac{1}{n_1^2} + \frac{1}{n_2^2} \right) \quad (2)$$

Z - Atomic number

$n_i (i=1,2,\dots)$  - Principal quantum numbers

The ground state energy is found to be -108.8 electron-volt. It is very much less relative to the experimentally found value (-79 electron-volt).

The second one is to include the correlation term  $\frac{1}{r_{12}}$  in the Hamiltonian [  $\vec{r}_{12} = |\vec{r}_1 - \vec{r}_2|$  ].  $r_1$  and  $r_2$  are the positions of the electrons relative to the nucleus. While put in the energy shift formulation of the 1st-order perturbation one gets the energy value to be  $\frac{5}{8}Z$ . The calculational procedure follows as [10]. The result is improved, still there is excess in value as compared to the experiment.

Third approach considers of screening effect. It is thought that the charge of the nucleus is screened to each due to occurrence of the other electron in the Helium atom. The nuclear charge  $Z$  is replaced with  $Z_{eff} = Z - S$  where,  $S$  is the screening parameter. The first order calculation gives the energy shift to be-

$$\Delta E = -2 Z_{eff} (Z - Z_{eff}) \quad (3)$$

Combining with above two formalisms one obtains the total energy to be-

$$E = -Z_{eff}^2 + \frac{5}{8}Z - 2 Z_{eff} (Z - Z_{eff}) \quad (4)$$

The application of Ritz variational principle [11] the value of  $Z_{eff}$  is found to be  $Z - \frac{5}{16}$ . So, the value of screening parameter  $S$  will be  $\frac{5}{16}$ . The corrected value of energy of ground state Helium has been found to be  $\approx -74.8$  electron-volt which is much more conforming with the experimentally obtained value -79 electron-volt.

As electron is a fermion, it has to obey Pauli's exclusion principle. Electron is a spin- $\frac{1}{2}$  particle. It is identified by two different co-ordinates; one is position and the other is spin. In case of Helium atom two electrons do exist. They may align parallelly to produce total spin  $S=1$  or antiparallelly to produce total spin  $S=0$ . The former is called Ortho-Helium and the latter is called Para-Helium. In case of Ortho-Helium, less amount of repulsion occurs. So, the energy of this state is lower relative to Para-Helium. The energy levels in Para and Ortho states are not similar. In Ortho state the energy levels start from 2s rather than 1s. The inter-state distances are different for the two sets.

Now, it's time to enter more into many electron atoms. Here also the same two factors i.e. inter-electronic correlation and screening effect are challenges. Due to occurrence of the screening parameter the simple Coulombic potential is modified as-

$$V(r_i) = - \quad (5)$$

Where,  $Z$ -Nuclear Charge,  $r_i$ -Distance of the  $i$ -th electron from the nucleus,  $S(r_i)$ - Screening experienced by the  $i$ -th electron of the atom. So, the screening parameter is no more a constant as Helium. It is function of distance from the nucleus. This non-Coulombic but symmetric potential removes l-degeneracy. If one discards the correlation term

along with a contribution from the non-Coulombic symmetric term (residual Coulomb Hamiltonian) the

$$\sum_{i=1}^N \frac{Z - S(r_i)}{r_i}$$

Schrodinger's equation is satisfied. In the eigenvalue equation the energy is function of both  $n$  and  $l$ . In actual sense, correlation term is an electrostatic term. Consideration of relativistic correction [12] introduces a term-

$$\sum_i \frac{1}{2m_e^2 c^2} \frac{1}{r_i} \frac{dV(r_i)}{dr_i} (\vec{L} \cdot \vec{S}) \quad (6)$$

This is called LS-coupling term which is of paramount importance in case of many-electron target. In case of lighter atoms the electrostatic repulsion is not too strong to break L-S coupling. But, in case of heavier atoms the pairwise electrostatic repulsion is strong enough to break L-S coupling. So, jj-coupling is important here. It can be also inferred that in case of lighter atom the L-S term acts as perturbation whereas in case of heavier atoms the residual Coulomb Hamiltonian acts as perturbation (fine-structure consideration) [13].

The discussion remains incomplete without inclusion of Alkali spectra. These metals are highly metallic in character. Elementary knowledge says that outside the complete core, here lies only one unpaired electron. So, the core contributes as optically inactive  $^1S_0$  structure and the unpaired electron is mostly responsible for the corresponding spectral structure. The core electrons here do have a screening effect for which the outermost electron cannot see the entire nuclear charge. Again this is responsible for removal of l-degeneracy which was absent for Hydrogen atom. This very comparison is given due to the fact that both the Hydrogen atom and Alkali atoms have one active electron. The energy already has been given previously where quantum defect is an implication of screening.

