INTERNATIONAL JOURNAL OF CIRCUIT THEORY AND APPLICATIONS
Int. J. Circ. Theor. Appl. 2001; **29**:73–91

Computing architecture composed of next-neighbour-coupled optically pumped nanodevices

György Csaba[1], Árpád Csurgay[2,‡] and Wolfgang Porod[1,*,†]

[1]*Center for Nano Science and Technology, University of Notre Dame, 275 Fitzpatrick Hall, Notre Dame, IN 46556-5637, U.S.A.*
[2]*Department of Electromagnetic Theory, Budapest University of Technology and Economics, Egry József utca 18, Budapest H-1111, Hungary*

SUMMARY

A new computing architecture composed of Coulomb-coupled optically pumped two-state nanodevices is proposed. A new four-level device, built from two coupled two-state devices called 'molecular pair', is introduced. Simulations on the equivalent circuit models of two-dimensional arrays composed of molecular pairs suggest that non-reciprocal pipelining wires, ring oscillators and NAND gates can be built. It is shown that the optically pumped molecular pairs do not stack in meta-stable states, and they restore logic levels. Copyright © 2001 John Wiley & Sons, Ltd.

KEY WORDS: nanotechnology; molecular electronics; coherent control; device modelling; circuit simulation

1. INTRODUCTION

Techniques for modelling and simulation of nanodevice arrays composed of Coulomb-coupled molecules or 'artificial molecules' have been developed [1]. In this paper a new computing architecture composed of Coulomb-coupled optically pumped two-state nanodevices is proposed.

A new four-level device built from two coupled two-state molecules, called 'molecular pair', is introduced. The properties of the two molecules in a pair are designed to be different. We design the molecular pair in such a way that the first molecule of the pair is sensitive to optical excitation and its relaxation to its stationary state is slow. On the other hand the second molecule, called 'dissipative', is not sensitive to the optical excitation, and its relaxation is a few times faster. These conditions can be achieved by choosing the frequency of the excitation

*Correspondence to: Wolfgang Porod, Center for Nano Science and Technology, University of Notre Dame, 275 Fitzpatrick Hall, Notre Dame, IN 46556-5637, U.S.A.
†E-mail: porod@nd.edu
‡Currently at University of Notre Dame; on leave from Budapest University of Technology and Economics.

Contract/grant sponsor: National Science Foundation; Contract/grant number: INT96-15497
Contract/grant sponsor: Hungarian Academy of Sciences.

Received 28 May 2000
Revised 10 July 2000

very close to the transition frequency of the first molecule, but relatively far from that of the second molecule. The two molecules in a pair are Coulomb-coupled, and we assume that the pair has only two stationary states belonging to logic state '0' and '1'.

It is known [2,3] that population in two-state quantum systems can be inverted adiabatically with the application of a resonant external electromagnetic field, called 'optical pump'. We show that a molecular pair switches between logic states if it was irradiated with a resonant optical 'flash'. The spectrum of the flash should correspond to the energy splitting of the optically pumped molecule in a pair. After switching the optically sensitive molecule of the pair, the dissipative molecule will follow the optically switched one, because of the Coulomb-coupling between them. The pair relaxes to its switched stationary state, and it stays in it until the next optical flash forces its switch again. We have developed an equivalent circuit for the molecular pair, thus we are in a position to perform simulations on two-dimensional spatial arrays of molecular pairs subjected to a series of optical flashes.

Simulations on the equivalent circuit models suggest that non-reciprocal pipelining wires, ring oscillators and NAND gates can be built. We found that arrays of optically pumped molecular pairs do not stack in meta-stable states, and they restore logic levels. Based on these experiences, in this paper we propose a new computing architecture composed of Coulomb-coupled and optically pumped molecular pairs.

In Section 2 we outline the model used by us for two-state quantum systems subjected to the Coulomb-coupling of its neighbours and the excitation of close to resonant external electromagnetic field [1,4–8]. In Section 3 we introduce the molecular pair, we show its switching properties, and present its equivalent circuit. Section 4 presents three simple case studies on the design of pipelined logic circuits composed of molecular pairs. The layout of the molecular pairs and the spectrum of the optical flashes are presented. Non-reciprocal 'wire', inverter, ring oscillator and NAND gate are the selected examples.

Finally, Section 5 illustrates the design of the molecular pair itself, and scrutinizes about the feasibility of the proposed architecture.

2. MODELLING ARRAYS COMPOSED OF TWO-STATE MOLECULES

The basic element of the architecture proposed in this paper, is not a single Coulomb-coupled QCA-type nanodevice, but a pair of devices, called a 'molecular pair'. It is a four-state quantum system (Figure 1) consisting of two-state nanodevices, referred to as 'molecules'. First, we introduce the model for the two-state molecule, used in this paper.

The term 'molecule', its state characterization, its dynamical model and its equivalent circuit are introduced and used in this paper in accordance with Reference [1] (see in this special issue). Coherence vectors describe the state of two-state molecules. In their pure states molecules possess a Hermitean Hamiltonian

$$\mathbf{H} = \begin{bmatrix} H_{11} & H_{12} \\ H_{12}^* & H_{22} \end{bmatrix} = \begin{bmatrix} 0 & 0 \\ 0 & \Delta E_0 \end{bmatrix} + \mathbf{H}^{(\text{neighbours})} + \mathbf{H}^{(\text{external field})}, \quad (1)$$

where $\mathbf{H}^{(\text{neighbours})}$ represents the perturbation caused by the Coulomb-coupling of the neighbours, and $\mathbf{H}^{(\text{external field})}$ is the contribution of the external electric field generated by an optical pump. Without loss of generality, we assigned zero energy to the first state of the molecule,

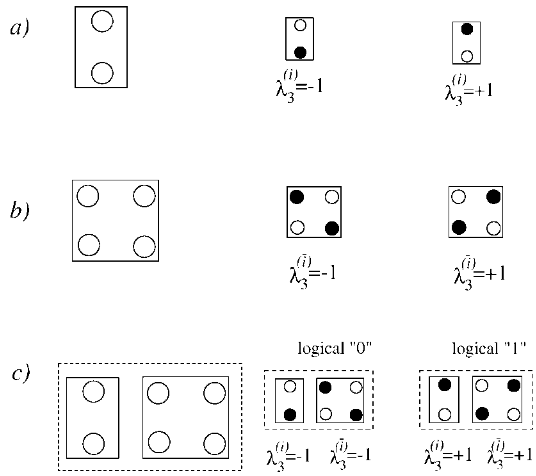

Figure 1. The molecular pair composed of two strongly coupled two-level molecules. The figure illustrates the case, when the optically-driven molecule is a dipole of polarization $\lambda_3^{(i)}$ and the dissipative molecule is a quadrupole of polarization $\lambda_3^{(\bar{i})}$. (a) and (b), respectively show the two stationary states of molecule (i) and $(\bar{i})$, (c) illustrates the two stationary states of the molecular pair.

