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Smallest artificial molecular neural-net for collective and emergent information processing

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While exploring the random diffusion of 2 bit molecular switches (we define as molecular neuron) on an atomic flat Au (111) substrate, we have found that at least four molecules are required to construct a functional neural net. Surface electron density wave enables communication of one to many molecules at a time—a prerequisite for the parallel processing. Here we have shown that in a neural net of several molecules, some of them could dynamically store information as memory and consistently replicate the fundamental relationship that is found only in a collective and emergent computing system like our brain. © *2009 American Institute of Physics*. [doi:10.1063/1.3227887]

The ever-growing complexity of information processing requires a unique paradigm shift from the existing electronics and computation, where the solution is predefined.[1] Following strict instructions, a computer sends current through its well-defined circuits and solves a problem. However, randomly connected systems might generate an emergent logic in a dynamic network as explicitly observed in our central nervous system, cellular regulatory machinery, ecosystems, etc.[2] Intelligent and massively parallel computations would be possible if such unique processors are built artificially. Using quantum dots or molecules,[3] several processors have been designed to realize unconventional computing.[4] The critical condition for such hardware is that all components should communicate with each other and influence their decision making, which is not possible without a wireless connection. Surface electron density wave has been proposed to serve the role of the wire for communication.[5] However, no such practical devices have been developed to this date, though literatures are rich in conceptual realization of the neural circuit.[6] Here, the fundamentals of a wireless dynamic molecular neural network (MNN) have been experimentally realized using molecular switches.

Using the MNN, two major problems are addressed. The statistical methods can be used for solving dynamic problems in an infinite size network. However, they are not applicable to the realistic central nervous system like networks where constituent subnetworks regulate the operation using periodic and chaotic dynamics. Recently, Zhigulin[7] has proposed a parametric relation to explain this dynamics. The MNN built here follows this relation; thus one fundamental aspect of brain function is replicated. Following White *et al.*,[8] we have also experimentally shown[9] how a network's storage capacity for temporal memory varies with the system size. Therefore, we have mapped processing capability of MNN as number of participants (molecules) increase in an assembly of diffusive molecules.

A submonolayer of 2,3,5,6-tetramethyl-1,4-benzoquinone [Duroquinone or DRQ, Fig. 1(a)] molecule is grown on a Au(111) substrate. DRQ reversibly switches among its four conformers (denoted as 0, 1, 2, and 3) during RAM and ROM operations,[9] and exchange states when they come in contact so DRQ is considered as a molecular neuron. DRQs movement is attributed to the rotation of alkyl groups by $\pm 60°$ and they do not assemble into an integrated architecture if scanned by scanning tunneling microscope (STM) at less than 0.6 V (V_t) tip bias.[10,11] V_t is always kept at less than 0.6 V.

Input signals are given vertically by using STM tip. To read the state, typical conductance of the conformer is matched with its reported value.[9] The STM image of the conformer computed using Green's formalism is found consistent with the experimental one [Fig. 1(c)].[12] Thus, we have detected ~100 consecutive events; however, within ~15 scans (~10 min) the rules of logic-state change become evident. Information processing is defined as change in the

