Single-Molecule Quantum-Effect Electronic Devices

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ABSTRACT

This paper studies single-molecule electronic devices in which quantum *state* transitions should be utilized to achieve processing tasks. Some biomolecules, molecules and supramolecules exhibit practical luminescence, photoelectric effects, photon absorption, resonance and other quantum effects. We study device physics using processable *observables*. The evolutions of *observables* must be controlled, and, the *microscopic* device *controllability* is studied.

Keywords: electronics, molecule, quantum mechanics

1. INTRODUCTION

Single-molecule *microscopic* devices and molecular processing *fabrics* are envisioned departing from conventional microelectronics [1]. These molecular inroads and solutions may emerge as progress will be achieved on many fronts. Despite debatable prospects on the engineering solutions and technology readiness, the major reasons to conduct research in single-molecule devices and *microscopic* processing are to:

- 1. Contribute to fundamental knowledge;
- 2. Enable transformative transitions towards innovative inroads of science, engineering and technology ;
- 3. Advance prospective technologies and solutions;
- Potentially enable understanding of phenomena, mechanisms and principles of information and data processing in living organisms;
- 5. Advance device physics of *microscopic* devices;
- Comprehend mechanisms and principles of sensing, processing and memory by biomolecules and biomolecular assemblies;
- 7. Devise and examine practical enabling paradigms of sensing and processing by *engineered* devices and *fabrics*.

Enormous advantages are foreseen because the aforementioned advances and premises promise to ensure a leap in sensing as well as enable data processing preeminence, computing superiority and memory supremacy.

2. MACROSCOPIC AND MICROSCOPIC DEVICES

The majority of microelectronic devices are *macroscopic* from device physics, phenomena utilized and analysis prospects [1]. In these microelectronic devices:

- Quantum phenomena are not utilized;
- Quantum effects and mechanics are not usually applied to examine the baseline characteristics and capabilities;
- Quantum effects significantly degrade the overall capabilities and functionality.

Microscopic systems are examined by applying quantum mechanics. The device, module and system testing, characterization and evaluation are imperative tasks. The Heisenberg uncertainty principle provides the fundamental limits on the measurements implying constraints on the testability, characterization, evaluation, etc. The position-momentum and energy-time limits on the measurements are given by using the standard deviations on the *observables* as

 $\sigma_x \sigma_p \geq \frac{1}{2}\hbar$ and $\sigma_E \sigma_E \geq \frac{1}{2}\hbar$.

and $\sigma_E \sigma_E^{-1/2} \hbar$. (1) The implications and consequences of the Heisenberg uncertainty principle are:

- Depending on devices and device physics, quantummechanical sensing, memory and processing processes, mechanism and phenomena exhibited by *natural* and *engineered microscopic* systems may or may not be directly and explicitly ascertained, characterized, demonstrated and verified;
- Some theoretical premises, postulates and hypothesis can be indecisive, unverifiable or impractical.

Despite the limits and complexity emphasized, practical and functional *microscopic* devices and systems can be designed, tested and verified. For example, lasers, resonanttunneling transistors and other devices, which utilize quantum effects, were characterized, used and implemented.

3. QUANTUM-EFFECT MOLECULAR DEVICES

Various cyclic molecules, organic and inorganic compounds have being synthesized, and, their chemical and physical properties are reported [2]. Supramolecular assemblies and complexness can be synthesized using supramolecular chemistry procedures. These assemblies are synthesized from molecular subunits and components. The exhibited phenomena, characteristics and spatial threedimensional assembly topology are predefined by the electronic coupling between atoms and molecules, bonds, molecular orbitals, weak forces (electrostatic, intermolecular, hydrogen bonding, hydrophobic, pi-pi interaction, van der Waals and other), strong forces (covalent bonding between atoms which share electrons), etc. There are reversible noncovalent interactions between molecules in supramolecular assemblies and complexes. Using molecular building blocks and structural units, supramolecular assemblies and complexes are synthesized usin macrocycles, self-assembly, molecular recognition, complexation, template-directed motifs, etc. The *engineered microscopic* devices, which exhibit the specified quantum phenomena, can be synthesized using cyclic molecules, compounds, cages and other structures with carbon interconnecting frameworks and scaffolds [2, 3].