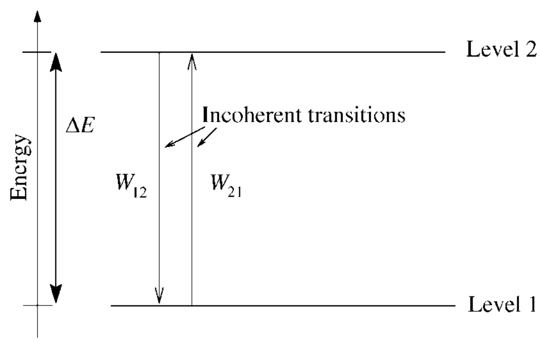

Figure 2. Energy levels of a two-state system. The energy difference between the levels is denoted by ΔE. The incoherent (dissipative) transitions are also marked.

and denoted the energy difference between the unperturbed states by ΔE_0. Figure 2 shows the energy levels of a two-state molecule and marks its dissipative transitions.

In this paper we assume that the neighbours change the energy levels of a molecule only, thus their perturbation is described by a diagonal Hamiltonian. The external field causes the transition between states, thus $\mathbf{H}^{(\text{external field})}$ has only off diagonal elements [1–3]. The dynamics of a two-state molecule, including the damping effects of the environment, can be described by the coherence vector composed of three real variables, $\boldsymbol{\lambda} = [\lambda_1(t), \lambda_2(t), \lambda_3(t)]$ satisfying the generalized Bloch equations [1,4]

$$\hbar\frac{\partial\boldsymbol{\lambda}}{\partial t} = \boldsymbol{\Omega}\boldsymbol{\lambda} + \left.\frac{\partial\boldsymbol{\lambda}}{\partial t}\right|_{\text{dissipation}} = \hbar\frac{\partial\boldsymbol{\lambda}}{\partial t} = \boldsymbol{\Omega}\boldsymbol{\lambda} + \eta\boldsymbol{\lambda} + \xi \tag{2}$$

where

$$\mathbf{\Omega} = \begin{bmatrix} 0 & -H_{22} + H_{11} & -j(H_{12}^* - H_{12}) \\ H_{22} - H_{11} & 0 & -H_{12} - H_{12}^* \\ j(H_{12}^* - H_{12}) & H_{12} + H_{12}^* & 0 \end{bmatrix} \quad (3)$$

The Hamiltonian according to Equation (1) is composed of three terms. The contribution of the external optical pumping is approximated according to the semi-classical model. It describes the interaction energy as a product of the external electrical field and the dipole moment of the two-state molecule. Thus

$$\mathbf{H}^{(\text{external field})} = \begin{bmatrix} 0 & \langle \phi_1 | e\mathbf{d} | \phi_2 \rangle \\ \langle \phi_2 | e\mathbf{d} | \phi_1 \rangle & 0 \end{bmatrix} \mathbf{E} \quad (4)$$

where **d** is the dipole-moment operator of the molecule, φ_1 and φ_2 are the wave functions of states (1) and (2), respectively, and **E** is the electric field at the position of the molecule. In case of monochromatic excitation with slowly varying amplitude the operator takes the form

$$\mathbf{H}^{(\text{external field})} = \begin{bmatrix} 0 & g(t)\sin(\omega t) \\ g(t)\sin(\omega t) & 0 \end{bmatrix} \quad (5)$$

where $g(t)$ depends on the field amplitude, polarization, and transition dipole moment of the molecule. In the following the Fourier transform of $g(t)$ sin ωt will be denoted by $A(\omega)$.

The damping matrix and vector can be approximated (according to the Glauber model [4,5]) as a function of the transition rates between two sates

$$W_{12} = \frac{1}{2\tau}\left(1 - \tanh\left(\frac{+\Delta E}{2\mathrm{k_B}T}\right)\right) \quad (6)$$

and

$$W_{21} = \frac{1}{2\tau}\left(1 - \tanh\left(\frac{-\Delta E}{2\mathrm{k_B}T}\right)\right) \quad (7)$$

as

$$\boldsymbol{\eta} = \begin{bmatrix} -\frac{1}{2}\hbar\,(W_{12} + W_{21}) & 0 & 0 \\ 0 & -\frac{1}{2}\hbar\,(W_{12} + W_{21}) & 0 \\ 0 & 0 & -\hbar\,(W_{12} + W_{21}) \end{bmatrix} \quad (8)$$

and

$$\xi = \begin{bmatrix} 0 \\ 0 \\ \hbar(W_{21} - W_{12}) \end{bmatrix} \quad (9)$$

Note that the occupation probability of states (in case of $t \to \infty$ and no optical excitation) should obey the Boltzmann distribution. This implies for the rates that

$$\frac{W_{12}}{W_{21}} = \exp\left(\frac{\Delta E}{\mathrm{k_B}T}\right) \quad (10)$$

Note that Equations (6) and (7) are consistent with Equation (10). If $|\Delta E| \gg kT$, the transition rate to the upper energy-state is negligible compared to the rate to the lower state ('downward arrow' in Figure 2). It means, that the molecule relaxes to its ground state with time constant τ.

The contribution of the neighbours in Equation (1) is

$$\mathbf{H}^{(\text{neighbours})} = \begin{bmatrix} 0 & 0 \\ 0 & \sum_{j\in\text{neighbours}} E_0^{ij} + \sum_{j\in\text{neighbour}} E_{\text{int}}^{ij} \lambda_3^j \end{bmatrix} \tag{11}$$

The coefficients E_0^{ij} and E_{int}^{ij} are time independent constants, and they depend only on geometry (i.e. the internal geometry of molecules and their relative position in the molecular cluster).

Obviously, the Coulomb interactions are reciprocal, thus

$$E_{\text{int}}^{ij} = E_{\text{int}}^{ji} \tag{12}$$

We assume that the molecules are neutral, they are not ions. Thus, the interaction energy is a result of dipole–dipole, dipole–qaudrupole, quadrupole–quadrupole, etc., interactions. If the intermolecular distance is r then the dipole–dipole coupling weakens with $1/r^3$, and the quadrupole–quadrupole interaction with $1/r^5$. Therefore, to achieve the given accuracy while calculating the effect of neighbours, it is always enough to extend the summation for a finite number of nearest molecules.

The molecular model introduced above can be applied to a molecular array. It leads to a set of coupled ordinary nonlinear differential equations. If the number of molecules is n, the number of equations is $3n$. The explicit form of the three Bloch equations for the ith molecule is

$$\hbar \begin{bmatrix} \dfrac{\mathrm{d}\lambda_1^i}{\mathrm{d}t} \\ \dfrac{\mathrm{d}\lambda_2^i}{\mathrm{d}t} \\ \dfrac{\mathrm{d}\lambda_3^i}{\mathrm{d}t} \end{bmatrix} = \begin{bmatrix} -\frac{1}{2}\hbar(W_{12}^i + W_{21}^i) & \left[\Delta E_0 + \sum_{j\in\text{neighbours}} E_0^{ij} + \sum_{j\in\text{neighbours}} \lambda_3^{(j)} E_{\text{int}}^{ij}\right] & 0 \\ -\left[\Delta E_0 + \sum_{j\in\text{neighbours}} E_0^{ij} + \sum_{j\in\text{neighbours}} \lambda_3^{(j)} E_{\text{int}}^{ij}\right] & -\frac{1}{2}\hbar(W_{12}^i + W_{21}^i) & 2g(t)\sin(\omega t) \\ 0 & -2g(t)\sin(\omega t) & -\hbar(W_{12}^i + W_{21}^i) \end{bmatrix} \begin{bmatrix} \lambda_1^i \\ \lambda_2^i \\ \lambda_3^i \end{bmatrix} + \begin{bmatrix} 0 \\ 0 \\ \hbar(W_{21}^i - W_{12}^i) \end{bmatrix} \tag{13}$$

3. THE MOLECULAR PAIR AS A SWITCH

The basic element of the architecture proposed in this paper, is not a single molecule but a 'molecular pair', a four-level quantum system, consisting of two-state molecules. The properties of the two molecules in a pair are different. We design the molecular pair in such a way that the first molecule of the pair is sensitive to the optical excitation and its relaxation to its stationary state is slow. On the other hand, the second molecule is not sensitive to the optical excitation, and its relaxation is a few times faster. (The first, optically sensitive molecule in a pair #i will be referred to as i, and the second, the so-called 'dissipative' molecule will be referred to as $\bar{i}$.)

