Logic gates using high Rydberg states

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Contributed by R. D. Levine, January 11, 2001

Connected logic gates can be operated on the levels of one molecule by making use of the special properties of high Rydberg states. Explicit experimental results for the NO molecule are provided as an example. A number of other options, including that of several gates concatenated so as to operate as a full adder, are discussed. Specific properties of high Rydberg states that are used are: their autoionization is delayed so that they can be distinguished from direct multiphoton ionization, during their long life such states also can decay by energy transfer to the molecular core in a way that can be controlled by the judicious application of very weak external electrical fields, and the Rydberg states can be detected by the application of an ionizing electrical field. The combination of two (or three) color photons with and without external weak fields allows the construction of quite elaborate logic circuit diagrams and shows that taking advantage of the different intramolecular dynamics of levels that differ by their excitation enables the compounding of logic operations on one molecular frame.

molecular computing | concatenated circuits | zero electron kinetic energy spectroscopy

The dynamics of high molecular Rydberg states, of principal quantum number n of 100 or more, has received much recent interest (1–6). The essential findings that are relevant to this paper are mentioned briefly in the abstract and are summarized further below. The studies of the dynamics were motivated both by the applications to ZEKE (zero electron kinetic energy) spectroscopy and by the inherent interest in the unusual dynamical regime exhibited by such states (7). We show here how this new understanding and the technological capabilities developed for these experiments allow a novel type of application: the concatenation of several logic gates by using only one molecular frame (8).

Computing with molecules is becoming an active area of research. Single-molecule-based rectifiers (9) and switches (10, 11) have been reported recently and offer a quite different approach (12) to molecule-based computing. The switches can be turned on or off by both redox and chemical inputs (13). The switch allows an electrical current to flow or not by causing a conformational change in [2]catanane-type molecules. The mechanochemical mechanism is that a pair of interlocking rings can be switched between two conformations in an oxidation/reduction cycle. The molecule was incorporated in a solid-state device and could be recycled many times (11). Chemical inputs and optical outputs also have been discussed (14, 15) for molecules in solution. To be clear, we emphasize that in this paper the logical response is due to one molecule but that the observed signal is due to a finite but not enormous number of molecules (as few as 100 per laser shot and even 10 molecules per shot if one is working at the peak of the spectrum). These molecules are acting independently of one another, and, because of the unusually large size of a high Rydberg state, they need to be kept as far apart as possible. This is why working the least possible number of molecules is experimentally advantageous.

This paper discusses the integration of reasonably large logic circuits on one molecule. We believe this is a worthwhile goal in its own right and, furthermore, that it is an essential first stage of developments that would become practical only if means can be devised to communicate the logical output of one molecule as an input to another.

In the background to this work is the suggestion that molecular spectroscopy can be thought of in terms of logic gates and that the intramolecular dynamics are logically equivalent to the concatenation of several gates (8). Here we apply this idea to ZEKE spectroscopy and provide experimental results showing that such integration indeed is possible. Not discussed in this paper is the integration of different molecules into even larger circuits.

Many of the special dynamical properties of high Rydberg states are a result of the very high density of quantum states of the system (6, 16, 17). [There are n^3 states of n per unit (au) of energy. Each value of n corresponds to n^2 quantum states, and n is in the range of 100. Compounding this are the states of the molecular core.] This high density and the ability to take fine spectral slices (see, e.g., refs. 18 and 19) suggest that Rydberg states could be used to perform quantum computations (20–22). In this paper we do not take advantage of quantum parallelism (23) and confine ourselves to classical two-valued logic. What this means is that the logic gates compute the value of a Boolean function (24). This is a function of variables that take on only one of two possible values, say "on" and "off." The variables are connected by the familiar logical operations of Boolean algebra and its axioms. The complete representation of such a function is by a truth table, where the value of the function is given for every possible combination of the values of the variables. An equivalent representation is by means of switches (25). These switches can be grouped into slightly larger units, known as logical gates, and we also will use the representation in terms of such gates. The point of our paper is that more than one such gate, in succession, can be operated on the molecule so that the molecular response is equivalent to quite a number of switches.