The π electrons move between multielectron atoms over which the π bonds resonate. These *delocalized* π electrons in a molecule are not attributed to a specific atom or covalent bond. *Delocalized* electrons are contained within an orbital which extends over several adjacent atoms. *Delocalized* electrons in conjugated systems of double bonds, aromatic and other systems are of our particular interest.

In the aromatic ring of benzene, there are six *delocalized* π electrons. There are overlaps of the orbitals in the *p*-*p* π system in benzene and in the *p*-*d* π system in the phosphhazene ring. The motion and propagation of *delocalized* electrons in molecules may be controlled. Figures 1 report single-molecule devices with overlapping atomic orbitals. These devices may have the *input*, *control* and *output ports*. The orbitals of the terminal atoms are hybridized to the adjacent atoms (not shown) by electron-pair covalent bonds. E.g., molecules are covalently bonded *directly* or using linkers and spacers to form molecular *fabrics*.



Figure 1. Multi-terminal cyclic molecule

The motion and *states* of electrons can be controlled by using external excitations. The device physics of singlemolecule *microscopic* devices is based on quantum interactions, perturbations, controlled electron transport, charge variations, field changes and other controlled quantummechanical or quantum-phenomena-induced evolutions. The applied excitation

$\boldsymbol{\mathcal{E}}(t,\mathbf{r}) = [V_{\varepsilon}(t,\mathbf{r}), \mathbf{E}_{\varepsilon}(t,\mathbf{r}), \mathbf{B}_{\varepsilon}(t,\mathbf{r})]$

changes the time- and spatial-varying potential function $\Pi(t,\mathbf{r})$, volume charge density $\rho(t,\mathbf{r})$, electric field intensity $\mathbf{E}(t,\mathbf{r})$, vector magnetic potential $\mathbf{A}(t,\mathbf{r})$ as well as quantummechanical quantities (absorption, emission, probabilities, rates, reflection, transmission and other). Time- and spatiallyvarying $\mathbf{g}(t,\mathbf{r})$ could be potential $V_{\varepsilon}(t,\mathbf{r})$ and electromagnetic fields $\mathbf{E}_{\varepsilon}(t,\mathbf{r})$ and $\mathbf{B}_{\varepsilon}(t,\mathbf{r})$.

Molecular devices may operate in the controlled electronexchangeable environment. The controlled super-fast potentialand field-assisted propagation of electrons, charge variations, field evolutions and quantum effects can be achieved. The *state* transitions, system evolutions, interactions and other phenomena quantitatively define the device physics, quantummechanical statistics, etc. The device controllability should be achieved by varying $\boldsymbol{\mathcal{E}}(t,\mathbf{r})$. Metastable evolutions, quantum interactions, photon emission and absorption as well as other transductions were observed and implemented.

4. EVOLUTION OF *MICROSCOPIC* SYSTEMS: Formulation and Solutions of Quantum-Mechanical Problems

Quantum mechanics is applied to describe and examine the evolutions and transitions of *microscopic* particles (electrons) and systems. Analysis of electron propagation and quantum *state* transitions ultimately results in the analyses of device functionality, controllability and capabilities.

To examine *microscopic* systems, consider the evolution of electrons. The unperturbed time-dependent Schrödinger equation is

$$-\frac{\hbar^2}{2m}\nabla^2\Psi(t,\mathbf{r}) + \Pi(t,\mathbf{r})\Psi(t,\mathbf{r}) = i\hbar\frac{\partial\Psi(t,\mathbf{r})}{\partial t},$$
(2)

where $\Psi(t,\mathbf{r})$ is the wave function; $\Pi(t,\mathbf{r})$ is the potential.

In general, the time-dependent and time-independent Schrödinger equations are

$$H\Psi(\mathbf{r},t) = i\hbar \frac{\partial \Psi(\mathbf{r},t)}{\partial t}, \ \Psi(\mathbf{r},t) = \psi(\mathbf{r})\varphi(t), \text{ and } H\psi = E\psi, \ (3)$$

where *H* is the *total* Hamiltonian operator, $H=H_0+H_I+H_F+H_E$; H_0 is the unperturbed Hamiltonian; H_I , H_P and H_E are the interaction, perturbation (disturbances, interferences and other) and excitation (control) Hamiltonian terms.

