Coherent Control of an Atomic Collision in a Cavity

S. Osnaghi,¹ P. Bertet,¹ A. Auffeves,¹ P. Maioli,¹ M. Brune,¹ J. M. Raimond,¹ and S. Haroche^{1,2}

¹*Laboratoire Kastler Brossel, Département de Physique de l'Ecole Normale Supérieure, 24 rue Lhomond,*

F-75231, Paris Cedex 05, France

²*Collège de France, 11 place Marcelin-Berthelot, F-75005 Paris, France*

(Received 21 March 2001; published 27 June 2001)

Following a recent proposal by S. B. Zheng and G. C. Guo [Phys. Rev. Lett. **85**, 2392 (2000)], we report an experiment in which two Rydberg atoms crossing a nonresonant cavity are entangled by coherent energy exchange. The process, mediated by the virtual emission and absorption of a microwave photon, is characterized by a collision mixing angle 4 orders of magnitude larger than for atoms colliding in free space with the same impact parameter. The final entangled state is controlled by adjusting the atom-cavity detuning. This procedure, essentially insensitive to thermal fields and to photon decay, opens promising perspectives for complex entanglement manipulations.

DOI: 10.1103/PhysRevLett.87.037902 PACS numbers: 03.67. –a, 34.60. +z, 42.50.Gy

Conditional dynamics and quantum gates [1] involving individually addressable particles have been demonstrated in various quantum optics experiments. Beyond their fundamental interest to test basic aspects of quantum theory, these studies open new perspectives in quantum information processing [2]. In most schemes, information is carried by internal degrees of freedom of atomic particles, called "qubits." Logic gates are realized via coherent "collisions" between them. The qubits are put in contact on demand, coupled for a given time, then separated while the interaction with the environment, causing decoherence [3], is kept to a minimum. Such processes are quite different from usual collisions whose output is determined only statistically.

In ion trap experiments, the collision is achieved by establishing the contact between the qubits with lasers, through Raman processes involving the excitation of vibrational modes of motion of the ions [4]. In cavity quantum electrodynamics (CQED), a primary collision involves an atom and a cavity mode in exact resonance. The two systems are entangled as the atom exits the cavity [5,6]. Atom-atom entanglement is obtained by combining such atom-field collisions [7]. Other proposals involve collisions between cold atoms trapped by light, which are put in contact, then separated, by adiabatically changing the laser beam parameters [8]. Several schemes involve the coupling between atoms momentarily excited into Rydberg states [9]. Owing to their large electric dipoles, these states are ideal to achieve strong qubit interactions. Although it has not yet been used to build quantum gates, the van der Waals interaction between excited atoms has been investigated in the early days of Rydberg atom physics [10] and recently revisited in the context of cold atom studies [11].

Following a recent proposal [12], we describe in this Letter an experiment in which we control the collision of two Rydberg atoms in a process assisted by a nonresonant cavity. The atoms exchange their energy and get entangled while they cross together the cavity. This process bears similarities with light-induced atomic collisions [13], with the difference that, in the present case, the field modes enhancing the collision rate are essentially empty (vacuum field effect). The cavity makes the entanglement process about $10⁴$ times more efficient than a free space collision with the same impact parameter. The final atomic entangled state is tailored by adjusting the atom-cavity detuning. Contrary to previous CQED experiments [5–7], this cavityassisted entanglement process leaves the field unexcited and is essentially insensitive to thermal cavity excitations and to cavity losses. These features make this method very promising for quantum information processing [2,12]. This cavity-assisted collison process can also be related to recently proposed [14,15] and implemented [16] schemes in ion trap physics, in which atomic entanglement is realized via virtual vibrational excitations of the ions.