High Rydberg states exhibit unusual dynamics. Some familiarity with the recent findings is needed for the development below. The essential background is discussed first. Next, we show the gates that can be realized by using the two-color, two-field experiments on NO (26). Finally, we explore what can be done with experiments by using three colors (27), including a so-called full adder, which typically is realized with five logic gates, two of which are the already nontrivial Exclusive-OR gates (24). The paper concludes by pointing out the many options for further concatenation that are open when one realizes that it is possible to combine all of the already available state-preparation options with the more detailed probing that also is currently practical.

Spectroscopy and Dynamics of High Rydberg States

Rydberg states of molecules often are prepared by a two-color excitation (see Figs. 1 and 2). The first photon brings the molecule to its first excited (S_1) electronic state. The combined energy of the two photons is just below a threshold for direct ionization of the molecule. One of the essential points of ZEKE

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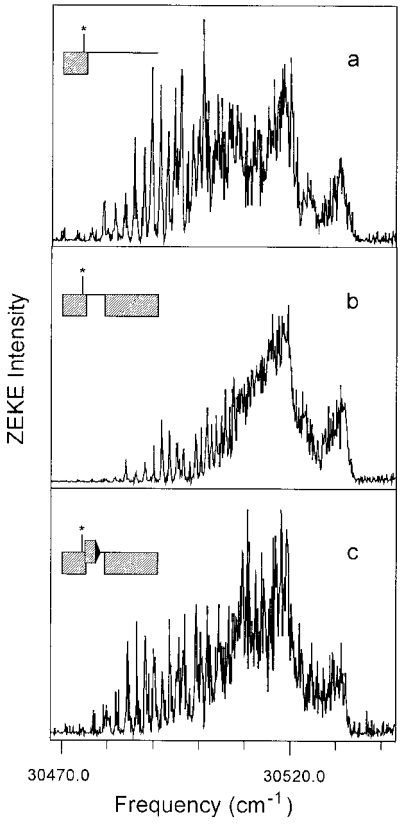
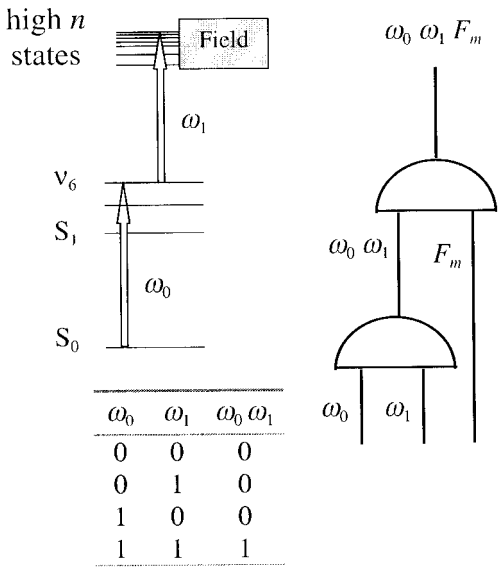


Fig. 1. An experimental ZEKE (pulsed-field ionization) spectrum plotted vs. the frequency ω_1 of the second photon (see Fig. 2) for NO with three different configurations of weak (300 mV/cm) DC fields as shown. (*a*) The field F_ℓ present only for 10 ns during excitation, indicated by an asterisk. (*b*) The field F_ℓ is present during excitation and is switched off for several microseconds and switched on again. (*c*) The field F_ℓ is present during excitation and is switched off and then switched on again. While the field F_ℓ is off, the field F_m is switched on for 10 ns. See ref. 26 for more details about such experiments.



ω_0	ω_1	$\omega_0\,\omega_1$
0	0	0
0	1	0
1	0	0
1	1	1

Fig. 2. The principle of the two-color, three-field experiment and the concatenated logic gates that can be built by using only one of the fields. The actual experimental results are shown in Fig. 1. These show that two or even all three fields can be used for the building of logic circuits. More elaborate schemes will be discussed below. (*Left*) A two-photon absorption leading to a high Rydberg state via an intermediate state, which is the v_6 vibration of the first electronically excited state (S_1). The two photons are of different colors. The Rydberg state is accessed in a region in which a weak DC electrical field, F_ℓ, is present. This field is switched off after 10 ns but can be switched on again, several microseconds later (see Fig. 1 and ref. 26). Next, a second DC field, F_m, which is in a direction perpendicular to the first field, is or is not switched on. The measured spectrum can be used to indicate whether the field F_m has been ''on'' or ''off.'' Thereby, the output of the first AND gate (shown in the usual notation as a half-moon, with its truth table below it) can be fed to a second AND gate, and the output depends on whether the field F_m was or was not on. Note that each AND gate is equivalent to two switches (25) so that even this simple scheme has a single-molecule response equivalent to four switches. As discussed for Fig. 3, using two photons and one field is really a four-gate logic circuit. We use the simpler circuit shown in this figure to introduce the ideas.