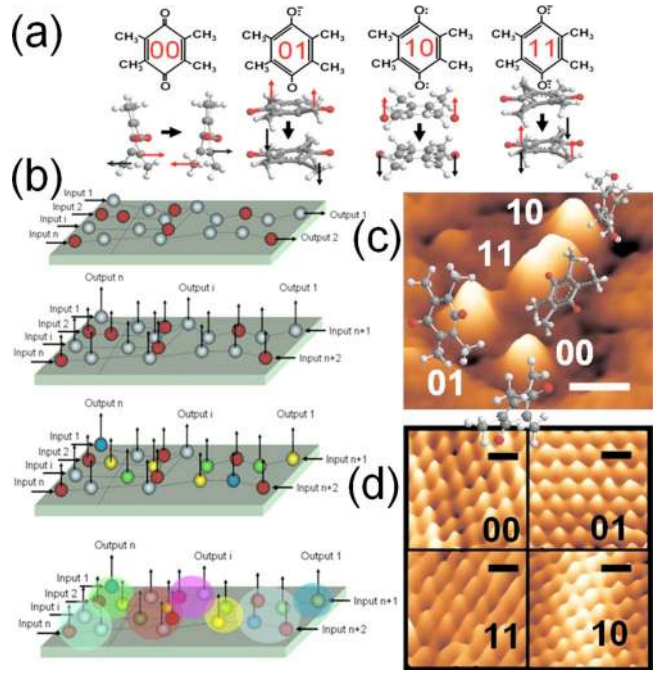

FIG. 1. (Color online) (a) Schematic presentation of the four molecular states (top). Two frames of walking of the molecule are shown by red and black arrow. (b) Schematic of Amemiya model is at the top, and the other three models are developed from the present work, here the red and black balls represent one and zero, respectively. (c) It shows random logical interchange between four DRQ molecules. (d) All 48 molecules inside monolayer is converted to 00, 01, 10, and 11 states. The scale bar is 1.2 nm.

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number assigned to each molecule between two STM scans and this data is used to calculate information entropy.

Instead of calculating entropy of the symmetry of atomic structure,[11] the information entropy or Shannon's entropy (SE) associated with the multilevel states[13] of molecular switches is calculated, which is the entropy of distributed numbers or multivalued logic states.[14]

The generalized SE formulation for a dynamic molecular system developed here is also applicable to any neural system consisting of multilevel switches.[15] At any given time, say N DRQ molecules, occupying states 0, 1, 2, or 3 are randomly distributed (separated by ~9 nm) on the Au (111) surface [Fig. 1(b)]. Then, as the molecular diversity or number of conformer M is 4, the maximum information registered from Shannon expression is $SE=\ln_e(M^N)$ ~5.52 nats for $N=4$ molecules.[13,16] If the surface is scanned by STM at threshold tip biases for conductance switching, all scanned molecules acquire the corresponding logic state i.e., all DRQs switch to the state 0, 1, 2, or 3. Then the information for one molecule represents the other $N-1$ molecules. Thus, the system can have only four distinct information [Fig. 1(d)]. Using expression for SE we get entropy 1.39 nats.

While implementing the SE formulation above, three conceptual changes are adopted in the classical quantum Boltzman machine (QBM) model. Instead of lateral, output is taken vertically via STM tip. The system processes four instead of two decisions (0 and 1) and a dynamic local symmetry[17] is generated here which turns the system significantly advanced than the fixed-site random distribution of QBM [Fig. 1(b)].

When DRQs move and exchange logic state on Au (111) substrate, by assigning four numbers, a matrix could be created for each STM scan. Information in this matrix/string is calculated using Shannon entropy equation $H(V)=-\Sigma_{i=0}^{N-1} p_i \log_2 p_i$ and an average minimum number of bits required to encode a string or matrix (H), which is determined by the frequency of their occurrence (p_i). Here, p_i $[P_i=P_{\mathrm{NDR}}(V)^*P_{\mathrm{Neu}}(V)]$ is the frequency of a particular logic state. P_{NDR} is the probability of a state generated by NDR with a Gaussian peak at the threshold bias V_{th} and is given by $P_{\mathrm{NDR}}=(2\pi\sigma^2)^{-1/2}e^{-(V_{\mathrm{th}}-V)^2/2\pi\sigma^2}$, here σ is $(2\pi)^{-1/2}$. P_{Neu} is the probability of logic state caused by wireless interaction of molecules, considering sigmoid Boltzman neuron, and is represented by $P_{\mathrm{Neu}}=(1+e^{S/C})^{-1}$, where S is weighted sum of input signals, $S=\Sigma_{i=1}^{N-1}x_i w_i+w_0$, x_i is distance between interacting molecules, w_i is energy difference between them, w_0 $(w_0=x_{\mathrm{rms}}V)$ is energy induced by external bias V. The constant C depends on the substrate and it's a function of temperature, sets limiting value of S as S_{min}, at less than S_{min} logic states change [$S\sim60$, Figs. 2(a)–2(c)].

