D1.2.2 Implementation of parallel computation and use of observables.

M26, WP1

Lead Beneficiary P2 (RDL-HUJI)

Three different implementations and simulations of parallel computing by observables have been demonstrated in PR2, in close collaboration with the systems studied by the experimental partners of **WP2 (**Optical addressing) and **WP3** (Electrical addressing)**.**

The partners of **WP1**, **P1** and **P2** proposed the concept of molecular decision trees in PR1, jointly with **P3** of **WP2**. This design was implemented experimentally in PR2 by a joint work of the partners of **WP1** and **WP2** on a DNA-Rhodamine complex, addressed in a 2D photo echo set up. The design was extended to the electrical addressing of an array of three dopants by a pulse gate voltage in collaboration with **P4** of **WP3**. The third contribution to this deliverable for PR2 is the design in collaboration with **P3** of a parallel computation that takes advantage of all the observables that correspond to phase matching conditions.

A decision tree can be viewed as a graphical representation of the truth table of a function. The design relies on the fact that each time the system interacts with a perturbation, there is a change in time of the observables that is intrinsically parallel: all the observables of the system respond in parallel to the perturbation. The output is a function of the observables and different paths connect in parallel the initial state of the system to the read out. Since the interaction between the system and the perturbation is bilinear, one can implement parallel bilinear classical logical operations. The advantage of computing with observables is that there are N^2-1 matrix elements of the density matrix of a system with N states which completely characterize its time evolution, while there are only N occupation numbers. The scheme is scalable. The number of physical interactions with the perturbation determines the number of logic variables and the number of transitions between states at each interaction the maximum radix for the logic variable.

1. A physical realization based on the 2D photo echo spectroscopy was demonstrated. In this set up, there are three-photon interactions with a chromophoric molecule (the rhodamine dye TAMRA) for which there are four accessible states which leads to 15 observables. Each time a photon interacts with the chromophoric system, four different paths are possible, which leads to quaternary logic or in a simplified version to Boolean logic. We demonstrate the mode of operation of the machine using Lie algebra to describe the light-matter interaction. An example for a three variable Boolean function is given in Figure 1 below. In the case of the DNA-TAMRA complex, the analysis of the photophysics jointly with WP2 and the simulations of the 2D PE maps by **P1** show that the four states are the S0 electronic ground state, two vibrational states of the S1 first excited electronic state (v=0 and v=1) and a higher excited electronic state, S2. The transitions frequencies of the S0-S1 and S1-S2 manifolds are of the same order of magnitude and fall in the experimental spectral window. The characterization of the involvement of excited vibrational states opens a new dimension for the complexity of the logic operations that can be implemented. The involvement of vibrational states increases the number of positions on the 2D maps that corresponds to transitions between states, since the ladder of physically available states is extended to

vibrational states. For example the identification of 8 positions on the map would allow to map all the 256 Boolean functions of 3 variables. Work to harvest the increase of complexity will be pursued in PR3. The simulation of the mode of operation of the molecular decision tree required extensive modelling. The electronic structure of the excited electronic states of the isolated dye was carried out at the quantum chemistry level (TD-DFT) and molecular dynamics simulations were necessary to elucidate the role of environment (solvent and DNA scaffold). The operation of the machine was simulated using a master equation description, taking into account the role of environment in a phenomenological way. The agreement between the computed maps and the experimental ones shown in Figure 1 demonstrates that the interaction of the DNA complex with the 3 laser pulses of the applied sequence corresponds to resolve a Boolean variable at each step, thereby implementing a molecular decision tree. Non Boolean decision tree will be implemented in PR3 in collaboration with WP2 on the DNA-bichromophoric complex under study in this WP.