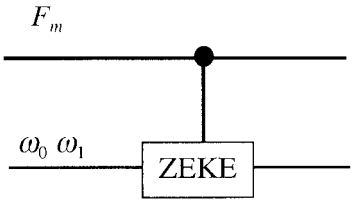
spectroscopy is that this need not be the lowest threshold. If it is not, the second photon can induce direct ionization or it can reach a Rydberg electron that is revolving around an excited ionic core. By waiting for tens of nanoseconds, the prompt electrons depart, pushed away by a weak DC electrical field. The kinetic energy of the direct electrons can be measured, and this is the basis for photo electron spectroscopy. The neutral high Rydberg states that are left can be ionized later by the rapid application of an electrical field.

The high Rydberg states are long-lived because most of the time the electron is away from the positively charged molecular core. The electron initially is promoted into a Bohr–Sommerfeld orbit of low angular momentum, ℓ. Once per its (long) period such an orbit brings the electron close to the core, where it may gain or lose energy. The period of the orbit scales with the principal quantum number n as n^3, so that low ℓ and high but not very high n states do interact with the core. To stabilize the Rydberg electron, it should be made to move into a circular-like orbit of high angular momentum, where it is never near the core. The presence of a static electrical-field F_ℓ breaks the spherical symmetry and allows the angular momentum to increase, which stabilizes the orbit (28–31). However, the angular momentum ℓ increases and then decreases (16) so that the electron is shielded from the core only for part of the time. To fully isolate the electron, one must make the magnetic quantum number m increase because $\ell \geq m$. This is possible (26) by the application of a second electrical field, F_m, which is in an orthogonal direction in space to that of the first field, whose role is to change ℓ. We will emphasize the on/off control made possible by the second field, but one also could use the first field as a Boolean variable.

Experimental Realization of Concatenated Gates

Fig. 1 shows the experimental ZEKE spectrum that forms the basis for the discussion. The plot is vs. the frequency of the second photon. (In our proposed logic circuit, this frequency would be fixed.) Fig. 1 *b* and *c* shows that the spectrum without and with the m changing field, F_m, being off and on, is readily detectable. The point is that when m is higher, the condition $\ell \geq m$ stabilizes Rydberg states of lower n and, therefore, allows their detection, which occurs for a lower frequency of the second photon. The Fig. 1 *a* and *b* shows that the condition that the ℓ changing field is reapplied or not is also experimentally detectable. Either field or both therefore can be used as Boolean variables.

The construction in Fig. 2 begins with the result that a two-photon transition, pumped with two photons of different color, already is an "AND" gate (8). This is because only if both lasers are "on" does one observe a ZEKE signal. The truth table for that part of the experiment is shown. In terms of Boolean variables, the output of the AND gate is w_0w_1, where w_i is 0 or 1 depending on whether the laser is off or on. The output of the AND gate is one input into a second AND gate. The other input to the second gate is the Boolean variable F_m that indicates whether the m changing



