

Conditional dynamics induced by new configurations for Rydberg dipole-dipole interactions

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We suggest a way to use strong Rydberg dipole-dipole interactions in order to induce nontrivial conditional dynamics in individual-atom systems and mesoscopic ensembles. Contrary to previous works, we suggest exciting atoms into different Rydberg states, which results in a potentially richer dynamical behavior. Specifically, we investigate systems of individual hydrogenlike atoms or mesoscopic ensembles excited into high-lying hydrogenlike s , p , or d states, and show how to perform three-qubit conditional dynamics on the information they contain through a proper use of dipole-dipole-interaction-induced energy shifts.

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I. INTRODUCTION

Due to their large dipole moments [1], Rydberg atoms experience strong long-range dipole-dipole interactions. These interactions strongly mix and shift the multiply Rydberg excited collective states of an atomic sample. This phenomenon has recently been put forward as the key ingredient of different promising atomic quantum-processing scenarios. For instance, Rydberg-Rydberg interactions can be used to perform two-qubit logic operations in individual-atom systems by shifting a transition off resonance in an atom, depending on the internal state of another atom in its immediate neighborhood [2–4]. In a mesoscopic ensemble, dipole-dipole interactions are able to inhibit transitions into collective states that contain more than one Rydberg excitation, thus leading to the so-called Rydberg blockade. First predicted in [5], this phenomenon was locally observed in laser-cooled atomic systems [6–11] and could in principle be used in the future to manipulate and entangle collective excitation states of mesoscopic ensembles of cold atoms [5].

So far, the schemes based on Rydberg-Rydberg interactions have focused on the coupling between atoms excited into the same high-energy state. Typically, in these proposals, atoms in the sample are (simultaneously or not) excited to the same Rydberg state $|ns\rangle$. When the so-called Förster process $ns+ns \rightarrow np+(n-1)p$ is resonant, the dipole-dipole interaction is enhanced: one can then diagonalize the dipole-dipole interaction operator V_{dd} in the subspace $\{|ns, ns\rangle, |np, (n-1)p\rangle, |(n-1)p, np\rangle\}$, which leads to shifted new eigenstates (see [3] for a detailed discussion). In the present paper, we propose to investigate other settings in which atoms can be excited into several Rydberg states of different l 's. In these configurations, V_{dd} mixes and shifts some of the several-atom states through Förster-like processes, whereas it leaves the others unchanged: figuratively speaking, depending on the Rydberg state they are excited into, atoms will *see* each other or not. This can be used to selectively hinder certain transitions into multiply Rydberg excited states, while allowing for the others. This, in turn, leads to richer dynamical behaviors than considered in pre-

vious theoretical proposals. Through exciting the information-carrying ground states of the atoms into properly chosen different Rydberg states, one can, for example, induce conditional logic dynamics involving more than two qubits.

To be more specific, in the present paper, we shall first focus on the dipole-dipole interactions which take place in a system of three hydrogenlike atoms submitted to different laser beams coupling their ground levels to different Rydberg states $|r_1\rangle$, $|r_2\rangle$, and $|r_3\rangle$, respectively. We shall carefully examine the interaction-induced energy shifts and the resulting blockade of unwanted transitions, and in particular we shall show that the desired performance is satisfactorily met by rubidium atoms. We shall then demonstrate how to use this physical setting in order to perform nontrivial conditional logic operations involving three qubits of information stored either in a three-atom system or in mesoscopic ensembles, thus generalizing the pioneering work by Lukin *et al.* [5].

II. DIPOLE-DIPOLE INTERACTIONS BETWEEN THREE HYDROGENLIKE ATOMS IN DIFFERENT RYDBERG STATES

Let us consider a system of three identical hydrogenlike atoms, denoted 1, 2, and 3. We let $\hat{H}_{0,i}$ be the unperturbed Hamiltonian of atom i and

$$\hat{V}_{i,j} \equiv \frac{1}{4\pi\epsilon_0} \left(\frac{\hat{\mu}_i \cdot \hat{\mu}_j}{R_{ij}^3} - 3 \frac{(\hat{\mu}_i \cdot \vec{R}_{ij})(\hat{\mu}_j \cdot \vec{R}_{ij})}{R_{ij}^5} \right) \quad (1)$$