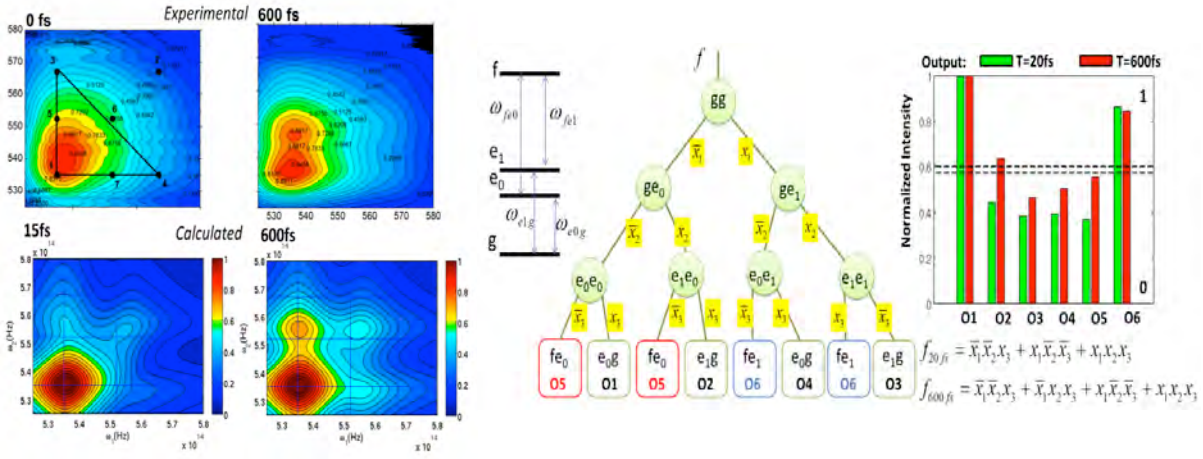

Figure 1. Experimental and computed 2D spectra of the DNA-Tamra complex and molecular logic tree implementation, joint work between **WP1** and **WP2.**

2. In PR2, the design of molecular decision trees was extended to the electrical addressing of an array of exchanged coupled dopant atoms in Si. The design was elaborated in collaboration with WP3. It will be experimentally implemented in PR3. The scheme of the design is shown in Figure 2, together with the corresponding electronic structure. In the electrically addressed decision tree, the perturbation applied is a pulse of gate voltage and the read out will be performed either by sensing of the charge of a tip induced QD, or by direct spatially resolved measurement of the charge on the dots and between the dots by the tip. This second design provides a direct access to the measurement of each observable.

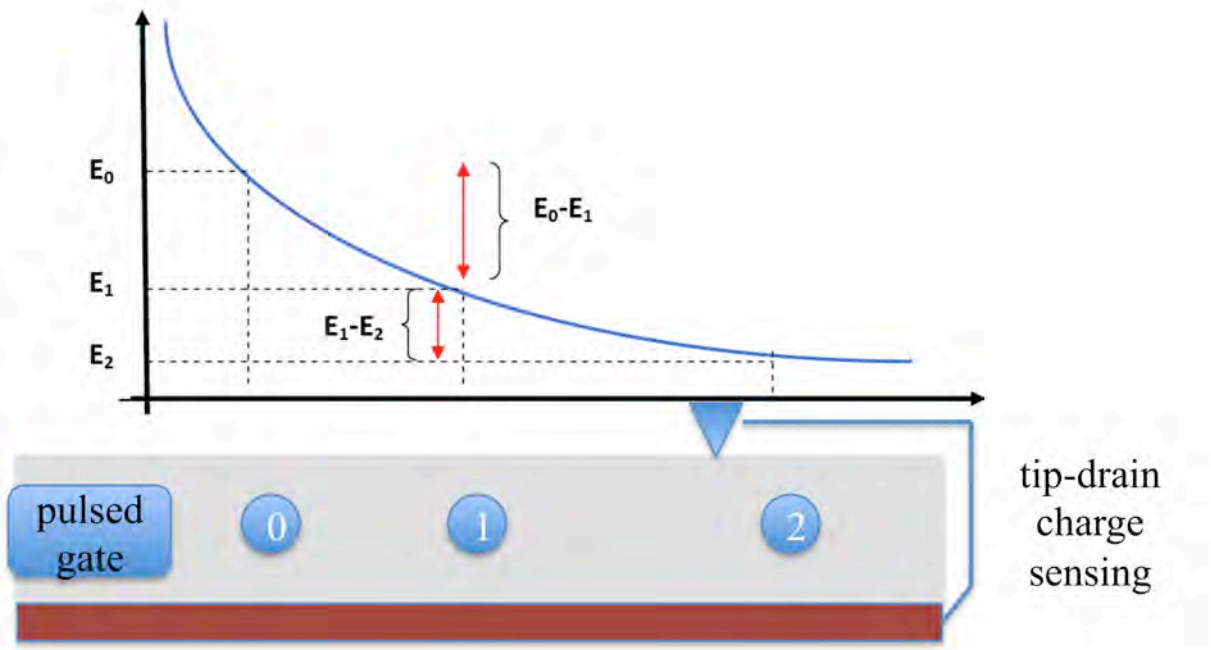

Figure 2. Scheme of the design of an electrically addressed molecular decision tree and associated level structure. A single level is taken into account for each dopant atom.