Let us first recall some orders of magnitude relevant to van der Waals collisions between Rydberg atoms in free space. We consider the resonant energy exchange $|e_1, g_2\rangle \rightarrow |g_1, e_2\rangle$ between two atoms A_1 and A_2 initially in states *e* and *g*, respectively (*e* and *g* correspond to large principal quantum numbers *n* and $n - 1$). The atoms interact via the dipole-dipole coupling $W_{vdW}(R, \mathbf{u}) =$ $\left[\mathbf{r}_1 \cdot \mathbf{r}_2 - 3(\mathbf{r}_1 \cdot \mathbf{u}) (\mathbf{r}_2 \cdot \mathbf{u}) \right] q^2 / 4 \pi \epsilon_0 R^3$, where \mathbf{r}_i (*i* = 1, 2) are the valence electron (charge *q*) coordinates in each atom, *R* and **u** the distance between the atoms and the unit vector along the interatomic direction, respectively. The expression of W_{vdW} , simply derived from electrostatic laws, can also be viewed as resulting from virtual photon exchange between the atoms, summed over all possible field modes. A collision is characterized by the functions of time $R(t)$ and $\mathbf{u}(t)$ which determine the "collision mixing" \arg e" $\theta = (1/\hbar) \int dt \, |\langle e_1, g_2 | W_{\text{vdW}}[R(t), \mathbf{u}(t)] | g_1, e_2 \rangle|.$ The atoms emerge from the collision in the generally entangled state:

$$
|\Psi\rangle = \cos\theta |e_1, g_2\rangle + \exp(i\Phi)\sin\theta |g_1, e_2\rangle, \quad (1)
$$

where Φ is a phase depending on $R(t)$ and $\mathbf{u}(t)$ which we need not specify.

Even in free space, the large-sized Rydberg atoms are very sensitive to the van der Waals interaction. For a collision with an impact parameter b_0 , involving atoms with relative velocity v , an order of magnitude estimate yields

$$
\theta_0 = n^4 \frac{q^2}{4\pi\varepsilon_0 \hbar v} \frac{a_0^2}{b_0^2} = \alpha \frac{c}{v} \left(\frac{a_0 n^2}{b_0}\right)^2, \qquad (2)
$$

where $a_0 = 0.53 \times 10^{-10}$ m is the Bohr radius and $\alpha =$ $q^2/4\pi\epsilon_0\hbar c = 1/137$ the fine structure constant. For $n = 51$ and $v/c = 10^{-6}$ (typical atomic beam velocity), the condition $\theta_0 = \pi/4$ of maximum entanglement is achieved for $b_0 = 13 \mu m$, a huge distance at atomic scale. We show in this Letter that, by having the atoms interact not in free space, but in a cavity, the Rydberg-Rydberg collision angle is enhanced by a huge factor, making it possible for the atoms to get entangled while they collide at millimetric distances.

Our setup is shown in Fig. $1(a)$. The Rb atoms, effusing from an oven *O*, propagate along a horizontal beam crossing the cavity *C* made of two superconducting niobium spherical mirrors placed at $L = 2.75$ cm from each other $[5-7,17]$. The setup is cooled to 1.3 K to minimize thermal radiation. The atoms are velocity selected by laser optical pumping, according to a procedure described elsewhere [5]. They are then prepared in box *B* by a combination of laser and radiofrequency excitation in the circular Rydberg states with principal quantum numbers $n = 51$ (*e*) or 50 (*g*). The atoms, prepared with different velocities, collide inside *C* which they cross at mid-distance between the mirrors. After exiting the cavity, they are detected by a state selective field ionization detector *D* (efficiency 40%), discriminating with less than 5% error rate *e* and *g*. An optional classical microwave pulse *R* coherently mixing *e*

FIG. 1. (a) Scheme of the experimental apparatus. (b) Spacetime diagram depicting the sequence of events. Atoms *A*¹ and A_2 , sent at different times with velocities v_1 and v_2 , simultaneously cross the cavity axis at time $t = 0$. They undergo an optional microwave pulse *R* before being detected by field ionization in *D*.