be the dipole-dipole interaction between atoms i and j , where $\vec{R}_{ij} \equiv R_{ij} \vec{u}_{ij} = R_{ij}(\sin \alpha_{ij} \cos \beta_{ij} \vec{e}_x + \sin \alpha_{ij} \sin \beta_{ij} \vec{e}_y + \cos \alpha_{ij} \vec{e}_z)$ is the vector from nucleus i to nucleus j . The total Hamiltonian of the system then takes the form $\hat{H} = \hat{H}_0 + \hat{V}_{dd}$ with $\hat{H}_0 \equiv \sum_{i=1}^3 \hat{H}_{0,i}$ and $\hat{V}_{dd} \equiv \sum_{i<j} \hat{V}_{i,j}$. Moreover, with electronic states $\psi_{nlm}(r, \theta, \phi) = R_{nl}(r) Y_{lm}(\theta, \phi)$ [12], one gets the general formula

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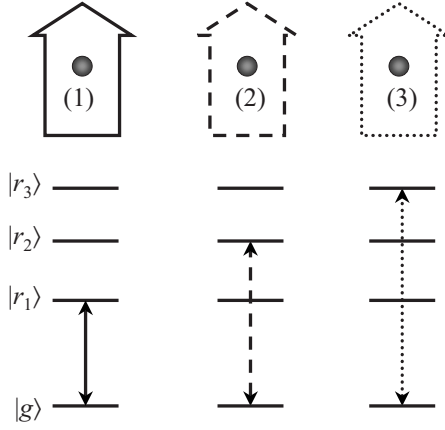


FIG. 1. Level scheme and laser excitations of the three-atom system.

$$\begin{aligned} & \langle n_i, l_i, m_i; n_j, l_j, m_j | \hat{V}_{ij} | n'_i, l'_i, m'_i; n'_j, l'_j, m'_j \rangle \\ &= \frac{e^2 \mathcal{R}_{n_i, l_i}^{n'_i, l'_i} \mathcal{R}_{n_j, l_j}^{n'_j, l'_j}}{4\pi\epsilon_0 R_{ij}^3} \\ & \times [\vec{\mathcal{A}}_{l_i, m_i}^{l'_i, m'_i} \cdot \vec{\mathcal{A}}_{l_j, m_j}^{l'_j, m'_j} - 3(\vec{\mathcal{A}}_{l_i, m_i}^{l'_i, m'_i} \cdot \vec{u}_{ij})(\vec{\mathcal{A}}_{l_j, m_j}^{l'_j, m'_j} \cdot \vec{u}_{ij})], \quad (2) \end{aligned}$$

where

$$\begin{aligned} \mathcal{R}_{n, l}^{n', l'} &\equiv \int_0^{+\infty} dr r^3 R_{nl}(r) R_{n'l'}(r), \\ \vec{\mathcal{A}}_{l, m}^{l', m'} &\equiv \int_0^\pi d\theta \sin \theta \int_0^{2\pi} d\phi \vec{e}_r(\theta, \phi) Y_{lm}^*(\theta, \phi) Y_{l'm'}(\theta, \phi) \end{aligned}$$

and $\vec{e}_r(\theta, \phi) = \sin \theta \cos \phi \vec{e}_x + \sin \theta \sin \phi \vec{e}_y + \cos \theta \vec{e}_z$ (see the Appendix for an explicit expression for $\mathcal{R}_{n, l}^{n', l'}$). Note that $\vec{\mathcal{A}}_{l, m}^{l', m'} \neq 0$ only if $l' = l \pm 1$ and $m' - m = 0, \pm 1$ (dipole selection rules).