The Hamiltonian matrix of the exchange coupled three dopant system is given by a 3x3 tridiagonal matrix. Unlike in the optical case, here, the input provided by the pulse of gate voltage is changing the energy of the levels of the dopant, the largest change being for the dopant next to the gate, as shown in Figure 2. The exchange coupling between the dopants is fixed and determined by the distance between them. This coupling varies exponentially with the inter-dopant distance. In order to be able to control the charge on the dopant and between them, the dopants should be placed at unequal distances, so that dopant 0 and 1 are more exchanged coupled than dopants 1 and 2. For this system, there are 8 observables: two populations on the dots, the third one being given by the normalization condition and three coherences which are complex numbers. In the linear arrangement shown in Fig. 2, the experimental read out by the tip-drain voltage directly probes 5 of these observables, 3 charge on the dot and 2 charge between the dots, that corresponds to bond orders. The bond orders correspond to the real part of the coherence. From a physics point of view, the essential difference is that the perturbation applied is not in the perturbative regime so there are no selection rules for the coupling. From a logic point of view, it translates into the fact that the five observables are in principle addressed simultaneously at each interaction, which leads in principle to a radix of 5 for the logic variable. An example is shown in figure 3 below. These preliminary results will be finalized in PR3, in collaboration with WP3.

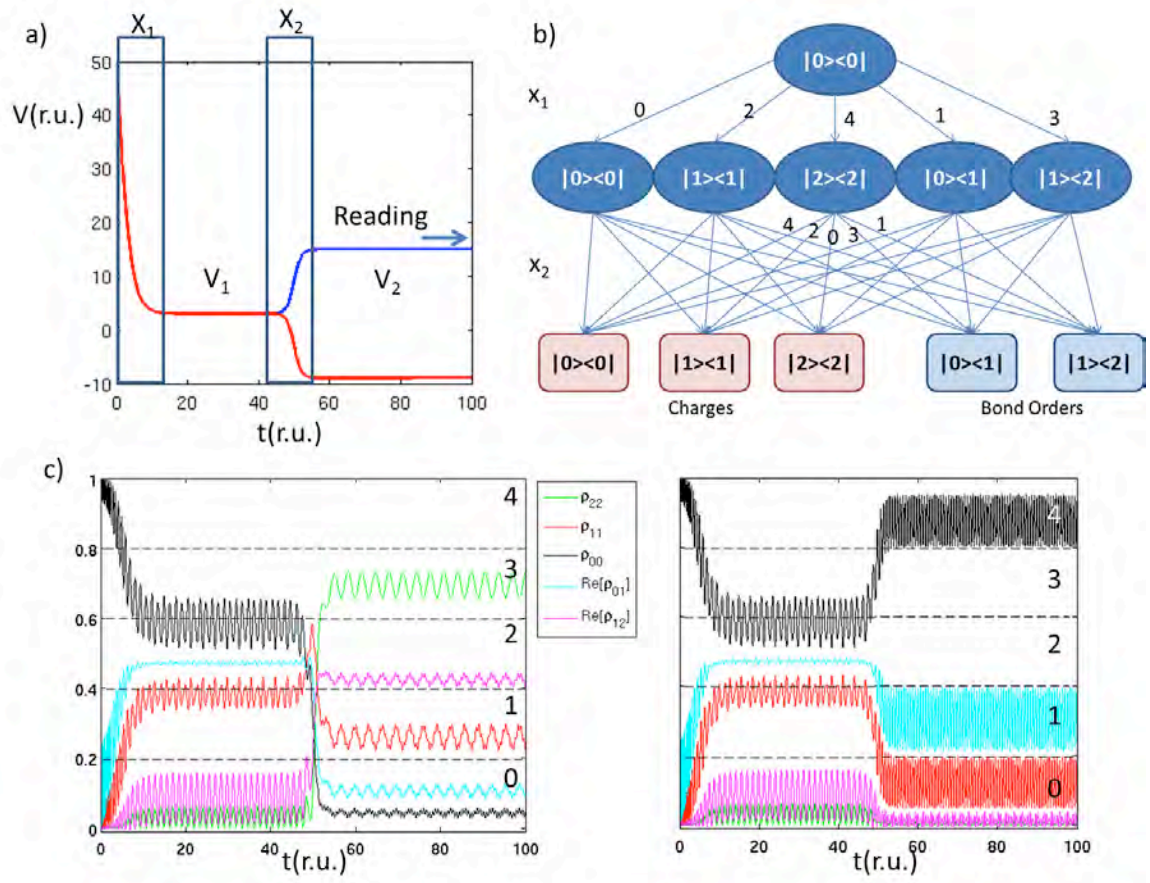

Figure 3: Voltage time profiles (a) applied to the side gate in order to implement two functions of two multi-valued logic variables in radix 5. The decision tree representing the functions is depicted in panel b. The nodes of the tree are the possible states (observables) of the system connected by the voltage switch. Panel c shows the time evolution of the observables under the application of the red (left) and blue (right) voltage profile, and indicates the 5-valued reading of the observables for the implemented function. The reduced unit of energy is defined by β_{12}, the smallest coupling constant (coupling constants are kept constant in the model), reduced units of time are given by h/β_{12}.