### B. Molecular Binding

The discussion will be limited to simple molecules only. What we understand as molecule is an amalgamation of at least two nuclei and distribution of electron cloud over them. The type of bonding and overlap act as decider of energy in this regard. The simplest molecular structure is  $H_2^+$ . It is a homonuclear molecular ion. Bohr-Oppenheimer Approximation [14] assumes the motion of electron is much faster than the motion of nucleus. Hence, the nucleus may be assumed to be at rest. This approximation is basic in all sorts of molecular energy spectra. A simple harmonic motion model is supposed to be in between two heavy nuclei and a molecule is in stable condition due to stable equilibrium there in. A small displacement from the equilibrium  $r_0$  position causes a restoring force  $F = -K(r - r_0)$ . Out of the Heisenberg's uncertainty principle  $\Delta p \Delta x \approx \hbar$ , from the dimension of molecule one may have an idea of energy of electron inside. Doing a bit of simple algebra electronic energy is found to be  $10^3$  times greater than that of vibrational energy and  $10^6$  times greater than that of

rotational energy [15]. In case of homonuclear molecule the external energy cannot change the rotational state of molecule due to not having permanent dipole moment. The

external energy also cannot trigger the vibrational energy also due to same reason. It is easier to disturb electrons in the molecule. The change of distribution is associated to change of dipole moment and so one can have rotational and vibrational energy band structure along with.

Another two very important factors are bonding and antibonding. Molecular orbital is just a wavefunction to find electron in a molecule i.e. concept of molecularized electron [16] comes into play. This is a probabilistic interpretation. So, the individual electronic entity is lost. In case of bonding orbital the electron clouds of two protons of a di-atomic molecule are accumulated between them, so, the overlap is pronounced. On the other hand, in case of antibonding orbital there occurs a depletion of electrons between the protons resulting in reduction of overlapping. So, the bonding orbital is of larger strength of bond and stabler than that of antibonding [17,18]. Another important parameter is the bond order [18] which determines the strength of bond.

The simplest representation of molecular orbital is Linear Combination of Atomic Orbitals [19,20] which is given as-

$$\begin{aligned} \varphi &= \dots\dots\dots C_1\psi_1 \pm C_2\psi_2 \pm \dots\dots\dots \pm C_N\psi_N \\ &= \sum_{i=1}^N C_i\psi_i \end{aligned} \quad (7)$$

Where, the (+) and (-) signs imply bonding and antibonding respectively. Here, the  $\psi_i$ 's represent component atomic orbitals with similar symmetry.

Another important and the most worth mentioning theory is Hartree-Fock Self Consistent Field (HF-SCF) theory. This theory is indispensable to understand many body physics. In case of molecule one has five type of energy terms- electronic kinetic energy, ionic kinetic energy, inter-ionic interaction, ion-electron interaction and inter-electronic correlation. As per Born-Oppenheimer Approximation one neglects kinetic energy of ions. The electrostatic interaction among bare ions (pairwise) are kept apart from calculation. The prime aim is to solve again the electronic Schrodinger's equation. The electronic correlation term acts as perturbation. The introduction of the Fock function is for the sake of minimization of the total energy [21]. Nevertheless, having the electronic interaction term the solution of the equation is approximated to be expressed as product of single electronic wave functions. But in case of electron (fermion) there is an association of spin co-ordinates along with spatial co-ordinates. The product form-

$$(8) \quad \varphi_i = \prod_i \psi_i(r_i)$$

is called Hartree's product. The one overlooks the General Hartree-Fock or Z-averaged Perturbation Theory [22] the wavefunction  $\psi_i(r_i)$  is expressed as product of spatial and spin co-ordinates [21]. But, there is a problem of antisymmetrization which is resolved by Slater's determinant. The occurrence of Permutation operator

appears as solution. The sequential as well as recurring combination of sum (subtraction) of products help minimize the inconsistency. Actually, H-F solution endorses to make possible the complex asymmetric part in form of eigenvalue equation with the alliance of Roothan equation. The overlap integral is annulled by the transformation of basis to orthogonal one. The description is given in [21]. The Hartree-Fock Roothan equation reads as [19].-

$$\sum_s C_{si} \hat{F} \chi_s = \varepsilon_i \sum_s C_{si} \chi_s \quad (9)$$

where,  $\hat{F}$  is Hartree-Fock operator such that-

$$\hat{F} = \hat{H}^{core}(1) + \sum_{j=1}^{n/2} [2\hat{J}_j(1) - \hat{K}_j(1)] \quad (10)$$

The first term denotes unperturbed part, J denotes Coulomb integral and K denotes exchange integral.  $\varepsilon_i$  is binding energy of electron obtained by diagonalizing the Slater's determinant.

### C. Crystalline Binding

In case of free/quasi-free electrons [23] there remains continuous states of accessibility. But, the scenario changes in case of complex type potentials. For the sake of simplicity we will concentrate on periodic type potentials, consisting of an array of potential wells and barriers.