These conditions can be achieved by choosing the frequency of the excitation very close to the transition frequency of the first molecule, but relatively far from that of the second molecule. Alternatively, we can design the transitional dipole moment of the second molecule perpendicular to the polarization of the electric filed to make transitions $1 \rightarrow 2$ and $2 \rightarrow 1$ in that molecule to be optically forbidden.

The state of the ith molecular pair depends on six real variables, $\lambda_1^{(i)}(t)$, $\lambda_2^{(i)}(t)$ and $\lambda_3^{(i)}(t)$ describing the optically pumped, and $\lambda_1^{(\bar{i})}(t)$, $\lambda_2^{(\bar{i})}(t)$ and $\lambda_3^{(\bar{i})}(t)$ characterizing the dissipative molecule in the pair. If an individual pair is in stationary state

$$\frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{\lambda}^{(i)} = \frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{\lambda}^{(\bar{i})} = 0$$

and the now six-dimensional Equation (13) has two stationary solutions. In both solutions $\lambda_1^{(\bar{i})}(t) = \lambda_2^{(\bar{i})}(t) = \lambda_1^{(i)}(t) = \lambda_2^{(i)}(t) = 0$, and

$$\left.\begin{aligned} \lambda_3^{(i)} &= -\tanh\left(2\frac{\Delta E_0^{(i)} + E_0^{(i)} + \lambda_3^{(\bar{i})} E_{\text{int}}^{(i,\bar{i})}}{k_{\text{B}} T}\right) \\ \lambda_3^{(\bar{i})} &= -\tanh\left(2\frac{\Delta E_0^{(\bar{i})} + E_0^{(\bar{i})} + \lambda_3^{(i)} E_{\text{int}}^{(i,\bar{i})}}{k_{\text{B}} T}\right) \end{aligned}\right\} \tag{14}$$

If the following three conditions are met, i.e.

$$\left.\begin{aligned} &\text{(a)} \quad |\Delta E_0^{(i)} + E_0^{(i,\bar{i})}| \ll |E_{\text{int}}^{(i,\bar{i})}| \\ &\text{(b)} \quad |\Delta E_0^{(\bar{i})} + E_0^{(i,\bar{i})}| \ll |E_{\text{int}}^{(i,\bar{i})}| \\ &\text{(c)} \quad k_{\text{B}} T \ll |E_{\text{int}}^{(i,\bar{i})}| \end{aligned}\right\} \tag{15}$$

then the stationary solutions could be well approximated by

$$\lambda_3^{(i)} = -1 \text{ and } \lambda_3^{(\bar{i})} = -1 \quad \text{or by} \quad \lambda_3^{(i)} = 1 \text{ and } \lambda_3^{(\bar{i})} = 1 \tag{16}$$

Note that, without loss of generality, we could assume that $E_{\text{int}}^{(i,\bar{i})}$ is always negative.

We will assign logical values to the two stationary states. Let the configuration described by $\lambda_3^{(i)} = \lambda_3^{(\bar{i})} = -1$ be the logical '0', and let the state characterized by $\lambda_3^{(i)} = \lambda_3^{(\bar{i})} = +1$ be the logical '1'. Note that according to Equation (14) the stationary state of each member of a molecular pair is determined by its pair. We have already illustrated these stationary states in Figure 1 in case of a dipole–quadrupole pair. Note that due to the highly non-linear character of the stationary $\lambda_3^{(i)}[\lambda_3^{(\bar{i})}]$ and $\lambda_3^{(\bar{i})}[\lambda_3^{(i)}]$ functions (see Equation (14)), the approximation given for the stationary states are very good. If the energy levels of the molecules are not degenerated ($\Delta E_0 \neq 0$) than only one of the two states in Equation (16) is the ground state of the molecular pair.

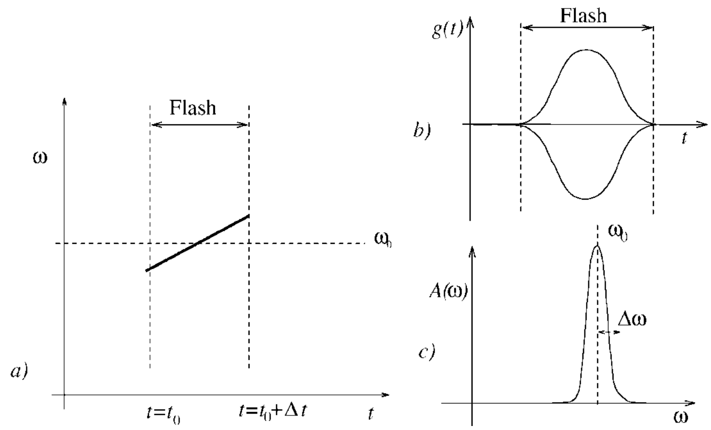

Figure 3. (a) The frequency of the optical chirp as the function of time; (b) the $g(t)$ envelope function of the chirp; (c) the $A(\omega)$ spectrum of the chirp.

The selective sensitivity for the optical excitation requires coupling only to the optically pumped molecule, and no coupling at all to the dissipative molecule, thus $g_{\bar{i}}$ should be zero, and $g_i(t)$ should be equal to the envelope function of the optical excitation. Thus the design of the optical excitation and its coupling to the molecules should insure that

$$g_i(t) \equiv g(t) \neq 0 \quad \text{and} \quad g_{\bar{i}} \equiv 0 \tag{17}$$

The relaxation time of the optically pumped molecule should be a few times greater than the relaxation times of the dissipative molecule, i.e.

$$\tau_i \gg \tau_{\bar{i}} \tag{18}$$

The technique of 'adiabatic inversion' will be used to switch a molecular pair from one logic state to the other. This will be done by inverting the population of a two-level system [6–8].

We irradiate the molecules by a monochromatic laser field. We vary the frequency of the beam slowly in time, while crossing the resonant frequency of the molecule, $\Delta E^{(i)}/\hbar$. In Figure 3 the frequency, the time-dependence, and the spectrum of the simplest inverting optical beam are illustrated.

