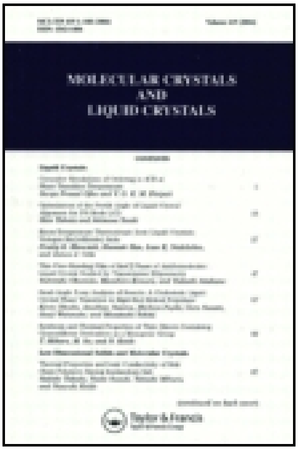

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Quantum Mechanical Processors Based on Organized Molecular Assemblies

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A practical model of a quantum logic gate is proposed, which consists of two photo-induced, charge-separating molecular systems. Basic concept of quantum computing and recent studies in ultrafast photophysical processes in highly organized molecular assemblies are reviewed.

Keywords; quantum computing, quantum interference, organized molecular system, energy transfer, electron transfer,

INTRODUCTION

In this decade, quantum computing is a research subject of advanced studies in general physics. Theoretical models have been proposed for quantum computation based on quantum mechanics[1-5]. A major problem is how the elementary quantum logic gate can be realized. A basic requirement for quantum processors is that quantum information must be processed under a condition that the coherence is maintained during processes and the quantum interference is controlled between neighboring quantum subsystems. Recent advance in photophysical studies revealed that photophysical processes like electron transfer and excitation transfer can occur as coherent process in highly organized molecular systems. In the present paper, we propose a model for a quantum logic gate by means of organized molecular systems.

BASIC CONCEPT OF QUANTUM COMPUTING

Let us consider a register consisting of molecular systems, a, b, c, ···, each of which has an information unit *bit* represented by the two states 0 and 1 as in the classical Boolean states. Practically, the two states can be represented in principle by the state of a two-state particle, for examples, an excited state and a ground state, a spin 1/2 state and a spin -1/2 state, or a charge-separated state and a neutral state. Therefore an n-bit number is then represented by a set of n-molecular systems involved in this register. A schematic model of a register and data processing is shown in Fig. 1. In quantum processors, the state of a register is described by a wavefunction represented by a state vector. Quantum data processing consists of applying a sequence of unitary transformations to the state vector, and the resultant quantum state can be read out by measuring the register state[1].

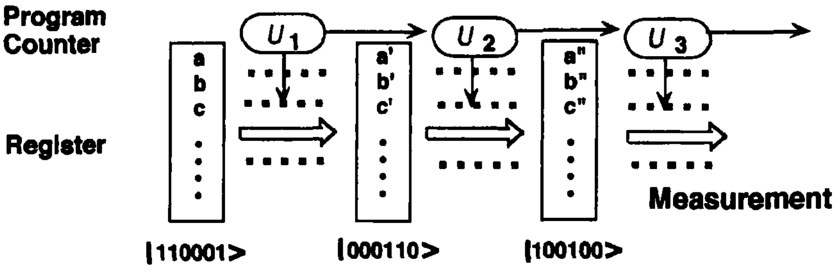

FIGURE 1 Basic concept of quantum mechanical processor.

Fundamental properties of the quantum data processing include striking features summarized as follows; (1) *Superposition*: A single bit can exist not only in the Boolean states but also in an arbitrary complex linear combination of orthogonal states | 0 > and | 1 >.

$$|a\rangle = \alpha\,|0\rangle + \beta\,|1\rangle, \quad \text{where} \quad |\alpha|^2+|\beta|^2=1 \qquad (1)$$

Schumacher[3] named such a quantum bit as "*qubit*". (2) *Interference*:

Quantum computation is characterized by parallel computation paths in individual qubits. If a particular pair of qubits has an interaction with each other, the time-evolving quantum states can interfere constructively or destructively depending on their phase factors. (3) *Entanglement*: A pair of qubits a and b can exist in an entangled state expressed as, for example, $\Psi_{a,b} = (1/\sqrt{2})\,(|0,1\rangle \pm |1,0\rangle)$. This means that the qubit by itself has no definite state, and the qubit is in both two states $| 0,1 >$ and $| 1,0 >$ simultaneously so long as that the state is not measured. More generally, a string of n-qubits can exist in any state described by a linear combination of 2^n basis functions in a 2^n -dimensional Hilbert space,

$$\Psi = c_1|1000\cdots\rangle + c_2|0100\cdots\rangle + \cdots + c_n|1111\cdots\rangle = \sum_{x=000\cdots0}^{111\cdots1} c_x|x\rangle \qquad (2)$$

where c_x's are complex numbers and the index x ranges over all 2^n classical values of an n-bit string.