The wave function Ψ is given as

$$\Psi(\mathbf{r},t) = \sum_{n=1}^{\infty} c_n \Psi_n(\mathbf{r},t) = \psi(\mathbf{r})\varphi(t) = \sum_{n=1}^{\infty} c_n \psi_n(\mathbf{r}) e^{-i\frac{E_n}{\hbar}t} = \sum_{n=1}^{\infty} c_n \psi_n(\mathbf{r}) e^{-i\omega_n t},$$
(4)

where $|c_n|^2$ is the probability that a *microscopic* system is in a state with E_n , $\sum_{n=1}^{\infty} |c_n|^2 = 1$.

Example 4.1.

In a magnetic field, the interaction of a magnetic moment μ with a magnetic field **B** changes the energy by $-\mu$ ·**B**. The external electromagnetic field affects the Hamiltonian. For a particle in a uniform magnetic field **B**, one has

$$H = \frac{1}{2m}\mathbf{p}^2 + \Pi(\mathbf{r}) - \frac{q}{2\mu c}\mathbf{B}\cdot\mathbf{L} + \frac{q^2}{8\mu c^2} \left[B^2r^2 - (\mathbf{B}\cdot\mathbf{r})^2\right],$$

where μ is the angular momentum; L is the orbital angular momentum.

Using the Hamiltonian operator H, one explicitly derives the evolution of the expectation value of an operator B as

$$\frac{d}{dt}\langle B\rangle = \left\langle \frac{\partial B}{\partial t} \right\rangle + \frac{1}{i\hbar} \langle [B,H] \rangle, \ [B,H] = BH - HB, \tag{5}$$

If B has no explicit time dependence, then, the change of the expectation value is

$$\frac{d}{dt}\langle B\rangle = \frac{1}{i\hbar}\langle [B,H]\rangle.$$
(6)

The operator B corresponds to a physical observable b as

$$\langle b \rangle = \int \Psi^*(t, \mathbf{r}) B \Psi(t, \mathbf{r}) dV$$
. (7)

For the wave function, which may characterizes the physical state for the system, one obtains

$$\Psi(t + \Delta t, \mathbf{r}) = \Psi(t, \mathbf{r}) + \frac{i\hbar\Delta t}{2m} \frac{\partial^2 \Psi(t, \mathbf{r})}{\partial \mathbf{r}^2}.$$
(8)

The wave function Ψ is not a physically measurable or observable quantity.

Example 4.2.

Consider a *microscopic* particle which moves in an external time-dependent electromagnetic field. The vector magnetic and scalar electric potentials are $\mathbf{A}(t,\mathbf{r})$ and $V(t,\mathbf{r})$.

The time rate of change of the expectation value of **r** is $d = \frac{1}{r} \left(\frac{1}{r} \right)^{1/r}$

$$\frac{d}{dt} \langle \mathbf{r} \rangle = \frac{1}{i\hbar} \langle [\mathbf{r}, H] \rangle = \frac{1}{m} \langle \mathbf{p} - \frac{q}{c} \mathbf{A} \rangle$$

From $\mathbf{v} = \frac{1}{m} \left(\mathbf{p} - \frac{q}{c} \mathbf{A} \right)$, one yields the Hamiltonian
 $H(t, \mathbf{r}, \mathbf{p}) = \frac{1}{2m} \left(\mathbf{p} - \frac{q}{c} \mathbf{A} \right)^2 + qV$.