We introduce a 'detuning' parameter, δ, to characterize the deviation of the resonant frequency from the applied instantaneous frequency

$$\delta = \frac{|\Delta E^{(i)}|}{\hbar} - \omega_0 \tag{19}$$

It can be shown by an approximate analytic solution of Equation (13) that by slowly varying the detuning $g^* = g/\hbar$

$$\text{from} \quad \frac{\delta}{g^*} \ll -1 \ \text{ to } \ \frac{\delta}{g^*} \gg +1 \tag{20}$$

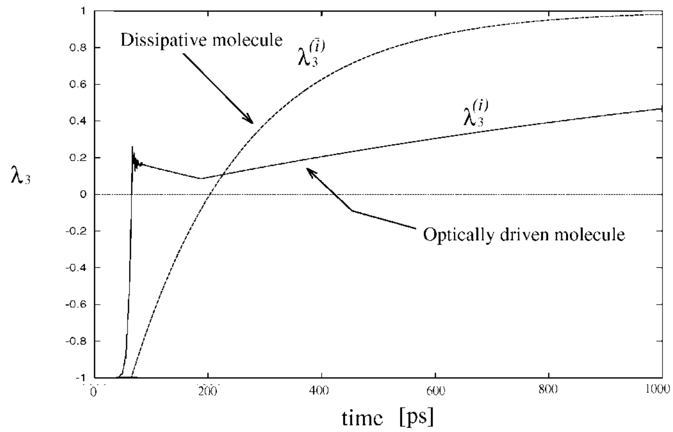

Figure 4. The molecular pair switches from stationary state $\lambda_3 = -1$ to the other stationary state $\lambda_3 = +1$. The optically-driven molecule switches its $\lambda_3^{(i)}$ state fast, and the dissipative molecule's state $\lambda_3^{(\bar{i})}$ slowly follows it.

the population of the molecule is inverted, and the sign of $\lambda_3^{(i)}$ changes. In order to achieve this, we must ensure that the 'chirping' will be slow enough to fulfill the adiabatic condition

$$\frac{1}{\lambda_3}\frac{\partial \lambda_3}{\partial t} \ll g^{*^2} + \delta^2 \tag{21}$$

but fast enough to minimize the effects of dissipation during the process.

In case of a molecular pair, the beam inverts the optically sensitive molecule. However, the ground state of the dissipative molecule is determined by its inverted closest neighbour, thus the relaxation of the dissipative molecule will follow the optically sensitive molecule. Therefore, after some time, the pair as a whole relaxes into its new stationary state. This justifies the $\tau_i \gg \tau_{\bar{i}}$ condition. The molecular pair would not switch if the dissipative molecule did not relax faster then the optically driven one, because the optically driven molecule decays before it could have switched the pair.

Figure 4 shows the polarization of both molecules $\lambda_3^{(i)}$ and $\lambda_3^{(\bar{i})}$, as the function of time. Note that due to the strongly nonlinear character of $\lambda_3^{(i)}[\lambda_3^{(\bar{i})}]$ given in Equation (14), a 'polarization gain' occurs. Even if the left molecule is inverted into a partially polarized state ($|\lambda_3^{(i)}| \ll 1$), the final state of the pair will be highly polarized ($|\lambda_3^{(i)}| \approx |\lambda_3^{(\bar{i})}| \approx 1$).

Consider an array of molecular pairs. The energy differences (ΔE) in any of the pairs depend on the state of its neighbours. Thus, the resonant frequency of a pair will be determined by the state of the pair's neighbours, because it is equal to the resonant frequency of its optically sensitive molecule. If we excite the whole array by a laser flash, all pairs, whose resonant frequency is matched by the frequency of the optical excitation, will be switched into a new state. However, all other pairs will stay in their original state, because their resonant frequencies are far from the frequency of the excitation.

In this paper, we suggest that a new type of computer architecture can be based on this very fact. First, we clarify the conditions of switching molecular pairs. Next, we demonstrate that logic input states can be pipelined through wires of molecular pairs by proper design of the spectrum of the optical excitation. Then we show that the same wire can alternate the logic states of molecular pairs in a sequence of them. This leads to the demonstration of a 'ring-oscillator'-type arrangement of molecular pairs. We also show that a NAND gate can be built from molecular pairs excited by a proper optical spectrum.

In all examples, an array of molecular pairs will be irradiated with a laser beam. The beam will be designed such that its composition from chirps with different frequencies insures the switching of those, and only those, pairs which should switch in order to realize a prescribed functionality. The contribution to the Hamiltonian of the external field composed of chirps with different frequencies, reads as

$$\mathbf{H}^{\text{exc}} = \begin{bmatrix} 0 & \sum\limits_k g_k(t)\sin(\omega_k t) \\ \sum\limits_k g_k(t)\sin(\omega_k t) & 0 \end{bmatrix} \tag{22}$$

In general, only beams with frequencies close to resonance have considerable effect on the state of molecules. Thus we choose the frequencies of the chirps such that $g_k(t)$ is non-zero only for a finite Δt time interval (during the flash), and at this time interval ω_k varies from $\omega_k^0 - \Delta\omega$ to $\omega_k^0 - \Delta\omega$, where $\Delta\omega$ should satisfy the adiabatic condition (21). By this choice we can adiabatically invert certain molecular pairs, and leave the state of all the others unchanged.

In case of a selected pair there are three possibilities depending on the state of the neighbours:

- The resonant frequency of the molecule is far from the frequency of each laser beam, i.e. $\omega_k^0 - \Delta\omega < \Delta E/\hbar < \omega_k^0 + \Delta\omega$ is not satisfied for any ω_k. In this case the molecular pair does not change its state;
- The resonant frequency of the molecular pair is close to the frequency of one of the laser beams, i.e. $\omega_k \approx \Delta E/\hbar$ is satisfied. In this case the molecular pair switches to its other stationary state;
- The flash initiates a transient in the molecular pair, but its effect is not strong enough to switch the pair, thus the pair relaxes back into its original state.

In order to study the transients of switching in a circuit composed of a few molecular pairs we shall introduce the equivalent circuit of the molecular pairs. Equivalent circuits of Coulomb-coupled and optically excited molecules have been introduced in Reference [1]. Here we apply the results for the molecular pair, and for circuits composed of pairs. Note that in the framework of the approximations, we are relying on the fact that the equivalent circuits are independent of the embedding circuits. Thus equivalent circuit models can be used to build models for integrated arrays, and thus simulation of arrays composed of molecular pairs can be performed.

In Figure 5 we show four pairs coupled to each other. Pair #1 is the one, whose state equations and circuit model is used as an example. The resonant frequency of pair #1 is equal to the resonant frequency of molecule (1) in the pair. This is determined mainly by the Coulomb field of its dissipative pair, molecule ($\bar{1}$), i.e. by the interaction energy between molecule 1 and $\bar{1}$, which is denoted as $E_{\text{int}}^{1,\bar{1}}$. But the interaction energies of the neighbouring pairs, $E_{\text{int}}^{1,2}, E_{\text{int}}^{1,\bar{2}}, E_{\text{int}}^{1,3}, E_{\text{int}}^{1,\bar{3}}, E_{\text{int}}^{1,4}, E_{\text{int}}^{1,\bar{4}}$, perturb the energy levels of molecule #1 as well.