Some quantum logic operations are simply extensions of classical Boolean operations to superpositions of the input states. In transformation on a single qubit, the following 2 × 2 matrix yields superposition of $| 0 > \sim(1, 0)^T$ and $| 1 > \sim(0, 1)^T$.

$$\sigma_{SQN} = \frac{1}{\sqrt{2}}\begin{pmatrix} 1 & 1 \\ -1 & 1 \end{pmatrix} \qquad (3)$$

$$\sigma_{SQN}|0\rangle = (1/\sqrt{2})(|0\rangle - |1\rangle) \quad \text{and} \quad \sigma_{SQN}|1\rangle = (1/\sqrt{2})(|0\rangle + |1\rangle) \qquad (4)$$

This operation is called "square root of NOT", in the sense that subsequent operation yields NOT operation which flips the Boolean state of a single bit.

$$\sigma_N = {\sigma_{SQN}}^2 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \sigma_N|0\rangle = |1\rangle \quad \text{and} \quad \sigma_N|1\rangle = |0\rangle \qquad (5)$$

All these features are intrinsically nonclassical, because they transform Boolean states into superpositioned states.

Next let us consider transformation in a two-qubit system which is necessary for implementation of an elementary quantum logic gate. In the two-qubit system consisting of a control qubit a and a target qubit b, an arbitrary quantum state $| a, b >$ can be represented by linear combination of four orthonormal basis functions; $| 0, 0 > \sim (1, 0, 0, 0)^T$, $| 0, 1 > \sim (0, 1, 0, 0)^T$, $| 1, 0 > \sim (0, 0, 1, 0)^T$ and $| 1, 1 > \sim (0, 0, 0, 1)^T$. It is known that an elementary and universal quantum logic gate is given by a two-qubits quantum controlled-NOT gate [6]. In this gate, the target qubit undergoes transformation that depends on the quantum state of the control qubit. This operation can be represented by the following unitary matrix,

$$U_{XOR}^{(\tau)} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & e^{i(\pi/4)}\cos(\pi\tau/2) & e^{-i(\pi/4)}\sin(\pi\tau/2) \\ 0 & 0 & e^{-i(\pi/4)}\sin(\pi\tau/2) & e^{i(\pi/4)}\cos(\pi\tau/2) \end{pmatrix} \tag{6}$$

where τ is a parameter related to some rotation of the phase angle involved in the coherent reaction. Here we take a special case $\tau = 1$ for convenience of discussion and obtain much simpler matrix.

$$U_{XOR}^{(1)} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \end{pmatrix} \tag{7}$$

If the initial state is in any basis state, the result yields an exclusive OR (XOR) logic of the input state.

$$U_{XOR}^{(1)}|0,0\rangle = |0,0\rangle,\quad U_{XOR}^{(1)}|0,1\rangle = |0,1\rangle,\quad U_{XOR}^{(1)}|1,0\rangle = |1,1\rangle,\ \text{and}$$
$$U_{XOR}^{(1)}|1,1\rangle = |1,0\rangle \tag{8}$$

where the state of target bit b flips if the control bit $a = 1$, otherwise b is left unchanged. In every case, the control bit a remains unchanged.

We should note here that the quantum processing is a coherent process, and that the quantum mechanical phase must be conserved during the processes. Photophysical reactions in suitably arranged molecular systems can occur as a coherent process in a time range of subpicosecond, and it allows us to make the quantum controlled-NOT gate by means of organized molecular systems.