In the forthcoming discussion we will concentrate on Bloch Theorem and Kronig-Penney model. These two theorems are intertwined with each others and the common ground lies in the periodicity of potential in the solid-state crystalline structure. Bloch's theorem expresses the solution of Schrodinger's equation to be expressed as-

$$\psi(r) = e^{\pm ikr} \mu_k(r) \quad 11.(A)$$

subject to  $\mu_k(\vec{r} + \vec{a}) = \mu_k(\vec{r}) \quad 11.(B)$

The Schrodinger's equation is a second order differential equation. It comprises of two independent solutions- f(r) and g(r). From the periodicity of potential there will be also two more solutions - f(r+a) and g(r+a) but from the rule of mathematics they must depend on f(r) and g(r). what

happens as a consequence is that the ratio of  $\psi(r+a)$  and  $\psi(r)$  is a constant say ( $\delta$ ). Here, the summary will only be retained without mentioning the entire calculation [24,25]. The invariance of the Wronskian [26] allows the parameter

$\delta$  to be expressed in form of a simple quadratic equation.

Application of Sridharacharya's rule, one finds two types of determinants; one producing real root and the other producing imaginary root. Both are correct mathematically. But, the real root obstacles physical acceptance because of the pre-assumed finite nature of physically acceptable wavefunction in quantum mechanics. This does disallow the energy values to be accessible everywhere as was in classical mechanics and the quantal picture so far. There are forbidden regions also. Nowfrom, two modifications of paradigm with take place- firstly, the existence of allowed energy bands will be separated by forbidden bands and the frequency will no more remain proportional with wave number [27].

Obviously the energy of electron is greater than the uniform potential  $V_0$ . Two schrodinger's equations are found. Imposing Bloch theorem with periodicity (a+b) and the conditions for continuity of logarithmic differential gives four linear equations with four unknown parameters. For the non-zero solution to exist the determinants whose elements are the coefficients of four unknowns to be zero, which gives [28]-

$$P \frac{\sin \alpha a}{\alpha a} + \cos \alpha a = \cos ka \quad 12.(A)$$

where,

$$P = \frac{mV_0 b a}{\hbar^2} \quad 12.(B)$$

Now, array of consecutive wells and barriers will be taken in account. Here the periodicity will be (a+b) [a for well and b for barrier]. Actually well stands for distance between two mobile ions and the barriers occur at the position of ions.



( $m$  is electron's mass and  $\hbar$  is reduced Plank's constant,  $k$  is the associated wave vector,  $\alpha$  is supposed to be measure of electronic energy).  $P$  is a measure of how strongly the electron is bound to the respective potential well. The right hand part of the equation 12.(A) is a cosine function. This confines the left hand portion between -1 and +1 which is again a restriction or constraint whatever one may say on the energy i.e. the E-k curve should not be continuous. An alternation of allowed and forbidden energy bands are formed. In case of infinitely strong bond ( $P \rightarrow \infty$ ) the energy bands are squeezed into lines separated by gaps i.e. line spectrum is formed. On the other hand in case of infinitely weak bond ( $P \rightarrow 0$ ) energy band is restored. In the intermediate zone one finds band width to be decreasing with strengthening of the bond.

It is already discussed that the E-K diagram is discontinuous in nature. So, Brillouin zones [29] are formed. Let's concentrate on the first Brillouin zone [30]. Combining simple electrodynamic equation with the quantum with simple quantal formalism a wonderful expression of mass is obtained which is reduced mass. This reduced mass is actually the indicator of how much bound is the electron in the crystalline structure. It is denoted as-

$$= \frac{\hbar^2}{d^2 E / dk^2} \quad (13)$$

 $m^*$ 

The binding energy of the electron in the lattice is best understood by the factor-

$$f_k = \frac{m}{m^*} \quad 14.(A)$$

$$= \quad 14.(B)$$

$$\frac{m}{\hbar^2} \frac{d^2 E}{dk^2}$$

This denotes that upto the point of inflection of E-K curve i.e. upto highest level one happens to obtain free electron. These free electrons are the only elements which take essential as well as sufficient part in electrical conduction process. So, it is quite evident that the extent to which the allowed energy bands in a solid remains filled up is the determinant of how good or poor conductor the material is [31,32]. In case of filled or nearly filled allowed energy

bands one obtains less effectively free electrons, hence the material is insulating in nature. Whereas, the materials in which energy bands are almost empty the number of effectively free electrons are much probable resulting it to be metallic in character. In the intermediate range one obtains semiconductor. In the standard notation the above definitions are defined as absolute zero temperature.

### III. CONCLUSION

In the entire article two types of character of electron; bound and unbound are discussed. In atomic and molecular level the binding energy opens the door for research toward ionization, capture, excitation [33-38]. In the solid state the different acoustic, electrical, thermal and optical properties may be studied. Immense scopes of research are there involving various type of potentials in this regard.

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