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Experimental coherent computation of a multiple-input AND gate using pure molecular superpositions

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Abstract

Pure coherent superpositions of Li_2 rovibrational eigenstates are used for the *experimental* coherent computation of a classical multiple-input AND logical gate. The input sets of Boolean values are imprinted into the phases of the eigenstates by shaped femtosecond pulses. The computation is carried out by the coherence and field-free time evolution. The Boolean result is given by the existence/nonexistence of a specific pre-determined superposition at a given delay time; it is read via projection onto an ionic state by a second femtosecond pulse. As an example, the gate is used in solving a yes/no computational decision problem. © 2002 Published by Elsevier Science B.V.

1. Introduction

The design and active control of the dynamics of photo-excited coherent superpositions of quantum states (wave packets) is now possible by tailoring broadband ultrashort laser pulses. The control knobs are the phase, amplitude, and polarization of the various spectral frequencies of the radiation field [1,2]. These principles have been exploited in the field of coherent control of atomic and molecular systems [3–8], with the objective of optimizing the outcome of chemical and physical processes. Recently these same principles have been exploited experimentally for storage and retrieval of information through the quantum phase in Rydberg atoms that can be used for a database search based on the Grover's quantum algorithm [9]. In this direction of information processing, in parallel to the present work, we have demonstrated the experimental implementation of the Deutsch–Jozsa quantum algorithm for two-qubit [10] and three-qubit [11] functions using a rovibrational molecular wave packet representation. The present study employs pure coherent superpositions of molecular Li_2 rovibrational eigenstates for the experimental coherent computation of a multiple-input AND logical gate. The gate is then used as a yes/no evaluation component in solving a computational decision problem. Both

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these works are part of exploring the computational potential of pure molecular superpositions of small molecules in a gas phase ensemble, which are controlled by shaped broad bandwidth pulses. Our general objective is creating an experimental platform for small-scale problem-specific quantum computation involving pure superpositions of hundreds of eigenstates. Below, in this section, we will elaborate on this objective framework.

Quantum computation [12,13] is aimed at utilizing the quantum nature of physical systems in order to solve computational problems in efficient ways, which are impossible in classical computation. Although quantum computation has been under extensive investigation in the last several years, the number of quantum algorithms that have been discovered to be significantly more efficient than their classical counterpart is rather limited. The most prominent quantum algorithm known today is Shor's algorithm for factorization of numbers [12–15] that is based upon the quantum Fourier transform. It provides exponential speedup over the best known classical factorization algorithm. The common formalism of quantum computation [12–18] is in terms of qubits, each of which is a two-state quantum system. The Hilbert space of N qubits is composed of 2^N states. The quantum computational task is carried out through a sequence of unitary transformations acting on superpositions of states within this Hilbert space. Every N-qubits unitary transformation is approximated to any required accuracy using a sequence of elementary gates, each acting on one or two qubits. Such a sequence is referred to as a quantum circuit. The gates comprising the circuit are all members within a universal finite set of basic gates. This model provides a powerful universal programming language for quantum computation. However, it acquires a significant drawback, since most unitary transformations can only be implemented inefficiently, i.e., they require a circuit of elementary universal gates whose size is exponential in N. Only special transformations, like the quantum Fourier transform, can be decomposed into polynomial-size networks. Thus, in parallel to efforts along the above universal quantum computation model, it seems highly desired to explore other frameworks for quantum computation, even at a price of losing the universality. This seems especially beneficial when considering the experimental implementation of a quantum computation device, beyond the study of the theoretical concept of quantum computation.