QUANTUM MECHANICAL ASPECT IN PHOTOPHYSICAL PROCESSES IN ORGANIZED MOLECULAR SYSTEMS

Photochemical processes such as electron transfer and excitation transfer are expected to occur very fast in organized molecular systems in which reactant molecules are arranged with close proximity and optimal orientation. Molecules in such systems can be coupled to an adjacent molecule with relatively strong intermolecular interaction, and therefore they may undergo ultrafast photochemical reaction. If the reaction rate exceeds the rate of vibrational relaxation leading to dephasing in the quantum state, a reaction occurs as a coherent process. If the reaction system consists of several molecules which are arranged linearly along the reaction pathway, a sequential reaction can occur through non-equilibrium excited states. We can see several examples such quantum coherent processes of sequential excitation transfer and electron transfer in natural photosynthetic reaction systems and in artificial organized molecular systems.

Sequential Transport of Electron and Exciton in Photosynthetic Reaction Center

Natural photosynthetic reaction centers and light-harvesting antenna have highly organized molecular arrangements[7,8] as shown in Fig. 2. In the antenna protein of a photosynthetic purple bacterium, 9~32 BChl *a* molecules form two types of rings, horizontally oriented ring (B800) and vertically oriented rings (B850 and B875). The excitation energy captured first by B800 is transferred to B850. Probably the

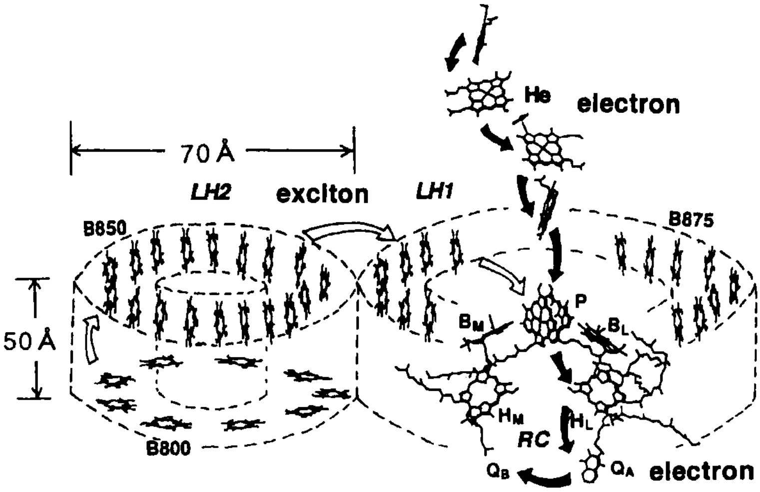

FIGURE 2 Molecular arrangement of photosynthetic reaction center.

excitation energy goes around the ring as a quantum wave of exciton, and at particular time the exciton moves to the adjacent ring B875 of LH1 protein. The exciton is reached finally to the special pair of BChl *a* (P), then an electron starts to move along the molecular channel $P \rightarrow B_L \rightarrow H_L \rightarrow Q_A \rightarrow Q_B$. The reactions are characterized by ultrafast reaction and high quantum efficiency of almost unity, and these features must be a consequence of relatively strong intermolecular interaction and the sequentialness and cooperativeness among sequentially arranged molecules.

Recent studies have revealed a explicit quantum nature in these electron transfer and exciton transfer reactions[9-11]. The fluorescence decay of the LH1 porphyrin ring involves a dumping oscillation with a time period of about 100~200 fs. The time courses of fluorescence and transient absorption of the special pair of bacteriochlorophyll *a* exhibit also similar dumping oscillation. Such oscillatory behaviors indicate that the primary steps of the exciton transfer and charge separation occur in coherent processes, and that the reaction phase maintains during the reactions.