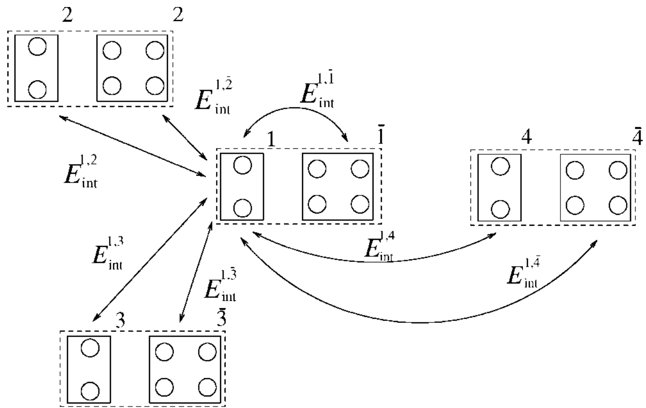

Figure 5. A molecular pair is subjected to its neighbours electrostatic field.

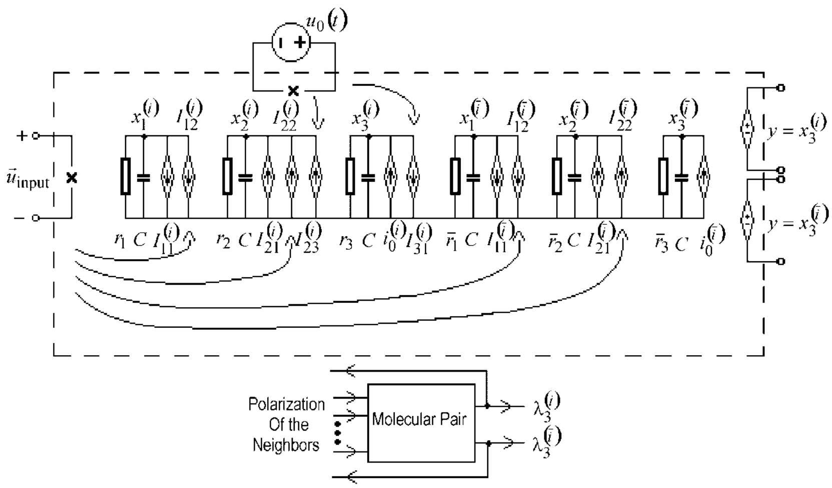

Figure 6. Equivalent circuit of a molecular pair.

The equivalent circuit of a molecular pair should represent the same dynamics as two sets of generalized Bloch equations of type (13). One set of equations stands for the dynamics of the optically driven, the other set for the dissipative molecule. To describe the dynamics of a pair we need six state variables, the six components of the two coherence vectors.

Figure 6 shows the equivalent circuit of molecular pair #i. The six state variables of the molecular pair are the components of the coherence vectors multiplied by (e/C), where e is the electron's charge, C is the given capacitance

$$\begin{aligned} x_1^{(i)} &= (e/C)\lambda_1^{(i)}, \quad x_2^{(i)} = (e/C)\lambda_2^{(i)}, \quad x_3^{(i)} = (e/C)\lambda_3^{(i)} \\ x_1^{(\bar{i})} &= (e/C)\lambda_1^{(\bar{i})}, \quad x_2^{(\bar{i})} = (e/C)\lambda_2^{(\bar{i})}, \quad x_3^{(\bar{i})} = (e/C)\lambda_3^{(\bar{i})} \end{aligned} \tag{23}$$

Note that the variables related to the optically-driven molecule are indexed again as (i), those related to the dissipative molecule as $(\bar{i})$. The variables $x_3^{(i)}$ and $x_3^{(\bar{i})}$ have a distinguished role in Coulomb-coupling, thus they will be called 'polarization voltages'. The input ports of the #ith pair are assigned to the polarization-voltages of the neighbours. The two output ports are the polarization voltages of the optically driven molecule $x_3^{(i)}$, and that of the dissipative molecule $x_3^{(\bar{i})}$. The excitation of the external field is represented as an independent voltage generator $u_0(t) = (1/e)\sum_k g_k(t)\cdot \sin \omega_k t$. The neighbours polarization voltages control two controlled sources of the optically driven and two controlled sources of the dissipative molecules. Using the notations of Figure 6, the values of the circuit elements can be learned from the state Equations (13), applied to the optically driven and to the dissipative molecule. This means that

$$r_1 = r_2 = 2\tau_i/C, \quad r_3 = \tau_i/C, \quad \bar{r}_1 = \bar{r}_2 = 2\tau_{\bar{i}}/C, \quad \bar{r}_3 = \tau_{\bar{i}}/C, \;\; C = 1$$

The two current generators, representing the damping vectors, 'attract' the dynamics to relax into ground states. Their source currents are

$$i_0^{(i)} = \frac{e}{\tau_i} \tanh\left(\frac{\Delta E^{(i)}}{2k_B T}\right) \tag{24}$$

$$i_0^{(\bar{i})} = \frac{e}{\tau_{\bar{i}}} \tanh\left(\frac{\Delta E^{(\bar{i})}}{2k_B T}\right) \tag{25}$$

There are three types of voltage-controlled-current generators in the equivalent circuit. The external field controls two of them, $I_{23}^{(i)}$ and $I_{31}^{(i)}$,

$$I_{23}^{(i)} = 2\frac{eC}{\hbar} u_0 x_3^{(i)} \quad \text{and} \quad I_{31}^{(i)} = 2\frac{eC}{\hbar} u_0 x_2^{(i)} \tag{26}$$

We shall present the source currents of the further voltage-controlled-current generators using the notation

$$Y^{ij}(x_3^j) = \frac{C}{\hbar}\left(E_0^{ij} + \frac{C}{e} E_{\text{int}}^{ij} x_3^j\right) \tag{27}$$

The voltage-controlled-current-sources relating the internal polarization voltages of the pair are

$$\begin{aligned} I_{11}^{i} &= Y^{i\bar{i}}(x_3^{\bar{i}}) x_2^{i}, \qquad I_{21}^{i} = Y^{i\bar{i}}(x_3^{\bar{i}}) x_1^{i}, \\ I_{11}^{\bar{i}} &= Y^{\bar{i}i}(x_3^{i}) x_2^{\bar{i}}, \qquad I_{21}^{\bar{i}} = Y^{\bar{i}i}(x_3^{i}) x_1^{\bar{i}} \end{aligned} \tag{28}$$

The current sources controlled by the polarization voltages of the neighbours are

$$\begin{aligned} I_{11}^{i} &= \sum_{j\,\in\,\text{neighbours}} Y^{\bar{i}j}(x_3^j) x_2^{i}, \qquad I_{21}^{i} = \sum_{j\in\text{neighbours}} Y^{ij}(x_3^j) x_1^{i}, \\ I_{11}^{\bar{i}} &= \sum_{j\in\text{neighbours}} Y^{\bar{i}j}(x_3^j) x_2^{\bar{i}}, \qquad I_{21}^{\bar{i}} = \sum_{j\in\text{neighbours}} Y^{ij}(x_3^j) x_2^{\bar{i}} \end{aligned} \tag{29}$$

The equivalent circuit of the optically pumped molecular pair is an open system with as many input polarization voltages as the number of neighbours with significant influence on energy splitting in pair #i, and two output voltages, the polarization vectors of the optically driven and that of the dissipative molecule, (i) and ($\bar{i}$), respectively. In the framework of a fixed array type, the equivalent circuit is invariant on interconnection. In Figure 6 we introduced a symbol to represent a molecular pair.