Hence

 $\frac{d}{dt} \langle \mathbf{v} \rangle = \left\langle \frac{\partial \mathbf{v}}{\partial t} \right\rangle + \frac{1}{i\hbar} \langle [\mathbf{v}, H] \rangle = -\frac{q}{mc} \left\langle \frac{\partial \mathbf{A}}{\partial t} \right\rangle + \frac{1}{i\hbar} \langle [\mathbf{v}, \frac{1}{2}m\mathbf{v} \cdot \mathbf{v}] \rangle + \frac{1}{i\hbar} \langle [\mathbf{v}, qV] \rangle$ From $\frac{d}{dt} \langle \mathbf{v} \rangle = -\frac{q}{mc} \left\langle \frac{\partial \mathbf{A}}{\partial t} \right\rangle + \frac{q}{2mc} \langle \mathbf{v} \times \mathbf{B} - \mathbf{B} \times \mathbf{v} \rangle - \frac{q}{m} \langle \nabla V \rangle$ and $\frac{1}{2} \partial \mathbf{A}$

and $\mathbf{E} = -\nabla V - \frac{1}{c} \frac{\partial \mathbf{A}}{\partial t}$,

the time rate change of the expectation value for the velocity operator is

$$\frac{d}{dt}\langle \mathbf{v}\rangle = \frac{q}{2mc}\langle \mathbf{v} \times \mathbf{B} - \mathbf{B} \times \mathbf{v}\rangle + \frac{q}{m}\langle \mathbf{E}\rangle$$

This equation describes the evolution of $\langle \mathbf{v} \rangle$.

The charged carriers can be controlled by changing the electromagnetic field. The time-dependent Schrödinger equations for two distinct states with

$$H_{1} = \frac{1}{2m} \left(\mathbf{p} - \frac{q}{c} \mathbf{A}_{1} \right)^{2} + \Pi_{1} + qV_{1}$$

and $H_{2} = \frac{1}{2m} \left(\mathbf{p} - \frac{q}{c} \mathbf{A}_{2} \right)^{2} + \Pi_{2} + qV_{2}$
are $-\frac{\hbar^{2}}{2m} \left(\nabla - i\frac{q}{\hbar c} \mathbf{A}_{1} \right)^{2} \Psi_{1}(t, \mathbf{r}) + \left(\Pi_{1} + qV_{1} \right) \Psi_{1}(t, \mathbf{r}) = i\hbar \frac{\partial \Psi_{1}(t, \mathbf{r})}{\partial t},$
 $-\frac{\hbar^{2}}{2m} \left(\nabla - i\frac{q}{\hbar c} \mathbf{A}_{2} \right)^{2} \Psi_{2}(t, \mathbf{r}) + \left(\Pi_{2} + qV_{2} \right) \Psi_{2}(t, \mathbf{r}) = i\hbar \frac{\partial \Psi_{2}(t, \mathbf{r})}{\partial t}.$
Here, $\mathbf{A}_{2} = \mathbf{A}_{1} + \nabla f(t, \mathbf{r})$ and $V_{2} = V_{1} - \frac{1}{c} \frac{\partial f(t, \mathbf{r})}{\partial t},$

where $f(t,\mathbf{r})$ is the continuous differentiable Lipshitz function. One derives

$$\Psi_2(t,\mathbf{r}) = e^{i\frac{q}{\hbar c}f(t,\mathbf{r})}\Psi_1(t,\mathbf{r})$$

which yields the transition operator $U(t, \mathbf{r}) = e^{i\frac{q}{\hbar c}f(t, \mathbf{r})}$.

We study the evolution of *observables* in *microscopic* systems. There is a need to examine the motion of *delocalized* electrons, probabilities, transition rate, etc. For example, the *delocalized* electrons may propagate within the bonding molecular orbitals, each occupied by two electrons of opposite spin leading to a covalent bond. Using $\Psi(t,\mathbf{r})$, one obtains the probability of finding the *delocalized* electrons at the specified spatial infinitesimal volume Δv as

$$P = \int \Psi^*(t, \mathbf{r}) \Psi(t, \mathbf{r}) dv.$$
(9)

(10)

This probability yields estimates for the *delocalized* electron velocity, *transit* time, transmission, etc. The probability current density $J(t,\mathbf{r})$ is frequently applied. The probability density and probability current density are

 $P(t,\mathbf{r}) = \Psi^{*}(t,\mathbf{r}) \Psi(t,\mathbf{r})$

and
$$\mathbf{J}(t,\mathbf{r}) = \frac{i\hbar}{2m} [\Psi(t,\mathbf{r})\nabla\Psi^*(t,\mathbf{r}) - \Psi^*(t,\mathbf{r})\nabla\Psi(t,\mathbf{r})].$$
 (11)

Example 4.3.