In our setting, the three atoms, initially prepared in the ground state $|g\rangle$, can be submitted to different (sets of) laser beams which couple $|g\rangle$ to the Rydberg states $|r_1\rangle \equiv |ns\rangle$, $|r_2\rangle \equiv |np, k_2\rangle$, and $|r_3\rangle \equiv |nd, k_3\rangle$ (see Fig. 1). Population of states with arbitrary magnetic quantum numbers k_2 and k_3 is achieved through a proper choice of the polarization of the laser beams. If the dipole-dipole interaction were absent, the only populated three-atom states would be $|g; g; g\rangle$, $|r_1; g; g\rangle$, $|g; r_2; g\rangle$, $|g; g; r_3\rangle$, $|r_1; r_2; g\rangle$, $|r_1; g; r_3\rangle$, $|g; r_2; r_3\rangle$ and $|r_1; r_2; r_3\rangle$. The effect of V_{dd} on the ground state $|g; g; g\rangle$ and the singly Rydberg excited states is very small, and we shall neglect it; in contrast, V_{dd} strongly couples the doubly and triply Rydberg excited states to the rest of the Hilbert space.

Nevertheless, choosing n in such a way that all the couplings listed in Table I are nonresonant, we shall assume that we can restrict ourselves to the two resonant couplings $|ns; np\rangle \leftrightarrow |np; ns\rangle$ and $|np; nd\rangle \leftrightarrow |nd; np\rangle$. Note that, by virtue of selection rules, the states $|ns, nd\rangle$ and $|nd, ns\rangle$, though resonant, are not coupled by V_{dd} . The applicability of the previous assumption will be discussed below and quantitative conditions for its validity will be identified. For now, let

TABLE I. Relevant states and unwanted couplings.

Unwanted couplings
$ ns, np\rangle \leftrightarrow n_1 p, n_2 s\rangle$ for $(n_1, n_2) \neq (n, n)$
$ ns, np\rangle \leftrightarrow n_1 p, n_2 d\rangle$
$ ns, nd\rangle \leftrightarrow n_1 p, n_2 p\rangle$
$ ns, nd\rangle \leftrightarrow n_1 p, n_2 f\rangle$
$ np, nd\rangle \leftrightarrow n_1 s, n_2 p\rangle$
$ np, nd\rangle \leftrightarrow n_1 s, n_2 f\rangle$
$ np, nd\rangle \leftrightarrow n_1 d, n_2 p\rangle$ for $(n_1, n_2) \neq (n, n)$
$ np, nd\rangle \leftrightarrow n_1 d, n_2 f\rangle$

us assume these conditions are met: the state $|r_1; g; r_3\rangle$ is then unaffected by V_{dd} (in first order), whereas $|r_1; r_2; g\rangle$, $|g; r_2; r_3\rangle$, and $|r_1; r_2; r_3\rangle$ are shifted. The (first-order) shifts can be calculated by diagonalizing V_{dd} in the three degenerate subspaces

$$\mathcal{H}_{sp} = \text{Span}\{|ns; np; g\rangle, |np; ns; g\rangle\},$$

$$\mathcal{H}_{pd} = \text{Span}\{|g; np; nd\rangle, |g; nd; np\rangle\},$$

$$\begin{aligned} \mathcal{H}_{spd} = \text{Span}\{|ns; np; nd\rangle, |np; nd; ns\rangle, |nd; ns; np\rangle, |ns; nd; np\rangle, \\ |nd; np; ns\rangle, |np; ns; nd\rangle\}, \end{aligned}$$

where the magnetic quantum numbers $m_p = -1, 0, 1$ and $m_d = -2, -1, 0, 1, 2$ are implicit. Using Eq. (2) and taking the selection rules into account, one derives the following expression for V_{dd} in \mathcal{H}_{sp} :

$$V_{sp} = \frac{e^2 (\mathcal{R}_{ns}^{np})^2}{4\pi\epsilon_0 R_{12}^3} A_{sp}, \quad A_{sp} = \begin{pmatrix} 0 & M_{sp}^\dagger \\ M_{sp} & 0 \end{pmatrix},$$

where M_{sp} is a 3×3 matrix which contains coupling terms between $|ns; np, m_p\rangle$ and $|np, m'_p; ns\rangle$ of the form $[\vec{\mathcal{A}}_s^{p, m'_p} \cdot \vec{\mathcal{A}}_p^{p, m_p} - 3(\vec{\mathcal{A}}_s^{p, m'_p} \cdot \vec{u}_{12})(\vec{\mathcal{A}}_p^{p, m_p} \cdot \vec{u}_{12})]$. A_{sp} has six nonzero eigenvalues $\{\pm \frac{1}{3}, \pm \frac{1}{3}, \pm \frac{2}{3}\}$ which do not depend on the geometric configuration of the system (i.e., the angles and distances between the atoms). The corresponding energy shifts are given by $\Delta_{sp} = [e^2 (\mathcal{R}_{ns}^{np})^2 / 4\pi\epsilon_0 R_{12}^3] \{\pm \frac{1}{3}, \pm \frac{2}{3}\}$.