4. COMPUTING WITH MOLECULAR PAIRS

Let us recall that the energy splitting of the levels in an optically driven molecule is dominated by the effect of its nearest neighbours. If the number of neighbouring molecules is n, then there is maximally 2^{n+1} different energy level splitting (ΔE^i) possible. Since the multipole interactions weaken rapidly with the increasing distance, n is a well- defined number. Symmetries (i.e. relations between the interaction energies) may significantly reduce this number, and since n is no more than three or four, we might say, that ΔE^i has only a few possible discrete values as a function of the state of its neighbours. We are proposing a technique to design the ω_k excitation frequencies and $g_k(t)$ envelop such that useful logic functions could be realized.

We shall demonstrate that it is possible to design a selective optical excitation in such a way that a series of consecutive, flashes irradiated on the whole array, will push the input logic state to the output resulting in the required functionality. In this paper we give examples illustrating the possibility of designing a class of 'pipelined' logic. Figure 7 illustrates the simplest example, a 'wire' composed of molecular pairs. After the wire, we show a 'ring-oscillator'-type arrangement, and a NAND gate, as examples.

4.1. Designing a molecular wire

First, we show that it is possible to design a non-reciprocal molecular wire. Figure 7 shows the layout of such a wire, and Figure 8 shows its circuit model. Consider an arbitrary molecular pair inside the wire. The wire transmits a signal from left to right if the energy splitting of molecular pair #(i) is determined mainly by its left neighbour #(i – 1). Its right neighbours

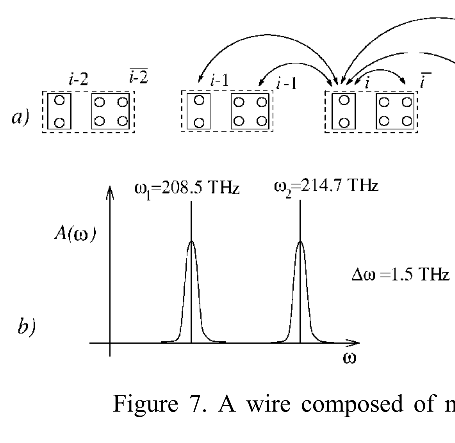

Figure 7. A wire composed of molecular pairs.

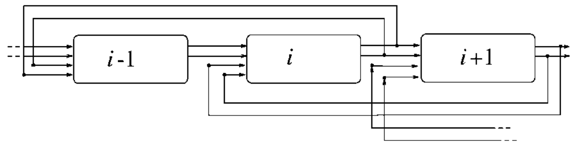

Figure 8. Equivalent circuit of a wire.

Table I.

Initial states of three pairs			States after optical flash #1			States after optical flash #2		
$(i-1)$	(i)	$(i+1)$	$(i-1)$	(i)	(i)	(i)	(i)	$(i+1)$
1	1	1	1	1	1	1	1	1
1	1	0	1	1	1	1	1	1
1	0	0	1	1	0	1	1	1
1	0	1	1	1	0	1	1	1
0	0	1	0	0	0	0	0	0
0	1	0	0	0	1	0	0	0
0	1	1	0	0	1	0	0	0
0	0	0	0	0	0	0	0	0

#$(i+1), (i+2)$, etc., have much smaller influence on the splitting than that of the left neighbour. In this case, the molecular pair #(i) will follow the stationary state of pair #$(i-1)$, if we flash the array by a chirp of properly designed frequency. The energy splitting is defined by the left neighbour, and there is a significant difference in the splitting depending on relative states of pairs #(i) and #$(i-1)$. The difference between the energy levels will significantly differ if both pairs are in the same state or they are in opposite states.

Let us look at a wire composed of molecular pairs, and let all pairs be in one of their stationary states, either in '0' or in '1'. The energy splitting of an optically pumped molecule is determined by all of its neighbours, including the closest molecule, which is its own dissipative pair. The stationary state of the optically pumped and that of the dissipative molecule in each pair is always the same. Nevertheless, the energy splitting of the optically pumped molecule strongly depends on its left neighbour's state as well.

We show that we can design the spectrum of the optical flash in such a way, that repeatedly applying the flash, the logic state of the left molecular pair will propagate from left to right, step by step. Each flash will push the signal to the right by one pair.

In Figure 8 pair #$(i-1)$ is the 'input' and pair #$(i+1)$ is the 'output'. The input state will be pushed to the output by two subsequent flashes. If a right neighbour is in the same state as the pair on its left side, the flash should not have any effect on the optically pumped molecule of the right neighbour. However, if its state is different, then the right neighbour should be inverted with a properly designed 'chirp'. Initially, the three pair of the wire can be in eight different stationary states, illustrated in the first column by the logic states (Table I). The second column shows the states after the first, the third column after the second optical flash:

Figure 9 shows the signal propagation on a wire. The wire initially was in logical '0' state. We switch the first pair on the left edge of the wire to state '1', then we irradiate the wire

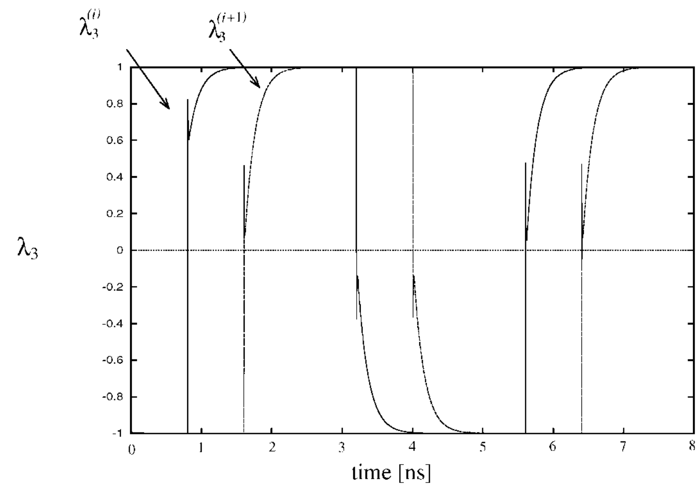

Figure 9. $\lambda_3(t)$ dynamics of two subsequent, optically-driven molecular pairs in a wire.

with consecutive flashes having the spectrum shown in Figure 7. After each flash, the signal steps one step toward the output. After a few flashes we can apply new inputs.

Note that the wire can be designed to be non-reciprocal if condition

$$\left| E_0 + \sum_{j \in \text{input cells}} E_{\text{int}}^{ij} \lambda_3^j \right| \neq \left| E_0 + \sum_{j \in \text{output cells}} E_{\text{int}}^{ij} \lambda_3^j \right| \tag{30}$$

is met for the optically excited molecules for all possible λ_3^j configuration. In this case a molecular pair can 'distinguish' between its input and output. The state of a molecule will not depend on the state of its right neighbour, but depends on its left neighbour.