In the Fig. 2(a), change in H with scan bias V, for two molecules participating in the random information exchange process is shown. During continuous scan at <0.4 V, the initial logic state distribution repeats several times as total number of electron τ (state 1 has one and state 2 has two extra electrons) remains constant [Fig. 2(e)]. At higher biases, τ changes randomly and a variable logic state distribution is observed. The H-V variation remained constant at $H=2$ with the change in scan bias. Value of H depicts a 2 bit parallel processing. The higher bias induces faster information exchange increasing the rate of DRQ diffusion (R) and thus number of collisions. Statistically, *rms* distance R trav-

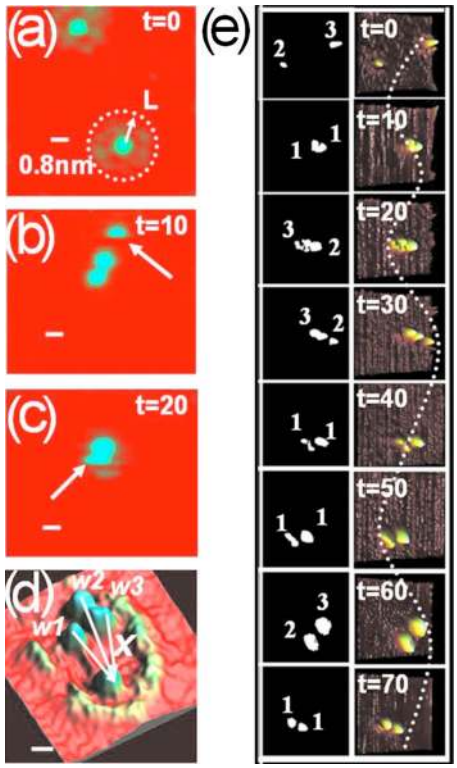

FIG. 2. (Color online) (a) Two isolated DRQ molecules at ~5 nm apart (top at state 2 and the bottom at state 3), surrounded by electron density barriers. [(b) and (c)] consecutive STM images taken ~10 s apart, arrow indicates sudden change in molecular positions. (d) Four DRQ molecules forming a group. Arrow points toward inner and outer walls of the molecular groups. (e) Eight 3D STM images taken ~10 s apart.

eled by each molecule between two STM scans at two different biases did not change significantly. The same conclusions are validated for three and four molecules.

The fact that, nearly constant R is independent of initial arrangement and number of interacting molecules, demand the existence of a wireless communication, possibly through the surface-propagating electron density wave. An apparent electron density wave boundary surrounds the molecules and prohibits surface electron density wave to interfere inside. Thus molecular network remains thermodynamically isolated (at constant $T\sim100$ K) which ensures collective information exchange among them. Inside, ripple period L around DRQ is changed particularly with its logic state. However, area within the boundary remained constant at ~50 nm^2 with the scan-bias variation [Fig. 2(d)]. This is an additional evidence for the wireless communication rather than physical collision induced diffusion.

Randomly connected wireless DRQ network exhibits a periodic and chaotic dynamics[7] at different local parts of the distribution, i.e., confined within a few participants. In Fig. 3(a), fraction of a network (F) is plotted with either mixed (periodic or chaotic) or purely chaotic dynamics with node-to-node connection probability (p) for 2, 3, and 4 molecules. Onset of $F=1$, beyond four molecular neurons support the fact that local clustering of DRQ neurons plays an important role in defining dynamical properties of a wireless network.[7] Thus, *one-to-many* weak and strong wireless connections among DRQ clusters enable mimicking the continuous firing of a real neural net and thus MNN satisfies Zhingulin's model consistently.

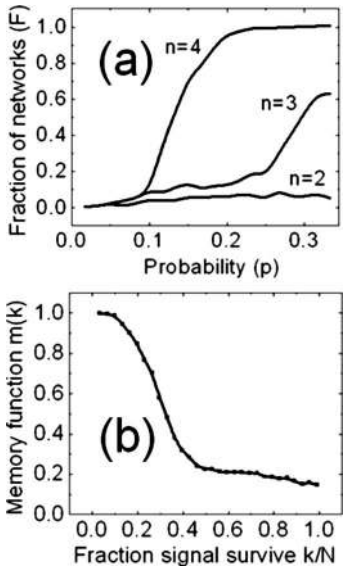

FIG. 3. (a) Using cANN networks, we have plotted fraction of networks (F) with periodic (red) or chaotic dynamic for three neurons and only chaotic dynamics with node to node connection probability (p) for the four molecules. (b) Shows the memory function $m(k)$ with the fraction of the input signals survived (k/N) within the symmetric region.