Similar results can be established in \mathcal{H}_{pd} : the energy shifts one obtains are nonzero and depend neither on distances nor on angles between atoms; their norms take values in the range $|\Delta_{pd}| = [e^2 (\mathcal{R}_{np}^{nd})^2 / 4\pi\epsilon_0 R_{23}^3] \{0.023 - 0.643\}$.

The case of \mathcal{H}_{spd} is more complicated, since V_{dd} now involves both sp - ps and pd - dp couplings: V_{spd} thus cannot be decomposed in as simple a way as V_{sp} and V_{pd} . The resulting eigenvalues will thus depend on the geometry of the three-atom system (one angle- and two distance variables, for instance). In principle, it is thus possible to find a specific arrangement so that one or more eigenvalues are null; in the generic situation, however, all the eigenvalues and the associated energy shifts Δ_{spd} are nonzero.

Finally, Δ_{sp} , Δ_{pd} , and Δ_{spd} prevent the states $|r_1; r_2; g\rangle$, $|g; r_2; r_3\rangle$, and $|r_1; r_2; r_3\rangle$ from being populated through resonant laser excitation of $|g; g; g\rangle$, whereas all the other unshifted states, and in particular $|r_1; g; r_3\rangle$, are accessible.

Before addressing the physical implementation of this situation in a rubidium atom system, let us turn back to the assumptions which allowed us to restrict ourselves to \mathcal{H}_{sp} , \mathcal{H}_{pd} , \mathcal{H}_{spd} . These assumptions are legitimate when the second-order shifts induced by the nonresonant couplings shown in Table I are negligible compared to the first-order shifts obtained above. To make sure this is satisfied, one has to verify that the smallest of the first-order shifts within each subspace, $\min(\Delta^{(1)})$, is much larger than the shifts obtained from the unwanted couplings. For instance, for the \mathcal{H}_{sp} subspace, the following condition must hold:

$$\frac{|\langle ns; np | V_{dd} | n_1 p; n_2 d \rangle|^2}{|E_{(ns; np)}^{(0)} - E_{(n_1 p; n_2 d)}^{(0)}|} \ll \min(\Delta_{sp}^{(1)}). \quad (3)$$

Let us now see how the previous situation can be implemented in a rubidium atom system. Assuming $n=42$ and $\vec{R}_{12}=\vec{R}_{23}=R\vec{e}_z$ with $R=5 \mu\text{m}$, and using atomic energy level data tables [13] we numerically checked both the nonresonance and the negligibility conditions Eq. (3) for the unwanted couplings listed in Table I. Then we calculated the dipole-dipole-interaction-induced shifts in the three degenerate subspaces \mathcal{H}_{sp} , \mathcal{H}_{pd} , and \mathcal{H}_{spd} , using $\mathcal{R}_{ns}^{np} \approx 2645a_0$ and $\mathcal{R}_{np}^{nd} \approx 2644a_0$ where a_0 is the Bohr radius ($a_0 \approx 5.3 \times 10^{-11} \text{m}$), which yielded $|\Delta_{sp}| \approx 6.1 \times 10^{-4} - 1.2 \times 10^{-3} \text{cm}^{-1} \approx 18-36 \text{MHz}$, $|\Delta_{pd}| \approx 4.2 \times 10^{-5} - 1.2 \times 10^{-3} \text{cm}^{-1} \approx 1.3-35 \text{MHz}$, and $|\Delta_{spd}| \approx 6.7 \times 10^{-5} - 2.2 \times 10^{-3} \text{cm}^{-1} \approx 2-67 \text{MHz}$. If the Rabi frequencies of the laser beams remain small compared to these shifts (here, typically, 1 MHz) the excitation of the corresponding states will be blocked. Conversely, it puts a lower bound of the order of 1 μs for the typical time duration of single-atom operations.