4.2. Designing a ring oscillator

A ring oscillator is as an example of autonomous circuits. The same structure that realizes a molecular wire, can be redesigned to realize an inverter chain. Let us invert the molecules if their state is the same as their left neighbour's, and let them unchanged, if their state is opposite. The wire becomes and 'inverter-chain'. In ground-state, the subsequent molecular pairs alternate.

We can design a nano-equivalent of the ring oscillator (Figure 10) if we form a loop from a wire consisting of odd number of molecular pairs, by connecting its input to its output. All molecules change their state after each flash. The results of simulation show, that the signal never stops propagating (Figure 11). As in the case of the 'classical' ring oscillator, the waveform may depend on the initial state of the device.

4.3. Designing Boolean logic

The considerations of the previous chapters can be generalized. The basic idea for the realization of an arbitrary binary logic is the following: first, calculate all possible energy splitting,

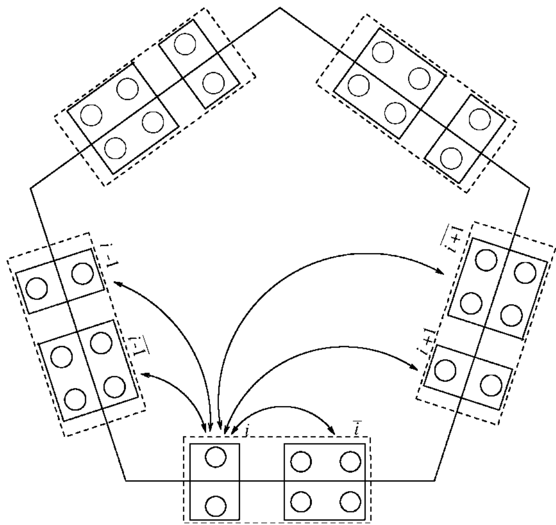

Figure 10. Ring oscillator: a loop (a pentagon) was formed from the wire. After each flash, at least one pair changes its state.

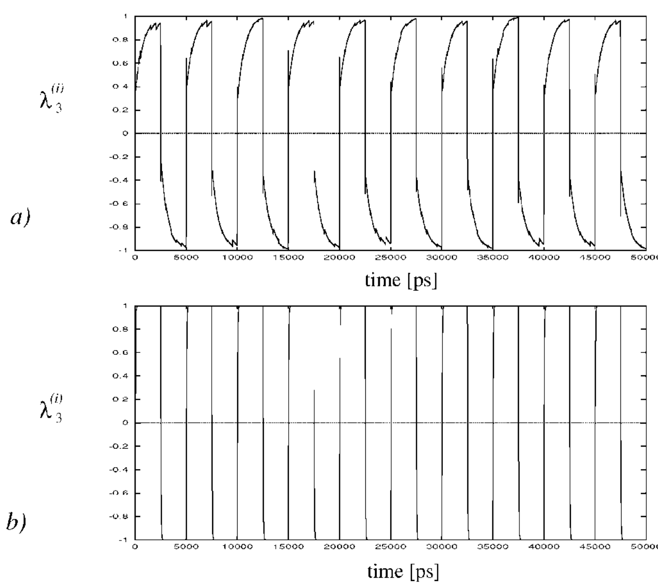

Figure 11. Waveforms (polarization as the function of time) of two subsequent molecules in the ring oscillator. Note how the dissipative molecule restores the signal levels.

ΔE^i, for a molecular pair as function of the state of its neighbours. Then design the spectrum of the optical flash in such a way, that it inverts the state of the molecular pair, if its present state does not satisfy the required logic function, but it does not invert it, if its state is the required one.

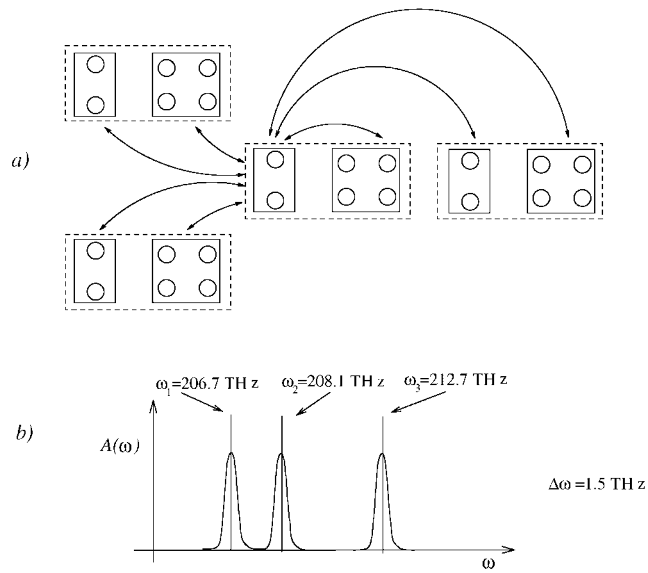

Figure 12. Layout of a NAND gate.

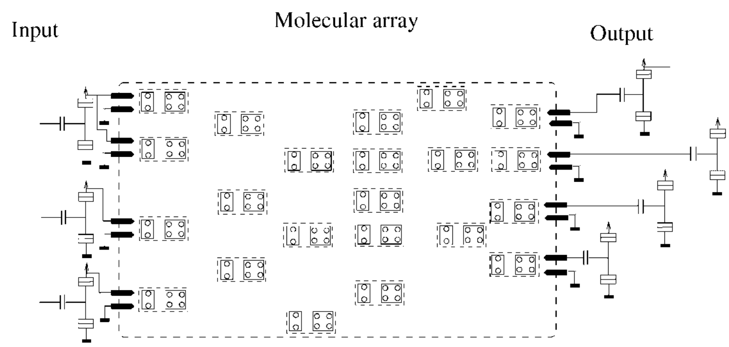

Figure 13. An array composed of optically pumped molecular pairs. Inputs are set by and outputs are measured by Single-Electron Transistors.

We illustrate the procedure by an example, the NAND gate. The proposed layout is shown in Figure 12, together with the optical spectrum performing the necessary inversions.

It was shown in the previous sections, that after each flash the signal ‘steps’ one molecule pair toward the output. The molecules, that the signal passed, should not be left idle: new inputs can be applied. The optical flash acts as a clock signal: after each flash, a new input ‘steps in’ the array, and an output ‘leaves’ the array. The proposed architecture shows a possibility to build highly parallel pipelined logic.

Figure 13 is a schematic of a larger array. The inputs and outputs can be e.g. single-electron transistors.

The steps of computation by an array are the following:

1. Apply new inputs, and read the outputs.
2. Apply an optical flash.
3. Wait, while the system finds its new stationary state.
4. Continue with step 1.

Note that there are no zero-delay elements in this structure, therefore the geometry of the system is in strong connection with the signal path. We do not need additional latches (as in most pipelined structure), since the wire is a memory element.

5. DESIGN OF A MOLECULAR PAIR

In order to illustrate the feasibility of the proposed architecture, in this section we calculate the interaction energies, and the frequencies of the optical excitation for a specific geometry.

Consider the structure illustrated in Figure 14. In this example, the right molecule is a pure quadrupole without transitional dipole moments. There are two electrons in the antipodal dots. The left molecule of the pair has dipole and quadrupole moments, and it can be excited applying a field with polarization having a significant z component. The wavefunction of the electrons is assumed to be well localized in the dots. In this case there is a unique connection between the state of the molecule and the geometry of its wave function.