The expectation value of x is

$$\langle x \rangle = \int_{-\infty}^{\infty} \Psi^*(t, x) x \Psi(t, x) dx$$

The evolution of $d\langle x \rangle/dt$ is govern by

$$\frac{d}{dt}\langle x\rangle = \frac{i\hbar}{2m}\int_{-\infty}^{\infty} \left(\frac{\partial\Psi^*}{\partial x}\Psi - \Psi^*\frac{\partial\Psi}{\partial x}\right)dx = -\frac{i\hbar}{m}\int_{-\infty}^{\infty}\Psi^*\frac{\partial\Psi}{\partial x}dx.$$

The probability to find a *microscopic* particle between a and b at time t is found as

$$\int^{b} \Psi^{*}(t,x) \Psi(t,x) dx$$

One obtains the evolution of the probability of finding a particle in $a \le x \le b$ as

$$\frac{dP_{ab}}{dt} = J(t,a) - J(t,b), J(t,x) = \frac{i\hbar}{2m} \left[\Psi(t,x) \frac{\partial \Psi^*(t,x)}{\partial x} - \Psi^*(t,x) \frac{\partial \Psi(t,x)}{\partial x} \right]$$

Example 4.5.

Consider a *microscopic* particle if its one-dimensional motion is confined in [0 L], and, $\Pi(x) = \begin{cases} 0 & \text{for } 0 < x < L \\ \infty & \text{for } x \le 0 \text{ and } x \ge L \end{cases}$.

By solving the Schrödinger equation

$$-\frac{\hbar^2}{2m}\frac{\partial^2\psi(x)}{\partial x^2} + \Pi(x)\psi(x) = E(x)\psi(x),$$

one obtains

$$E_n = \frac{\hbar^2 \pi^2}{2mL^2} n^2,$$

$$\psi_n(x) = \sqrt{\frac{2}{L}} \sin\left(\frac{n\pi}{L}x\right), \ 0 \le x \le L, \ n = 1, 2, 3, \dots$$

The probability of finding a particle within $x \in [0 \ l]$ is

$$P = \int_{0}^{l} \psi^{*}(x)\psi(x)dx = \frac{2}{L}\int_{0}^{l} \sin^{2}\left(\frac{n\pi}{L}x\right)dx$$

= $\frac{1}{L}\left[x - \frac{L}{2n\pi}\sin\left(\frac{2n\pi}{L}x\right)\right]_{0}^{l} = \frac{1}{L}\left[l - \frac{L}{2n\pi}\sin\left(\frac{2n\pi}{L}l\right)\right].$

5. MULTIELECTRON ATOMS AND MOLECULES

A neutral atom with atomic number Z contains a nucleus with electric charge eZ and Z electrons. The Hamiltonian for a single nonrelativisitic electron in the *central* Coulomb potential

$$\Pi(r) = -\frac{Ze^2}{4\pi\varepsilon_0 r}$$
 is
$$H = -\frac{\hbar^2}{2m}\nabla^2 - \frac{Ze^2}{4\pi\varepsilon_0 r}$$

For multielectron atom, the unperturbed Hamiltonian operator

$$H_{0} = \sum_{j=1}^{Z} \left(-\frac{\hbar^{2}}{2m} \nabla_{j}^{2} - \frac{Ze^{2}}{4\pi\varepsilon_{0}r_{j}} \right) + \frac{1}{2} \frac{e^{2}}{4\pi\varepsilon_{0}} \sum_{j\neq k} \sum_{j=1}^{Z} \frac{1}{\left|\mathbf{r}_{j} - \mathbf{r}_{k}\right|}$$
(12)

implies the need to solve the following partial differential equation in 3Z dimensions

 $H_0 \psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_Z) = E \psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_Z).$ (13)

It is impossible to analytically solve the Schrödinger equation (13) for even simple multielectron atoms due to the electron-electron and electron-nucleus interactions. The perturbations and excitations, which must be considered, further significantly complicate the problem. Electromagnetic fields (excitations, perturbations, disturbances, interferences and other) affect a *total H*. One yields the resulting equation

 $H\psi(\mathbf{r}_1,\mathbf{r}_2,...,\mathbf{r}_Z) = E\psi(\mathbf{r}_1,\mathbf{r}_2,...,\mathbf{r}_Z), H = H_0 + H_I + H_P + H_E,$ (14)

Example 5.1.