One such experimental framework is the one we are aiming at. It is based on an ensemble of small molecules in the gas phase interacting with a sequence of multiple shaped femtosecond laser pulses. The computational task is carried out through the time-dependent dynamics of the molecule in pure coherent superpositions, with short computation time and very low decoherence rate. Each pulse induces a desired molecular unitary transformation. The superpositions (wave packets) are composed of a manifold of molecular eigenstates, each acquiring independent amplitude and phase values. The molecular eigenstates span several (entangled) internal degrees of freedom – rotational, vibrational, and electronic. Typical decoherence times for rovibrational wave packets of small molecules in the gas phase at low pressure are several nanoseconds, compared to the time scale of a single coherent manipulation with an ultra-short laser pulse, which is in the femtosecond range. This framework does not provide a basis for universal quantum computation, but is expected to be suitable for small-scale computational tasks, involving up to hundreds of eigenstates spanning several internal degrees of freedom (as an illustration, the Hilbert space of 8–10 qubits is composed of an equivalent number of quantum states). Still, at this price, it might provide the possibility of efficiently implementing quantum algorithms and unitary transformations that under the above universal quantum computation model would be considered inefficient. The potential lies in the richness and complexity of the interaction between the molecule and the optical field of the broad bandwidth pulse, involving simultaneous single- and multi-photon, direct and Raman, resonant and nonresonant transitions among the whole manifold of quantum states within the superposition. The possibility of using additional molecular eigenstates, external to the specific computational Hilbert space, as intermediate states in the transitions adds significantly to this richness. At this stage we view our objective framework mainly as a

problem-specific small-scale computation device. By problem-specific we mean that the pulses inducing the molecular unitary transformations of the quantum algorithm will be specifically designed and shaped to implement the transformation in complete, without decomposition into a quantum circuit of elementary gates. It is expected that the useful theoretical computational methods for designing such pulses will include also optimal control theory [3,4]. Good candidates for quantum algorithms, which provide significant advantage over their classical counterpart even for a relatively small number of involved elements, are quantum simulation of quantum systems [19–21].

2. Coherent computation of a multiple-input AND gate

2.1. Principle of computation

An important property of a nonstationary time-dependent wave function is the underlying coherent parallelism in the time evolution of the various individual eigenstates. For $|\Psi(t)\rangle$, composed of a coherent superposition of eigenstates $|n\rangle$, $|\Psi(t)\rangle \propto \sum_n a_n e^{-iE_n t/\hbar}|n\rangle$ with $a_n = |a_n|e^{-i\Theta_n}$ as complex coefficients, and each state $|n\rangle$ evolves according to its eigenenergy E_n. The quantum dynamics of the involved physical system enables implementation of a computational logical operation between a set of input values. Each of these values is written into the phase Θ_n and/or amplitude $|a_n|$ of a corresponding eigenstate. The present implementation of a coherent multiple-input AND logical gate using coherent superpositions of molecular rovibrational eigenstates is based on this above principle.

(i) *Input.* The input to the AND gate, which is composed of a set of independent Boolean values, is imprinted into the relative phases Θ_n of the various eigenstates composing the superposition. The molecule is transformed into this coherent pure superposition as a result of its interaction with a phase-shaped broadband femtosecond laser pulse; different pulses apply different unitary transformations representing different input sets.

(ii) *Computation*. The machinery for implementing the computational operation of the AND logical relation between the input values is the coherent field-free time evolution of the molecule (after the imprinting pulse is over).

(iii) *Output*. The output Boolean result of the gate is given by the existence or nonexistence of a specific pre-determined coherent molecular superposition, $|\Psi^*(t = t^*)\rangle$, at a pre-selected time t^* from preparation.

(iv) *Readout*. The output is read out through the projection of the overall superposition at the pre-selected time onto an ionic final state. This is carried out by photoionizing the molecule, using a second femtosecond pulse, and measuring the resulting ionization signal.