Coherent Photophysical Processes in Artificial Molecular Systems

In specially designed molecular system for electron transfer, a quantum beat appears in the fluorescence decay of electron donor[12]. Fig. 3 shows the molecular structure and the experimental result. An electron donor (Zn-porphyrin) and an acceptor (Fe-porphyrin) are linked to anthracene with 4.9-Å distance. The electron transfer kinetics was probed with fluorescence decay measurement by a fluorescence up-conversion method. The electron transfer occurs very rapidly (τ_F = 231 fs) in benzene solution. On the other hand, the reaction in PMMA polymer film occurs with τ_F = 1.3 ps showing a dumping oscillation with a time period of 450 fs. In polymer matrix, the molecular environment is rather sparse so that the energy dissipation is slow and the decoherence time is much longer than in solution.

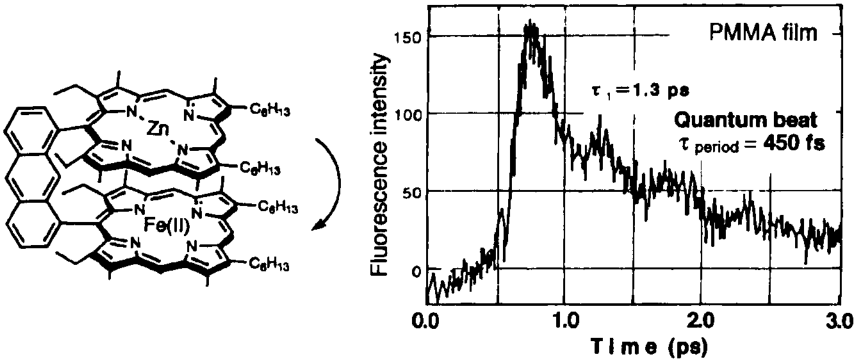

FIGURE 3 Molecular structure and fluorescence decay curve of zinc-porphyrin (donor) in metal porphyrin hetero dimer.

The coherent excitation transfer was suggested in sequentially stacking molecular architecture of Langmuir-Blodgett multilayers, as shown in Fig. 4[13]. The interlayer energy transfer among different types of cyanine dyes involves fast (~10 ps) and slow (0.1~1 ns) transfer processes. In the *slow* transfer process, the fluorescence polarization undergoes randomization, and then the interlayer transfer

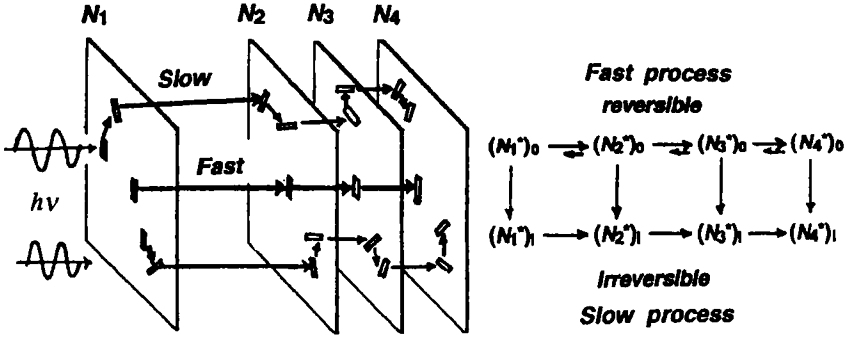

FIGURE 4 Sequential excitation transfer in Langmuir-Blogdett films. The transfer paths consists of fast and slow processes.

occurs in an equilibrium state of dyes in each layer. The kinetic behavior can be interpreted in terms of conventional Förster's very-weak interaction mechanism[14]. In the *fast* transfer process, the fluorescence polarization decays within ~10 ps which corresponds to the fluorescence rise time. Note that the intermolecular distance is 25 Å much shorter than the critical transfer distance of 50 ~ 70 Å. It follows that a much stronger interaction, Förster's *medium* interaction mechanism[14], is involved with an the interaction energy $|u|$, $10^2 < |u| < 10^3$ cm^{-1}. As is discussed in the next section, this mechanism is characterized by the reversible energy transfer associated with quantum recurrence (cyclic period of ~100 fs) between highly vibrational levels of S_1 in donor and acceptor. It is referred to as *non-equilibrium* excitation energy transfer[13].