III. CONDITIONAL DYNAMICS IN A SYSTEM OF INDIVIDUAL ATOMS

Let us now see how to use the spectroscopic situation described in the previous section in order to induce conditional dynamics in individual-atom systems. To be specific, here, we show how to implement a three-qubit Toffoli gate [14] in the three-atom system considered above. A qubit of information is encoded in each of the three atoms on the ground state $|0\rangle \equiv |g\rangle$ and a low-lying excited state $|1\rangle \equiv |q\rangle$ (the Rydberg states will be only temporarily populated during the gate), to achieve conditional dynamics through dipole-dipole-interaction-induced shifts (see Fig. 2). Atoms 1, 2, 3 will, respectively, play the roles of control 1 (C_1), target (T) and control 2 (C_2) qubits.

The Toffoli gate is then implemented through the following three-step procedure (see Fig. 3).

(A) One first submits control atoms (C_1) and (C_2) to π -laser pulses which couple $|0\rangle$ to $|r_1\rangle$, and $|r_3\rangle$, respectively.

(B) One successively applies three π -laser pulses on the

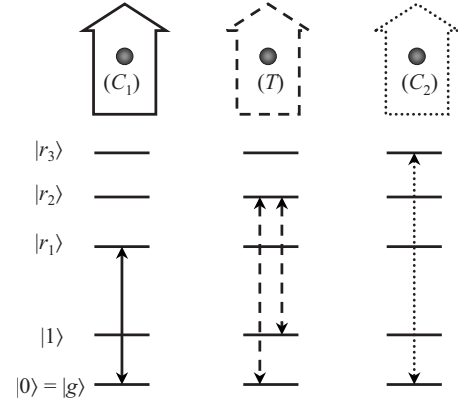


FIG. 2. Level scheme and laser excitations for implementing the Toffoli gate in the three-atom system.

target atom (T) which couple $|0\rangle$ to $|r_2\rangle$, $|1\rangle$ to $|r_2\rangle$, and $|0\rangle$ to $|r_2\rangle$, respectively (in the absence of the control atoms, this boils down to performing the Pauli matrix σ_x in the computational basis $|0\rangle, |1\rangle$).

(C) One repeats the first step.

If (at least) one of the atoms (C_1) and (C_2) is initially in the state $|0\rangle$, at the end of step A, (at least) one of the states $|r_1\rangle$ and $|r_3\rangle$ is excited: the shift induced by the dipole-dipole interaction between (T) and (C_1) and/or (C_2) will then prevent the target atom from being excited into the state $|r_2\rangle$, i.e., step B will not induce any change. In contrast, if both (C_1) and (C_2) are initially in $|1\rangle$, no interaction will shift the state $|r_2\rangle$, and step B will thus result in a σ_x gate on the target atom. Finally, the overall transformation is thus a Toffoli gate,

$$\text{TOFFOLI} = \text{CCNOT} = \begin{pmatrix} \mathbb{I}_6 & 0 \\ 0 & \sigma_x \end{pmatrix},$$

expressed in the computational basis $|c_1 c_2 t\rangle = |000\rangle, |001\rangle, |010\rangle, |011\rangle, |100\rangle, |101\rangle, |110\rangle, |111\rangle$. Note that, even though the same result can be achieved through combining one- and two-qubit elementary gates, our proposal involves only three steps and thus constitutes a more economical implementation of the Toffoli gate.