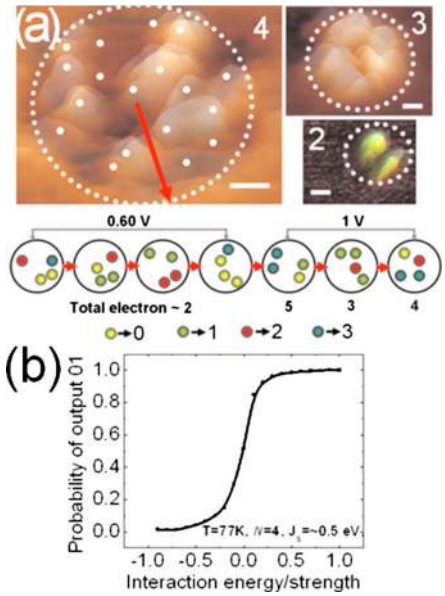

FIG. 4. (Color online) (a) Change in logic states for four molecules in eight events with 30% transparency for 2, 3, and 4 DRQ molecule. The radius of these molecules is 4, 7, and 10 Å, respectively. The scale bar is of 3.5 Å. White dots depict central position of DRQ molecules. (b) It shows the probability of particular state with the ratio of input energy (U) and exchange interaction coefficient (J) for four DRQ neurons.

Temporary memory storage capacity of a DRQ neural network is determined during continuous logic state exchange process of participants inside an electron density wave boundary. Matrices produced without switching any DRQ molecule externally are recorded, and then the same procedure is repeated by forcefully changing one DRQ's state. Finally, the memory function $m(k)$ $[m(k)=\alpha^k/\alpha^k+\varepsilon^1(1-\alpha^{k+1})]$ (Ref. 8) is plotted with the fraction of the input signals survived k/N, $(k=0,1,2\ldots N$, here $N=4)$ within the symmetric region in Fig. 3(b), where α varies from 0 to 1, ε is the variance constant and $\varepsilon^1=\varepsilon/(1-\alpha)$ (Ref. 8). Therefore, to test collective memory, formulation used by White *et al.*[8] is directly implemented.

In Fig. 4(a), change in connectivity of the neural net is plotted with time which shows that even if DRQs acquire states randomly, their motion is always confined within a sphere of diameter (number of molecule × dimension of molecule). For $N=2$, 3, and 4 molecules and for a given input logic set, *N input –N output* neural net is realized. To develop a general operational model, it is considered that a DRQ neuron should produce a particular output with the fourth power of the sigmoid probability function.

As classical QBMs have N input–one output configuration, to model an N input–N output processing device, we have parallely coupled N number of independent QBM nets. Considering homogeneous coupling, the results of one QBM is extrapolated to form a N input–N output neural net, this approach leads to a generalized parallel processor. In Fig. 4(b), the probability of particular distribution [Fig. 2(c)] is plotted with the ratio of input energy and exchange interaction coefficient for four DRQ neurons. These parameters are calculated as an output of QBM, where all molecules interact following a Hubbard model (spin is replaced by an imaginary electronic charge for the states $00=+2$, $01=-1$, $10=+1$, and $11=-2$), with the same interaction coefficients.[4,5] The sigmoid nature leads to a collective logical output of the N, N processing surface, which is an essential requirement to practically realize the peculiar brain function and the neural network formulation proposed by Zhingulin[7] and White *et al.*,[8] respectively.

Here, in a naturally formed thermodynamically quasi-closed system, we have observed a DRQ based N input and N output neural network which is essential for an emergent logical operation. This operation has now been extended for $N=730$ molecules, which increases the processing capability of existing QBMs by several orders in magnitude. Thus, the journey for molecular bioprocessing begins with the realization of the smallest neural net generated by spontaneous interactions of four DRQ molecular neurons.

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