2.2. Experimental implementation

In the experiment, the molecular rovibrational superposition state, $|\Psi(t)\rangle$, is prepared following the interaction of a molecule in a pure initial state i (with energy E_i) with a weak broadband ultrashort laser pulse. It is given at time t after preparation (when the pulse is over) as [7,8,22] $|\Psi(t)\rangle \propto \sum_n \mu_n \epsilon_n e^{-i\Phi_n} e^{-iE_n t/\hbar}|n\rangle$. The index n stands for a rovibrational eigenstate (v_n, J_n) in the superposition, $|n\rangle$ represents its eigenfunction, E_n represents its energy relative to state i, and the coeffcient μ_n is the dipole moment matrix element of the $i \to n$ transition. The quantities ϵ_n and Φ_n are the electric field amplitude and phase, respectively, of the preparing pulse at the transition frequency $\omega_{ni} = E_n - E_i$; these are the experimental pulse shaping parameters for tailoring the superposition. Here, only phase-control by laser electric field is employed, leaving the amplitude-control constant.

The AND gate implemented here has six input Boolean values, each is encoded into the phase Φ_n of a single rovibrational state $n \equiv (v_n, J_n)$ in a (six-state) superposition prepared on the E electronic state of Li_2 (see Fig. 1). An input value of 0 is encoded as $\Phi_n = 0$, while an input value of 1 is encoded as $\Phi_n = \Phi_n^*$, i.e., as a specific phase value (different than 0) depending on the state. The amplitude that each state acquires is invariant, i.e., independent of the input values. The Boolean output value of an AND gate is 1 only when all

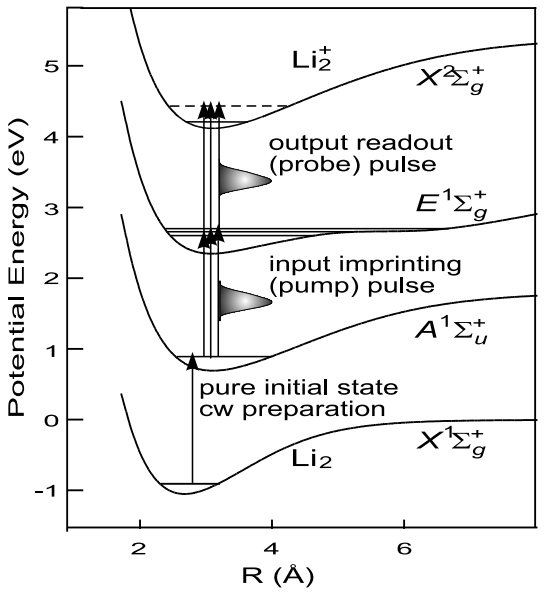

Fig. 1. The experimental excitation scheme, with the relevant potential energy curves of Li_2 and Li_2^+ [23,24], used for implementing the coherent multiple-input AND logical gate.

input values are 1, while in all other cases the output value is 0. Thus, the coherent molecular superposition at t^*, $|\Psi^*(t^*)\rangle$, that its existence or nonexistence provides, respectively, the 1 or 0 Boolean result of the AND gate (see above) is the one prepared with the specific set of phase values $\Omega^* = \{\Phi_n^*\}$. Explicity, it is $|\Psi^*(t^*)\rangle \propto \sum_n \mu_n \epsilon_n e^{-i\Phi_n^*} e^{-iE_n t^*/\hbar}|n\rangle$. The set of phases Ω^* used here is $\Omega^* = \{\Phi^*_{v_n,J_n}\} = \{\Phi^*_{13,17} = 143.5°,\ \Phi^*_{13.19} = 147.0°,\ \Phi^*_{14,17} = 244.4°,\ \Phi^*_{14,19} = 207.2°,\ \Phi^*_{15,17} = 108.0°,\ \Phi^*_{15,19} = 270.8°\}$, all of which differ significantly from zero. Only the relative phase values are important, not the absolute ones. These values are specific to the excited Li_2 states. They were chosen, based on previous detailed coherent control studies of Li_2 [7,8,22], such that the ionization of the corresponding Li_2 superposition (i.e., with Ω^*) at 7 ps delay time after its excitation (i.e., $t^* = 7$ ps) will result in a *global maximum* of the coherent ionization signal as measured here. The 7 ps time is chosen arbitrarily. Any superposition with one or more phases set to zero will have a significantly lower ionization amplitude at 7 ps than the global maximum. Consequently, following the readout step at 7 ps, a gate output value of 1 will result in a measured ionization signal amplitude that is equal to the global maximum, while an output value of 0 will produce a lower amplitude. This means that observing the maximum signal immediately identifies the output value of the AND gate to be 1. This identification is constrained by the signal-to-noise of the measurement (see below).