IV. CONDITIONAL DYNAMICS IN MESOSCOPIC ENSEMBLES

The same kind of conditional dynamics can also be performed on qubits stored in mesoscopic ensembles. To be specific, here, we shall show how to perform a controlled-controlled-PHASE (CCPHASE) gate in an ensemble made of the same atoms as in Sec. II. In addition to the ground state $|g\rangle$ and Rydberg states $|r_1\rangle$, $|r_2\rangle$, and $|r_3\rangle$, we shall need three extra long-lived atomic states $|q_{C_1}\rangle$, $|q_T\rangle$, and $|q_{C_2}\rangle$ (see Fig. 4). Following [15], we encode three qubits of information on the eight collective states $|000\rangle \equiv |g\rangle$, $|100\rangle \equiv |q_{C_1}^1\rangle$, $|010\rangle \equiv |q_{C_2}^1\rangle$, $|001\rangle \equiv |q_T^1\rangle$, $|110\rangle \equiv |q_{C_1}^1 q_{C_2}^1\rangle$, $|101\rangle \equiv |q_{C_1}^1 q_T^1\rangle$, $|011\rangle \equiv |q_{C_2}^1 q_T^1\rangle$, and $|111\rangle \equiv |q_{C_1}^1 q_{C_2}^1 q_T^1\rangle$ where $|q_{C_1}^1 q_{C_2}^1\rangle$ (for instance) denotes the symmetric collective state with one atom in $|q_{C_1}\rangle$ and another in $|q_{C_2}\rangle$.

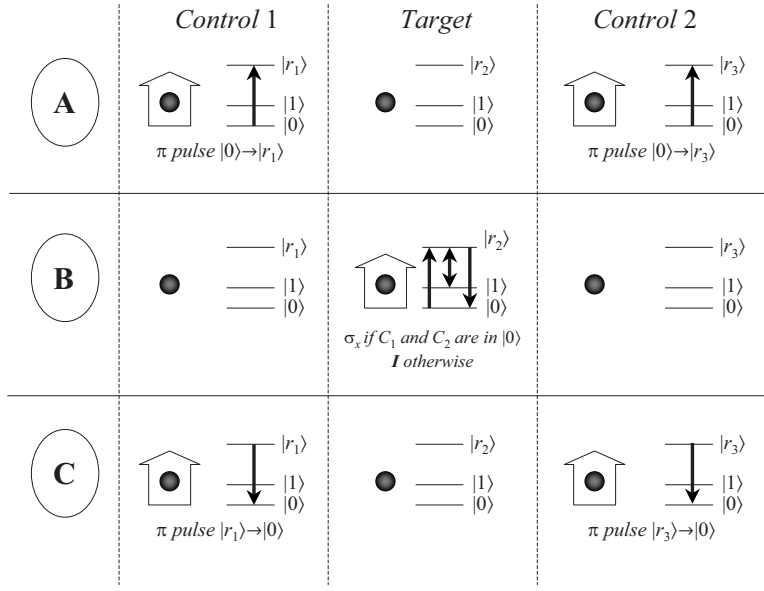


FIG. 3. Three-step implementation of a Toffoli gate in a three-atom system involving three different Rydberg states.

The following three-step procedure implements the desired CPHASE gate.

(A) One first submits the whole sample to two π laser pulses which couples $|q_{C_1}\rangle$ and $|q_{C_2}\rangle$ to the Rydberg states $|r_1\rangle$ and $|r_3\rangle$, respectively. This induces the following transformations: $|\mathbf{g}\rangle \rightarrow |\mathbf{g}\rangle$, $|\mathbf{q}_{C_1}^1\rangle \rightarrow |\mathbf{r}_1^1\rangle$, $|\mathbf{q}_{C_2}^1\rangle \rightarrow |\mathbf{r}_3^1\rangle$, $|\mathbf{q}_{C_1}^1 \mathbf{q}_{C_2}^1\rangle \rightarrow |\mathbf{r}_1^1 \mathbf{r}_3^1\rangle$, $|\mathbf{q}_{C_1}^1 \mathbf{q}_{C_2}^1\rangle \rightarrow |\mathbf{r}_1^1 \mathbf{r}_3^1\rangle$, $|\mathbf{q}_{C_1}^1 \mathbf{q}_{C_2}^1\rangle \rightarrow |\mathbf{q}_{C_1}^1 \mathbf{q}_{C_2}^1\rangle$, and $|\mathbf{q}_{C_1}^1 \mathbf{q}_{C_2}^1 \mathbf{q}_T^1\rangle \rightarrow |\mathbf{r}_1^1 \mathbf{r}_3^1 \mathbf{q}_T^1\rangle$.