The electric field intensity \mathbf{E} , which affects H, depends on the potential V. For the static case

 $\mathbf{E} = -\nabla V.$

In linear isotropic media

 $\nabla \cdot \varepsilon \mathbf{E} = \rho$,

where the volume charge density is $\rho = dq/dv$.

In general one uses $\varepsilon(\mathbf{r})$, while, in the homogeneous media ε is constant. The Poisson equation for the electric potential $\nabla^2 V = -\rho/\varepsilon$

is solved by using the boundary conditions.

In the spherical coordinate system one finds

$$\nabla^2 V(r) = \frac{1}{r^2} \frac{d}{dr} \left(r^2 \frac{dV(r)}{dr} \right) = -\frac{\rho}{\varepsilon},$$

and $\mathbf{E} = -\nabla V$.

We consider the *microscopic* system under the excitation (control) $\boldsymbol{\mathcal{E}}(t,\mathbf{r}) = [V_{\varepsilon}(t,\mathbf{r}), \mathbf{E}_{\varepsilon}(t,\mathbf{r}), \mathbf{B}_{\varepsilon}(t,\mathbf{r})]$ and perturbations. For time-varying electromagnetic fields, the *nonhomogeneous wave equations* for scalar and vector potentials are

$$\mathbf{E} = -\nabla V - \frac{\partial \mathbf{A}}{\partial t},$$

$$\nabla^{2} V - \mu \varepsilon \frac{\partial^{2} V}{\partial t^{2}} = -\frac{\rho}{\varepsilon},$$

$$\nabla^{2} \mathbf{A} - \mu \varepsilon \frac{\partial^{2} \mathbf{A}}{\partial t^{2}} = -\mu \mathbf{J}.$$
(15)

Equations (15) are reduced to

$$\mathbf{E} = -\nabla V,$$

$$\nabla^2 V = -\rho/\varepsilon,$$

$$\nabla^2 \mathbf{A} = -\mu \mathbf{j}$$
(16)

only for a static case which is not sufficient or adequate for many problems.

Hence, for *microscopic* systems one needs to solve the Schrödinger (14) and *nonhomogeneous wave* (15) equations in order to obtain time-varying transductions, *state* transitions, evolution of *observables*, etc. The aforementioned problem is analytically and numerically unsolvable even for atoms with a few electrons. We apply the Hartree-Fock paradigm examining the *delocalized* electron using the effective $Z_n(r)$ which is evaluated at the average value of r for the shell. The atomic wave function is represented as a product of single-electron orthonormalized wave functions

$$\psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_Z) = \psi_1(\mathbf{r}_1) \psi_2(\mathbf{r}_2) \dots \psi_Z(\mathbf{r}_Z).$$
(17)
Each-electron wave function satisfies the equation
$$H_i \psi_1(\mathbf{r}_i) = E_i \psi_1(\mathbf{r}_i), H = H_{i0} + H_{i/2} + H_{i/2} + H_{i/2}.$$
(18)

 $H_j \psi_j(\mathbf{r}_j) = E_j \psi_j(\mathbf{r}_j), H_j = H_{j0} + H_{j1} + H_{iP} + H_{jE}$, (18) yielding Z simultaneous integro-differential equations (18) for Z wave functions $\psi_j(\mathbf{r}_j)$. Equations (16) and (18) are solved simultaneously. The aforementioned iterative self-consistent scheme allows one to solve a quantum-mechanical problem for *delocalized*, optically- and field-active electrons which define or affect the device functionality, performance and capabilities.

6. CONCLUSIONS

Applying theoretical foundations of quantum mechanics, we examined quantum phenomena which may be utilized to ensure sensing and processing by single-molecule devices. The controlled evolutions of *microscopic* particles in *microscopic* devices were studied to research the overall functionality, processability, controllability and other features. A highfidelity modeling with consecutive heterogeneous simulations and data-intensive analysis were performed.

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