3. Experimental technique

The relevant potential energy curves of Li_2 and Li_2^+ [23,24] are shown in Fig. 1. The experiment [7,8] is conducted in a cell (1023 K) containing lithium sample. A single mode cw dye laser selects a specific state-to-state transition of the thermally populated Li_2 from $X^1\Sigma_g^+(v_X = 2, J_X = 17)$ to $A^1\Sigma_u^+(v_A = 14, J_A = 18)$, thus preparing the latter as a pure initial state. The given set of input Boolean indications to the AND gate is encoded, as described above, into a phase-shaped femtosecond laser pulse. The interaction of this shaped pulse with the *ensemble* of Li_2 molecules, which have been excited to the selected initial level on the A-state, transforms them into a corresponding pure coherent superposition (i.e., input-dependent shaped wave packet) on the electronically excited $E^1\Sigma_g^+$ shelf state (pump step). It is composed predominantly of six rovibrational states ($v_E = 13$–$15, J_E = 17, 19$). The existence or nonexistence of the pre-determined molecular coherent superposition $|\Psi^*(t^*)\rangle$, presented in Section 2.2, on the E-state at the pre-selected time delay from preparation, $t^* = 7$ ps, provides the 1 or 0 Boolean output to the AND gate. It is probed through time-delayed ionization of the lithium dimer using a second unshaped femtosecond pulse (probe step). The resulting pump–probe photoionization signal amplitude (ion and electron current) is the measured experimental observable; it is composed of a constant level and a part that depends on the delay-time between the pump and probe pulses. The pump and probe pulses originate from a Ti:sapphire laser system with ~160 fs duration, ~12 400 cm^{-1} central wavelength, ~150 cm^{-1} bandwidth, parallel polarization, and energies of ~0.5 and ~1.5 μJ, respectively. The phase-shaping

of the pump pulse is accomplished using an assembly incorporating a liquid crystal spatial light modulator to encode the desired phases on the pump pulse frequencies. The interaction of the lithium dimer with the laser pulses can be described within the weak field limit [22]. The decoherence of the rovibrational superposition prepared on the E-state occurs on a time scale longer than 5 ns. It is mainly due to collisions of the Li_2 molecules with Li and Ar atoms, resulting in pure dephasing and/or state-changing transitions.

3.1. Model computational problem

We demonstrate the experimental coherent computation of the multiple-input AND gate also by its incorporation within a complete model computational process. The gate is used in the last step of evaluating the answer to a computational decision problem for which the output is just 'yes' or 'no'. The input to the problem is translated by an oracle into a set of Boolean values that is used as an input to the AND gate, and the Boolean output of the gate provides the yes/no output of the decision problem. Decision problems are a fundamental class of computational problems [25]. The specific decision problem we consider involves graphs that are composed of vertices and edges between pairs of vertices (not every pair is necessarily connected by an edge) [26]. The computational task is to determine whether a given graph is free of isolated vertices. An isolated vertex is a vertex where no edge connects it to any other vertex (see Fig. 2). The input of our problem is a graph's data given as a list of vertices, $\{i, j, k, \ldots\}$, and a list of edges, $\{(i, j), (i, k), \ldots\}$. Each edge is represented by the pair of vertices it connects. The output of the problem is either 'yes' or 'no', depending on whether the input graph is identified to be without or with isolated vertices, respectively.