(B) One then applies a 2π pulse on the ensemble which couples $|q_T\rangle$ to $|r_2\rangle$. The pulse will cause a transition in the ensemble only if none of the Rydberg levels $|r_1\rangle$ and $|r_3\rangle$ are excited: it follows that only the state $|\mathbf{q}_T^1\rangle$ will be multiplied by -1 , the other states being left unchanged.

(C) Finally, one applies the same two π laser pulses as in the first step, which induces the inverse transformations. The overall transformation is thus a CPHASE gate, which imposes a σ_z gate on the target qubit initially stored in the T ensemble if and only if the control ensembles C_1 and C_2 are initially in state $|0\rangle$.

V. CONCLUSION

In this paper, we proposed new configurations for dipole-dipole Rydberg interactions, involving different coupled and noncoupled Rydberg states. We think that such configura-

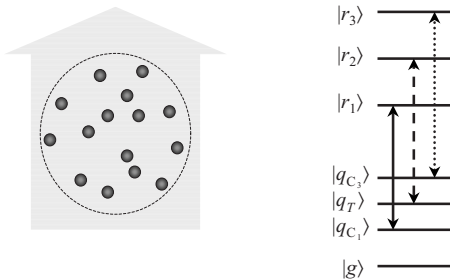


FIG. 4. Level scheme and laser excitations for implementing the CPHASE gate in an atomic ensemble.

tions are very promising and should allow for efficient implementation of sophisticated conditional dynamics beyond two-qubit gates. As first examples, we showed how to perform two specific three-qubit gates (the CCNOT and CPHASE gates) in an individual-atom system and in an atomic ensemble, through appropriately exciting atoms into three different Rydberg states. The feasibility of our schemes has been verified for the specific example of rubidium atoms.

We are currently investigating how such configurations with different Rydberg states could contribute, on the one hand, to extend the blockade phenomenon to macroscopic ensembles, and, on the other hand, to solve grid games like Latin squares quantum-mechanically.

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APPENDIX: CALCULATION OF THE RADIAL INTEGRAL

The radial part of the hydrogenic wave function takes the expression [16] $R_{nl}(r) = a^{-3/2} N_{nl} F_{nl}(2r/na)$, with $a = a_0/Z = \hbar^2/Zm'e^2$, $N_{nl} = (2/n^2)\sqrt{(n-l-1)!/(n+l)!^3}$ and $F_{nl}(x) = x^l e^{-x/2} L_{n-l-1}^{2l+1}(x)$, where $L_p^k(x)$ is an associated Laguerre polynomial. One can thus put the radial integral $\mathcal{R}_{n,l}^{n',l'}$ $= \int_0^{+\infty} dr r^3 R_{nl}(r) R_{n'l'}(r)$ in the form

$$\mathcal{R}_{n,l}^{n',l'} = a \frac{2^{l+l'} N_{nl} N_{n'l'} n^{4+l'} (n')^{4+l'}}{(n+n')^{4+l+l'}} \int_0^{+\infty} dx x^{3+l+l'} e^{-x} \times L_{n-l-1}^{2l+1} \left(\frac{2x}{1+n/n'} \right) L_{n'-l'-1}^{2l'+1} \left(\frac{2x}{1+n'/n} \right).$$

Using the definition $L_p^k(x) = \sum_{s=0}^p (-1)^s [(p+k)!] / [(p-s)!(k+s)!s!] x^s$, one finally gets the explicit expression

$$\mathcal{R}_{n,l}^{n',l'} = a 2^{l+l'+2} \frac{n^{2+l'} (n')^{2+l}}{(n+n')^{4+l+l'}} \sqrt{(n+l)! (n'+l')! (n-l-1)! (n'-l'-1)!} \sum_{r=0}^{n-l-1} \sum_{s=0}^{n'-l'-1} \left(-\frac{2}{n+n'}\right)^{r+s} \frac{n^s (n')^r}{r! s!} \\ \times \frac{(3+l+l'+r+s)!}{(n-l-r-1)! (2l+r+1)! (n'-l'-s-1)! (2l'+s+1)!}.$$

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