Parameter d is a scale factor, and Figure 15 shows the interaction energies and the corresponding frequencies as a function of the scaling factor.

A static electric field $\mathbf{E}_0$ is used to 'shift' the energy levels of the optically-driven molecule. It allows the adjustment of the value of ΔE_0.

The driving frequencies for a specific molecular wire can be estimated. Let $d = 4$ nm. The distances between the nearest dipoles and quadrupoles are $2.5d$. In this case the interaction energies are $E_{\text{int}}^{i,i-2} = E_{\text{int}}^{i,i+2} = 0.855$ meV, $E_{\text{int}}^{i,i+1} = -134.99$ meV, $E_{\text{int}}^{i,i+3} = -0.8996$ meV. Therefore $E_{\text{int}}^{\text{input}} = 4.311$ meV, and $E_{\text{int}}^{\text{output}} = -0.04$ meV (Figure 15). The effect of the 'far' molecules are negligible, the interaction energy to the next pair is less by a factor of 20.

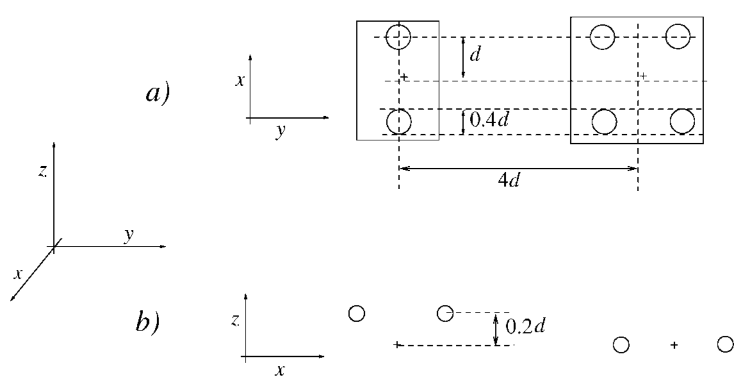

Figure 14. An idealized possibility for implementing a molecular pair.

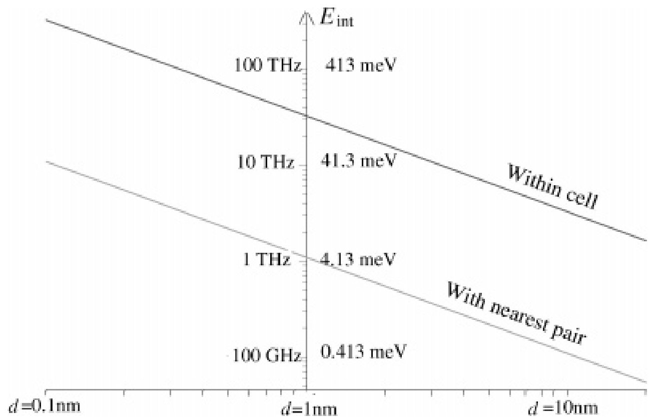

Figure 15. Energy differences and pumping frequencies as functions of d.

Note that in this special geometry the electrostatic effect of the nearest pair at the output side is very small compared to all other energies. This is not true for the input side. The system is non-reciprocal. If $\Delta E_0 = 2$ meV, the frequencies are $\omega_1 = 34.14$ THz, $\omega_2 = 33.18$ THz. Figure 9 shows the simulation results at $T = 100$ K. The coupling between the field and the molecule is $g = 0.25$ meV, and the bandwidth of the chirp is $\Delta\omega = 0.3$ THz. The duration of the chirps is 40 ps.

In the case of a NAND gate, if the distance of the inputs from the switching cell is $2.5d$ from both (x and y) direction, and $d = 1$ nm, then the driving frequencies are $\omega_1 = 33.86$ THz, $\omega_2 = 33.12$ THz and $\omega_3 = 32.89$ THz.

If d is a few nm, than we are in the 'molecular electronics' region, if d is a few tens of nm, than the structure may be realized in solids (artificial molecules).

These results suggest that the selectivity of the optical excitation is good enough to realize the proposed new architecture by solid-state technology, however, the molecular scale offers a much robust way to build these structures. In solid state, the noise of the background charge and the manufacturing inaccuracies could cause serious difficulties. Since the semiconductor structures are much larger than the molecular ones, the interaction energies are smaller, therefore the sensitivity of the system to errors could become critical.

Note that the dissipation mode, applied to individual molecules does not give accurate results for the time evolution of the processes. According to our approximation, the dissipation continuously takes the system into a mixed state. The model does not take into account dissipation in the form of 'quantum jumps' [4], and the model outlined above, describes the average behavior of many molecules. However, the exact time-evolution of dissipation is not crucial from the point of view of the architecture proposed in this paper.

The other approximation of our model is the assumption on the exactly two-state character of the molecules. They are two-state quantum-mechanical systems, and no transitions occur to any other states. It is hard to satisfy this condition, because we can excite additional levels easily with the application of several laser-beams with different frequencies.

Nevertheless, our simulations suggest that it is worth to pursue experimental studies on optically pumped nanoelectronic circuits, and our two-state model can be used to design experimental circuits.

ACKNOWLEDGEMENTS

This work was supported in part by a joint grant of NSF (National Science Foundation INT96-15497) and HAS (Hungarian Academy of Sciences), and also by a grant from the W.M. Keck Foundation. The authors like to acknowledge the stimulating discussions with G.H. Bernstein, D. Hall, G.J. Iafrate, C.S. Lent, A.C. Seabough, and G. Tóth.

REFERENCES

1. Csurgay ÁI, Porod W. Equivalent circuit representation of arrays composed of Coulomb-coupled nano-scale devices: modeling, simulation and realizability. *International Journal of Circuit Theory and Applications* 2000; **29**:3–35.
2. Allen L, Eberly JH. *Optical Resonance and Two Level Atoms*. Wiley: New York, 1975.
3. Meystre P, Sargent M. *Elements of Quantum Optics.* Springer: Berlin, 1995.
4. Mahler G, Weberruss V. *Quantum Networks*. Springer, Berlin, 1995.
5. Glauber RJ. Time-dependent statistics of the ising model. *Journal of Mathematical Physics* 1963; **4**:294.
6. Oreg J, Hioe FT, Eberly JH. Adiabatic following in multilevel systems. *Physics Reviews* A **29**(2).
7. Gordon J, Rice SA. Active control of the dynamic of atoms and molecules. *Annual Review of Physical Chemistry* 1997; **48**:601–641.
8. Mandel L, Wolf E. *Optical Coherence and Quantum Optics*. Cambridge University Press: Cambridge, 1995.
9. Csurgay ÁI, Porod W, Lent CS. Signal processing with next-neighbour-coupled time-varying quantum-dot arrays. *IEEE Transactions on Circuits and Systems* (Part I) 2000; **47**(8).
10. Porod W, Lent CS, Bernstein GH, Orlov AO, Amlani I, Sneider GI, Merz JL. Quantum-dot cellular automata: computing with coupled quantum dots. *International Journal of Electronics* 1999; **86**(5): 549–590.