The algorithm employed by the oracle to translate the data of a graph into a set of Boolean values (0/1) is as follows: each vertex of the graph is represented by a corresponding Boolean indication; all the indications are initialized with a value of 0; the graph data is read, edge after edge; for each edge, the Boolean indications of both its vertices are updated with a value of 1. This algorithm is such that a specific set of Boolean values corresponds to a family of graph configurations rather than to a specific graph. To make the yes/no decision does not require such uniqueness. Overall, a graph without any isolated vertices results in the existence of the $|\Psi^*(t^* = 7\text{ ps})\rangle$ superposition with the Ω^* set of phases defined above. A graph with isolated vertices results in a different superposition where some phases Φ_n are zero. The AND gate operation distinguishes between the graphs containing isolated vertices and the ones without.

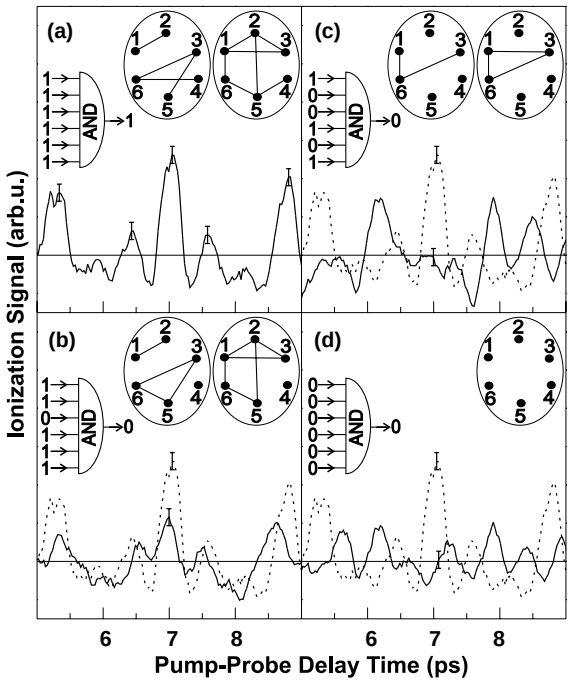

Fig. 2. Pump–probe experimental signals (solid lines) obtained from the time-delayed ionization of several phase-shaped (six-state) rovibrational wave packets of Li_2. Only the time-dependent part of the signal is shown; the thin solid lines indicate the constant signal level (see text). Each wave packet results from a different input to the AND gate shown in the figure, or, equivalently, from a different family of (six-vertex) graphs (see text). Graph examples are given in the ellipses: panel (a) shows graphs with no isolated vertices; panels (b)–(d) show graphs with isolated vertices. The transient of panel (a) is also shown (for comparison) in dashed lines in the other panels. The signal amplitude at 7 ps provides the correct output to the AND gate and whether the encoded graph has no isolated vertices [1/'yes', for panel (a); 0/'no', for panels (b)–(d)].

4. Results

Fig. 2 shows various pump–probe experimental transients obtained from the ionization of several different shaped wave packets at various time delays; only the time-dependent part of the signal is shown (the constant signal level is indicated). Each transient results from a different input to the AND gate or, equivalently, a different family of graph configurations. Some graph examples are shown next to each transient. Fig. 2a corresponds to the case where all the input values to the gate are 1, and, thus, it shows graphs without isolated vertices. Figs. 2b–d correspond to cases where at least one gate input value is 0, and, thus, show graphs with isolated vertices; in Fig. 2d all the input values are 0 and all the graph vertices are isolated. The transient for graphs without isolated vertices, is also shown for comparison (dashed lines) in Figs. 2b–d. As can be seen, the ionization signal transient distinguishes the various cases one from another, and the signal amplitude at 7 ps immediately identifies the AND gate output and whether the encoded graph is without isolated vertices [1/'yes', for Fig. 2a; 0/'no', for Fig. 2b–d]. Obviously, a single initial calibration of the amplitude that corresponds to the maximum signal level is required once and for all. Based on the experimental signals and the corresponding error bars, the probability of obtaining the correct answer is greater than 99%. The results demonstrate the reliable experimental operation of the coherent molecular AND gate in the time domain.