ω_0 ω_1	F_m	ZEKE
0	0	0
0	1	0
1	0	b
1	1	c

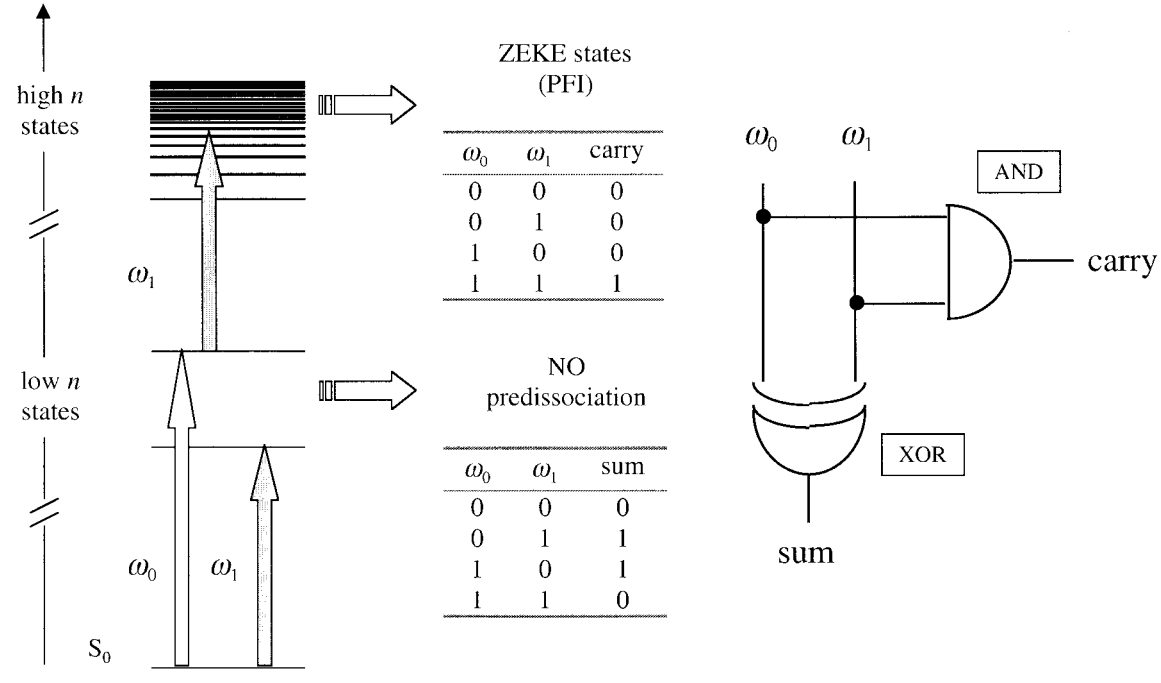
Fig. 3. A representation of the role of the control electrical field F_m as a control gate. The output, $\omega_0\omega_1$, of the first AND gate of Fig. 2 is controlled by the field. The gate is shown as a truth table, where the entries for the output refer to the panels in Fig. 1.

field is off or on. The complete gate evaluates $\omega_0\omega_1F_m$, as seen also from the truth table included in Fig. 3.

The higher frequency ω_0 could lead to a two-photon direct ionization. Because the absorption cross-section to S_1 is typically high, this is minimized by using an intensity that is low enough. In any case, direct ions are discriminated against by the weak DC field that is already present at the ionization stage as shown in Fig. 1. The lower frequency needs to be low enough (say, 30 cm^{-1} below the ionization potential) that one is in the spectral regime in which one can discern whether the control field F_m is on or off. In the same spectral range, one could also distinguish whether an ℓ changing field, F_ℓ, is or is not imposed for a second time (see Fig. 1). Therefore, one even has a three-stage gate that evaluates $w_0w_1F_mF_\ell$. Even $w_0w_1F_\ell F_mF_\ell$ is possible, where the order of terms indicates the sequence of physical operations.

One can also use the experimental arrangements to define a controlled gate, as shown in Fig. 3. The role of the control field, F_m, is to determine which one of the two possible ZEKE spectra is obtained as the output.

The gates based on delayed pulsed-field ionization are not fast (see Fig. 1 legend for the time scales, which are tens of nanoseconds). We have used them in illustrating the logical circuits because of ease of presentation. One can realize far faster gates [e.g., about 1 ps (32) or even faster (8)]. The Rydberg-based gates are also one time and irreversible because the molecule is destroyed. These problems can be avoided by making use of the already demonstrated experimental ability to slice the ZEKE spectrum by the application of a stepwise ionizing field (18, 19) or by proceeding via different vibronic states of the electronically excited intermediate state [$B^1\Pi_u$ in the case of Ag_2 (33); $A^2\Sigma^+$ for NO]. Other options include applying crossed electric and magnetic fields (34) so as to break the ℓ and m quantum numbers or to delay the second frequency with respect to the first. Time-resolved ZEKE spectroscopy (35) revealed that there is more than one time scale in the dynamics of high Rydberg states. This separation of time scales (6, 36), which is due to several bottlenecks in the time evolution of the optically accessed state (37), allows one further to add columns to the truth tables. A particular but important special case is the chemical transformations that are possible in the ionic core, about which the Rydberg electron revolves. These changes, such as dissociation, can be spontaneous (38) or light-induced (39). It is a testimonial to the remarkable stability of the Rydberg electron that one of the two fragments of the dissociation is, itself, a high Rydberg state. It too can be probed by ZEKE spectroscopy, and mass resolution (40) can distinguish it from the parent. Another option that we will discuss below is the