3. The third contribution to parallel computing with observables in PR2 is by optical addressing by a sequence of short laser pulses, as in the 2D PE set used in WP2. In the design that we propose, all the observables corresponding to the phase matching conditions of the ensemble are used, instead of only the 2D PE rephrasing and non rephrasing direction as is presently implemented by WP2. The phase matching conditions provide a complete basis set for decomposing the full polarization of the system, $\mathbf{P}(t)=\sum_l P_l(t)\exp(i\mathbf{k}_l\mathbf{r})$ with the phase matching conditions given by the set of integers $\mathbf{l}\equiv(l_1,..,l_i,...)$ with $\mathbf{k}_l=\sum_n l_n\mathbf{k}_n$. The full polarization of the system, $P(t)$, can be tuned by choosing the molecular system and for a given molecular system, by changing the pulse sequence which addresses it. We propose to use this approach to implement the decomposition of a function, the full polarization, on a discrete basis set, the set of integers that correspond to the phase matching conditions. The

computation of the transform is performed is parallel, since all the coefficients, the partial polarizations, $P_l(t)$, can be measured simultaneously. Another very appealing aspect of this logic scheme is that what is measured is directly a macroscopic signal, which is the response of the ensemble. In the logic scheme, the response of the ensemble of molecules does the interfacing with the macroscopic world. For random positions of the solute molecules in solution, the partial polarizations, $P_l(t)$, correspond to the rotationally averaged polarization of a single molecule of solute. Examples of the tunability of the transform are shown in Figure 4. They are computed by simulating the ensemble response of a given three level model system by changing the two time intervals between the three pulses, τ, the coherent time, defined as the time interval between the first two pulses and *T*, the relaxation time, time interval between pulse 2 and 3.

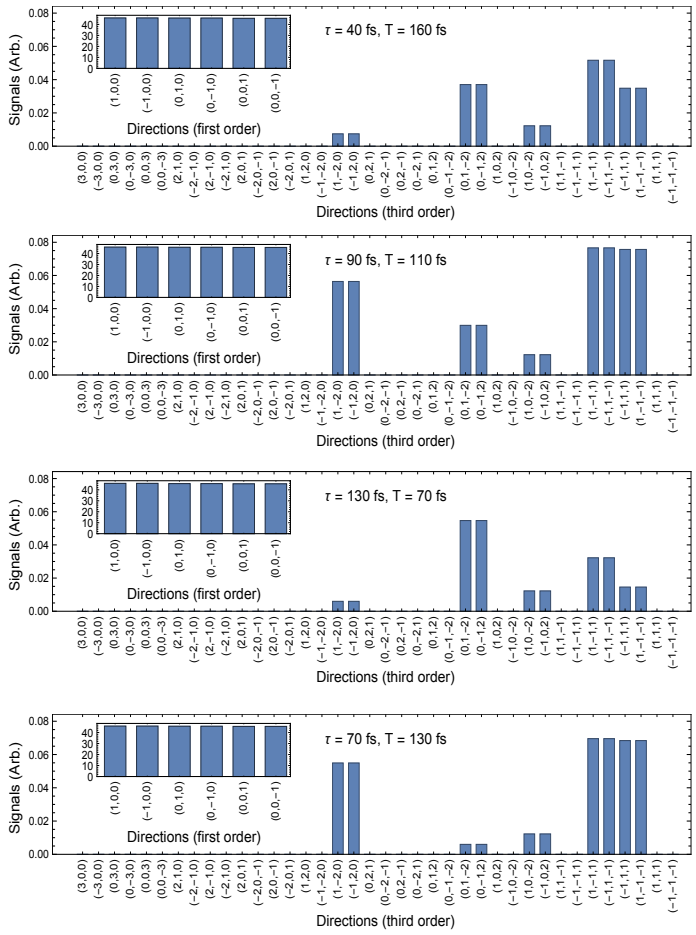

Figure 4. Coefficients $P_l(t)$ of the transform computed in all the phase matching directions *l* corresponding to first order (inset) and third order for various values of the time intervals between the pulses of the sequence τ and *T* as indicated. The model is a 3 level system in a V shape, comprising a ground state, |0> and two excited states, |1> at 15800 and |2> at 16200 cm^{-1} above the GS. The interaction with the electric field couples |0> to |1> and |2>. The three pulses of the sequence are identical, polarized along z in the laboratory frame and propagating in the (x,y) plane. The envelopes are Gaussian, with a width of 15 fs. The values reported correspond to the integrated values of the coefficients $P_l(t)$. A lot of coefficients are strictly zero because only 3 states are included in the model and the field strength is weak.