Quantum Coherency and Recurrence

Quantum beat which appears on the fluorescence or transient absorption time profiles is a phenomenon associated with the quantum recurrence between the initial and final states of a photoreacting molecular system. Originally the recurrence in the excitation transfer was predicted in the Förster's mechanism[14]. Let us consider a

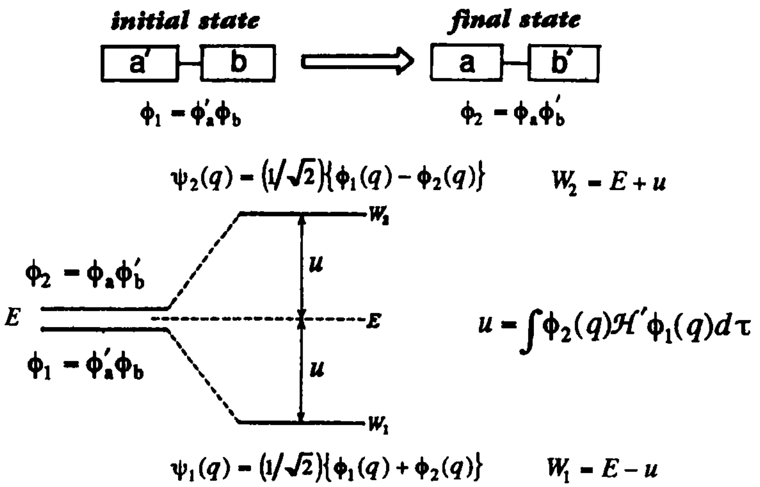

FIGURE 5 Molecular interaction in excitation energy transfer.

molecular system consisting of two molecules, donor and acceptor molecules in the excitation transfer. In zero order approximation, a singly excited state of the system is described as $\phi_1 = \phi_a'\phi_b$ and $\phi_2 = \phi_a\phi_b'$ for the initial and final states, respectively, where ϕ_1 and ϕ_2 are the eigenfunctions of H_0, and ϕ' is for an excited state of free molecule *a* or *b*. The initial and final states interact each other through a perturbation Hamiltonian H' and splitted by the interaction energy *u*, as is shown in Fig. 5. The interaction energy can be approximated as a Coulombic dipole-dipole resonance integral. Then we have a new basis functions, $\psi_{1,2} = (1/\sqrt{2})\{\phi_1(q) \pm \phi_2(q)\}$.

A time-evolution of the total wavefunction can be obtained as a general solution of the time-dependent Schrödinger equation.

$$\Psi(q,t) = (1/\sqrt{2})\,\psi_1(q)\mathrm{e}^{-iW_1t/\hbar} + (1/\sqrt{2})\,\psi_2(q)\mathrm{e}^{-iW_2t/\hbar} \qquad (9)$$

Hence we are concerned with a probability density of the quantum state, we take a square of Eq.(9).

$$|\Psi(q,t)|^2 = (1/2)\,\psi_1^2(q) + (1/2)\,\psi_2^2(q) + \psi_1(q)\psi_2(q)\cos[(W_1 - W_2)t/\hbar]$$

$$= \cos^2(ut/\hbar)\ \phi_1^2(q) + \sin^2(ut/\hbar)\ \phi_2^2(q) \tag{10}$$

This means that the quantum state undergoes cyclic evolution between the initial photoexcited state and the excitation transferred state with a time period of $h/2u$. In case of a molecular system in which the interaction energy is ~100 cm^{-1}, the time period of the recurrence is estimated to be 0.1 ~ 1 ps.

Necessary condition for appearance of the quantum beat is that there exits no interaction with the environment surrounding the molecular system during the reaction. The experimental results shown above indicate that the photophysical process in suitably arranged molecular systems occurs under such condition and the quantum mechanical phase is conserved in subpico-second time range. This is a consequence of relatively strong interaction between the reactant molecules leading to very fast photo-reaction which is comparable or faster than the energy dissipation to the environment.