ω_0	ω_1	carry
0	0	0
0	1	0
1	0	0
1	1	1

ω_0	ω_1	sum
0	0	0
0	1	1
1	0	1
1	1	0

Fig. 4. The implementation of a half-adder. Note that by monitoring the intermediate excited state (S_1) one is able to get an Exclusive OR (denoted as XOR) gate. This generates the ''sum,'' which is a sum of two Boolean variables, modulo 2. In Boolean notation, the ''sum'' of A and B equals $A\bar{B} + \bar{A}B$, where the bar denotes the logical negation. The forward carry bit is generated only when both laser colors are on by a ZEKE signal, which is obtained via pulsed-field ionization. Note that an XOR gate, shown with an additional semicircle, is a universal gate. The concatenation here is achieved without external fields by using only the intramolecular dynamics of S_1. By adding the role of the field, one can do more, as shown in Fig. 5. A full adder is shown in Figs. 6 and 7.

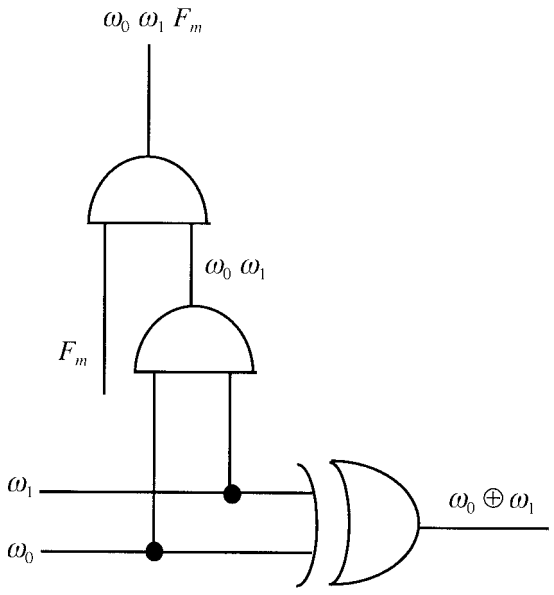


Fig. 5. A two-photon, one-field logical scheme. (It is a half-adder crossed with a field. The symbol $\oplus$ means addition of two Boolean variables, modulo 2. It is the Boolean variable called "sum" in Fig. 4.) This scheme uses two different aspects of molecular dynamics. As in the half-adder, Fig. 4, it takes advantage of the dynamics in the intermediate electronic state, S_1. As in the scheme of Fig. 2, it takes advantage of the more rapid quenching by the molecular core of high but not very high Rydberg states, unless these are protected by having a high m value, which is made possible by the application of the field F_m. Once m is not low, the condition $\ell \geq m$ on the angular momentum of the electron keeps it away from the core and stabilizes the state so that it gives rise to a ZEKE signal. In reality, one could (see Fig. 1) include the role of two fields.