5. Discussion and conclusions

The success probability for the correct answer based on the output signal is limited by the experimental signal-to-noise; for greater numbers of rovibrational states within the superposition, i.e., larger number of input Boolean values or vertices, higher signal-to-noise is required to achieve the same performance. For 95% success probability with N states, considering all ionization pathways of the Li_2 (i.e., pump–probe pathways as well as direct two-photon ionization by the pump and probe pulses individually), the required signal-to-noise ratio (S/σ) is $\approx 10N$. Since $S/\sigma = \sqrt{n_{\text{ions}}}$, where n_{ions} is the number of detected Li_2^+ ions, for 95% success at least $100N^2$ ions are required; for $N = 1000 : n_{\text{ions}} \approx 10^8$. With the present experimental conditions [7,8,22], about 10^4 pump–probe sequences (at t^*) are required to ionize 10^8 Li_2 molecules. However, the current usage of the available resources, i.e., $\sim 10^{12}$–10^{13} photons per pump or probe pulse and $\sim 10^{10}$ molecules in the initial state (v_A, J_A), is highly inefficient and can be improved by longer path and better detection. An improvement factor of $\sim 10^4$ seems feasible and would provide the required signal-to-noise ratio to handle hundreds of elements in the molecular superposition. In this context it is worth mentioning that laser pulses of ~ 1700 cm^{-1} bandwidth ($\sim$10–15 fs transform-limited duration) will allow the direct simultaneous coherent excitation, from a single selected Li_2 rovibrational state of the A-state, of about 50–60 rovibrational states centered spectroscopically around the current region of the E-state.

Obviously, there are other coherent and incoherent ways to implement an equivalent classical AND gate. It is done on any classical computer based on Boolean logic that is described by the Turing machine model [12,13]. It could also be implemented with purely optical techniques, for example: the phases will be encoded into a phase-shaped pulse; the set of phases, which corresponds to the case when all the input Boolean values are 1 and thus the gate output is 1, will result in a transform limited pulse, while any other input will result in a chirped pulse; the existence or nonexistence of a transform limited pulse will be determined based on whether or not the generation of second-harmonic light is maximal. The importance of the present work is in the ability to implement the AND gate in a coherent way, i.e., involving coherent information and coherent dynamics, with molecular quantum elements. It demonstrates experimentally one element out of many needed for the full implementation of computation with molecular superpositions controlled with sequences of multiple ultrashort pulses. Such a platform will not serve as a universal quantum computation device, but rather as a problem-specific computation device. In such a device the involved pulses will be

shaped to correspond to specific unitary transformations tailored for the specific problem under computation. The platform might also serve as a model system for experimental studies of quantum error correction [12].

To conclude, this work utilizes a detailed knowledge of the expected time-dependent molecular dynamics for the experimental coherent computation of a multiple-input AND gate using pure coherent molecular superpositions. The input information is stored in the phases of the individual rovibrational eigenstates composing the superposition. Since a weak optical field is used for the readout/probe operation the encoded information is kept primarily undamaged, and, in principle, can be further used within several nanoseconds. The coherent AND gate can be incorporated, as demonstrated above, as a yes/no output evaluation component, i.e., it can evaluate the existence or nonexistence of a pre-determined specific coherent superposition at the end of a quantum computational process. It might also be a basis for implementing conditional coherent molecular manipulation. In such a case, the molecular superposition prepared on a particular electronic state would not be projected onto an ionic final state, but rather, equivalently, onto one or more other neutral electronic states. The amplitude transferred to target superpositions on each of these electronic states will depend on the exact coherent superposition existing at the projection time; this time can be different for the different states. Extending the possible input and output values beyond the Boolean case, i.e., with more than two possible values, also seems feasible.

Acknowledgements

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