IMPLEMENTATION OF QUANTUM LOGIC GATE

The above mentioned situation in organized molecular systems satisfies basic requirement for implementation of a quantum logic gates.

An elementary quantum logic gate that we propose here is achieved by using a molecular system consisting of two charge-separating groups attached closely to a molecular base, as shown in Fig. 6. The two groups are denoted as *control qubit* (or 1st dot) and *target qubit* (2nd dot). Each dot can undergo a photo-induced electron transfer in an anti-parallel direction, and it may induce relatively large electric dipole due to the new charge distribution. When the 1st dot is photoexcited, resultant large electric field must affect the energy level of neighboring 2nd dot. Similarly, when the 2nd dot is photoexcited, the resultant electric field may shift the energy level of the 1st dot. In the energy level diagram and correlation diagram of this *quantum-confined Stark effect*, the electronic states are

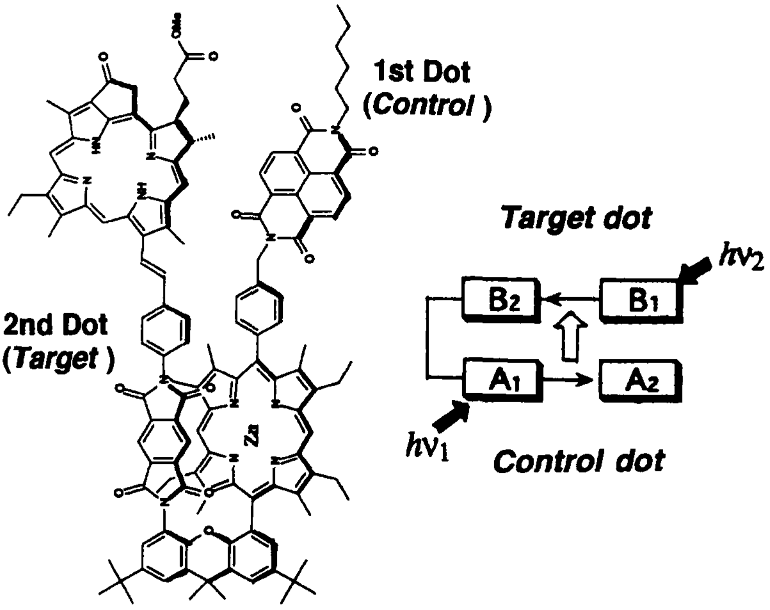

FIGURE 6 A model of quantum controlled NOT gate.

denoted as $|0>$ and $|1>$ respectively for the ground and excited states. If we are concerned with the resonant optical transition of the 2nd dot, the optical absorption corresponding to the transition $|1> |0> \rightarrow |0> |1>$ is allowed only if the quantum states of the two dots are $|1>$ and $|0>$. Namely the 2nd target dot undergoes the XOR logic operation from the 1st controlled dot.

Practically, the quantum *controlled-NOT* operation can be performed in the following procedure (Fig. 6). When A_1 is photoexcited at $h\nu_1$, an electron transfer takes place to A_2, and in other words, an initial neutral state $\psi_{a1} = \phi_{a1}\phi_{a2}$ is changed into an final ion-pair state $\psi_{a2} = \phi_{a1}{}^{+}\phi_{a2}{}^{-}$. The total wavefunction of system A, $\psi_a = c_{a1}\psi_{a1} + c_{a2}\psi_{a2}$, is time-evolved with changing coefficients c_{a1} and c_{a2}. When the second laser pulse of $h\nu_2$ is irradiated and B_1 is photoexcited, the electron transfer in system B (target dot) is started under the intermolecular interaction with the quantum state of system A (controlled dot). By changing the time delay between the two laser pulses which corresponds to the phase of the quantum recurrence involved in these coherent reactions, one can observe a controlled NOT operation. If another logic gate is integrated at the neighboring site, we do not need to measure the target dot in this stage, and the quantum state of this target bit can operate the next gate in a quantum dynamics fashion. In this way, we are now in the first step of realizing the quantum computer.

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