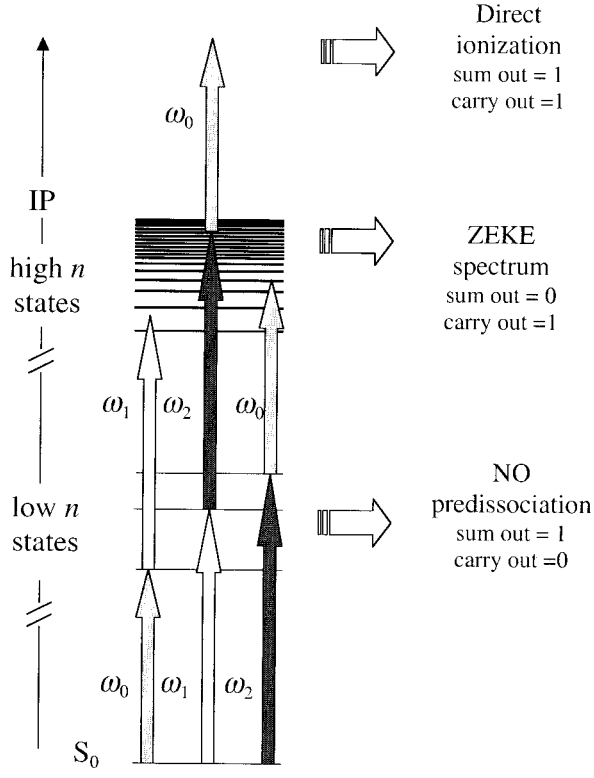
predissociation of the neutral excited state that is used as an intermediate in reaching the Rydberg state (31). An option that we do not use here is to induce transitions among the high Rydberg levels by millimeter radiation (41). This is a very promising direction because it makes a large number of states addressable by controlled fields. Instead of outlining all of the many options, we discuss next how to design a full adder.

The Full Adder

A full adder carries out a process somewhat more complicated than addition modulo 2. It must also add any carry digit brought from the previous adder and deliver any required carry digit to the next adder. Addition modulo 2, denoted by $\oplus$, is implemented by a so-called "half-adder." As shown in Fig. 4, a half-adder does not require a carry input bit. To implement a half-adder one needs to monitor S_n, n low (42). This provides the sum, as S_n lights up if either photon is present. Detecting the signal S_n functions as an XOR gate, because absence of the signal indicates either absence of both photons or the presence of both. The presence of one photon but not the other produces signal S_n. The forward carry bit is generated when both photons are present, which allows the molecule to reach a high Rydberg state, and this provides the needed signature.

The half-adder uses only two photons (here, of different colors) and the molecule. In Fig. 5, we increase the concatenation by adding the role of one field. This is a two-photon, one-field scheme.

A full adder is a half-adder supplemented by the ability to allow for an input carry bit. This input bit, if present, of course, contributes to the sum. Unlike the half-adder, the logical evaluation of the sum is now more complicated because there are three ways for the sum to be unity. Fig. 6 indicates how this can



ω_0	ω_1	ω_2	sum out	carry out	
0	0	0	0	0	no signal
1	0	0	1	0	pred
0	1	0	1	0	pred
0	0	1	1	0	pred
1	1	0	0	1	ZEKE
0	1	1	0	1	ZEKE
1	0	1	0	1	ZEKE
1	1	1	1	1	direct ion

Fig. 6. The implementation of a full adder, based on the half-adder of Fig. 4. The scheme calls for the proven experimental ability to fully discriminate between direct multiphoton ionization and delayed ZEKE spectra. The experiment uses photons of three different colors, as, e.g., in ref. 27. The corresponding logic circuit is shown in Fig. 7. The scheme requires the monitoring of the predissociation of the intermediate excited electronic state. This Boolean variable is denoted as "pred" in the figure and truth table.

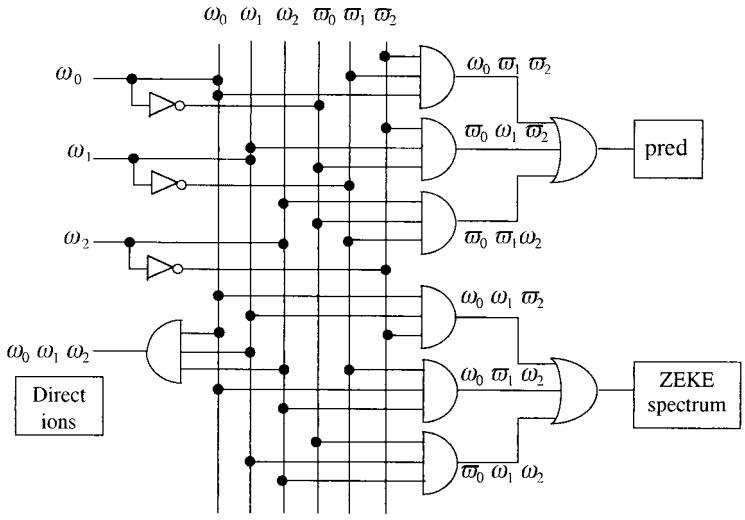


Fig. 7. The logic circuit corresponding to the full adder of Fig. 6. Three NOT gates, shown as triangles, are used to generate the logical complement (indicated by a bar) of the three Boolean variables (e.g., if the second laser is on, $\omega_2 = 1$ whereas $\bar{\omega}_2 = 0$). Two OR gates, shown as a quarter-moon, are used to collect output. The vertical and horizontal lines are drawn to show which of the six variables (three ω variables and their complements) are the actual inputs to any particular gate.

be realized by using three photons. Experimental ZEKE spectra with three photons have been reported by Held *et al.* (27). The third frequency needed for the scheme proposed in Fig. 6 would have to be somewhat lower than that used in the Held experiment, but there is no reason the method should not work. The logic circuit corresponding to Fig. 6 is shown in Fig. 7.

The circuit diagram in Fig. 7 is quite concatenated. This is seen from the vertical lines that are drawn to show what the inputs are to each gate. In fact, the circuit is even more concatenated than shown because we used seven AND gates, each with three inputs. Such a gate is really two concatenated AND gates of two inputs each. (A circuit showing how to concatenate two such AND gates to get a three-input AND gate is shown in Fig. 2.) There are also two OR gates with three inputs each. So the circuit contains 3 NOT gates, shown as triangles, 4 OR gates, and 14 AND gates. Of course, we do not propose to actually build such a circuit, just to indicate that the three-photon spectroscopy of a single molecule can be thought of as logically equivalent to such a circuit.

To summarize, just two photons suffice to implement an AND gate. The logic for this gate is done by the photons. The molecule is used for its resonance condition. Two photons plus a monitoring of the intermediate electronically excited state suffice for a connected circuit known as a half-adder (Fig. 4). For this scheme the key is the intramolecular dynamics. Two photons and a field is the experiment shown in Fig. 1 and discussed in Figs. 2 and 3. Here, too, the dynamics of the molecule play a key role. In the experiment of Fig. 1, the intermediate state is not monitored. If it were, the logical circuit is that shown in Fig. 5. With three photons (27), one already can do a full adder (Figs. 6 and 7). A circuit using as inputs three photons and one field is already beyond our graphical ability to display logic circuits on one page, so this is a good place to stop.[††] Before closing the discussion, we note that three photons and two fields (even three photons and three fields) is still within current experimental capabilities of what can be implemented by using the levels of a single molecule.

Fig. 7 shows a well connected circuit, so a clear comment on what we have *not* shown is also in order. We have demonstrated concatenation in the sense of logical equivalence between the dynamics of one molecule and a connected logical circuit. We have not shown how logical circuits of different molecules can be connected. Real integration ultimately will require that the logical output of one molecule is the input to another.

Concluding Remarks

The experiment on NO that is discussed and the many other options that are allowed by the existing technology demonstrate that several logic gates can be concatenated by using but one molecular frame. In other words, even relatively simple Hamiltonians can have a dynamical behavior that is rich enough for implementing nontrivial logic operations. These gates were shown to be possible even with the crudest experimental probes that are currently available. Refined detection methods allow the evaluation of even more complex Boolean functions. Some of the logical operations are implemented by the interaction of light with matter, and, so, it can be said that they are as much a result of the photons as they are of the molecule. Other operations require both the molecule and the external field. Other logical operations, such as the quenching of a ZEKE spectrum of Rydberg states of high but not very high values of n (see Fig. 1*b*) or the predissociation of low electronically excited states, are strictly a result of the nature of the dynamics of the isolated molecule itself.

The work of F.R. is supported by the Actions de Recherche Concertée (Direction de la Recherche Scientifique–Communauté Française de Belgique). This work was carried out under the auspices of the James Franck Program for Laser–Matter Interaction and by SFB 377. We thank Profs. E. E. B. Campbell and J. L. Kinsey for their extensive and useful comments on the manuscript.

[††]The reason the possible structures are so varied is that there are 2^{2^n} different Boolean functions of n variables.

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