037902-2 037902-2

and $g(\pi/2)$ pulse) can be applied to the atoms after *C*, for analyzing the final state of the collision process. The sequence of events is schematized in Fig. 1(b), which shows a space-time diagram depicting the evolution of the two atoms crossing the apparatus.

The Rydberg excitation is pulsed within a time of 2 μ s. In each atomic pulse, we prepare on the average 0.25 atom, with Poisson statistics. The probabilities for exciting 0, 1, and 2 atoms per pulse are, respectively, 0.78, 0.19, and 0.025. Events in which only one atom is detected in the two pulses (0.6% of the experimental sequences) are recorded. In approximately 25% of these events, there are in fact two atoms in one of the pulses, one of them escaping detection. These "three atom collision" events are a source of errors. We focus here on the case where a simple *A*1-*A*² pair has been prepared. We choose the delay *T* between the preparation pulses and the two atomic velocities v_1, v_2 such that A_1 overcomes A_2 at cavity center. This event defines the time origin $t = 0$. The $A_1 - A_2$ separation at $t = 0$ is of the order of the atomic beam diameter, about 0.5 mm. This would be the impact parameter for the same collision process in the absence of the cavity, corresponding to a negligible entanglement ($\theta_0 \approx 5.10^{-4}$).

The cavity sustains two TEM₉₀₀ modes, M_a and M_b , with linear orthogonal polarizations and transverse Gaussian profiles (common waist at center $w = 0.6$ cm). Because of a small mirror anisotropy, the mode degeneracy is lifted (frequency difference $\Delta/2\pi = 128$ kHz). The two frequencies ω_a and $\omega_b = \omega_a + \Delta$ can be tuned together by translating the mirrors with a piezostack. The modes are frequency shifted from the atomic $e \rightarrow g$ transition frequency $\omega/2\pi = 51.1$ GHz by variable detunings δ_a and $\delta_b = \delta_a + \Delta$. The field mode damping times are $T_{c,a} = 10^{-3}$ s and $T_{c,b} = 0.9 \times 10^{-3}$ s. The maximum vacuum field rms amplitude in each mode is $E_0 = (2\hbar\omega/\pi\epsilon_0 Lw^2)^{1/2} = 1.57 \times 10^{-3}$ V/m. At equilibrium, there is an average of about one thermal photon per mode, due to microwave leaks in *C*. These photons are erased at the beginning of each experimental sequence by sending a train of absorbing atoms across *C* [17]. During the 180 μ s delay between the end of the erasing sequence and the A_1 - A_2 collision, a field of about 0.25 photon builds up in each mode.

In a first experiment, the *R* pulse is not used. The delay between the atomic preparations is $T = 78 \mu s$, with $v_1 =$ 300 m/s, $v_2 = 243$ m/s. We sweep δ_a (and $\delta_b =$ $\delta_a + \Delta$). For each detuning value, we detect 1000 atomic pairs and we reconstruct the four detection probabilities *P*(e_1, g_2), *P*(g_1, e_2), *P*(e_1, e_2), and *P*(g_1, g_2). Figure 2 shows the variations of these probabilities as a function of the dimensionless detuning parameter $\eta = (\omega/\delta_a +$ ω/δ_b). We see that $P(e_1, g_2)$ and $P(g_1, e_2)$ (solid and open circles, respectively) oscillate in a symmetrical way as a function of η . These variations reflect the pattern described by Eq. (1), the detuning parameter η being—as we show below—directly related to the cavity-assisted

FIG. 2. Joint detection probabilities versus the detuning parameter η . *P*(e_1 , g_2) and *P*(g_1 , e_2) (solid and open circles) oscillate in a symmetrical way, reflecting the atom-atom energy exchange enhanced by the cavity. The solid line represents the predictions of Eq. (4), in the $\eta < 5 \times 10^5$ range where it applies. The dashed lines present the result of a numerical integration of the system's evolution. The spurious channel probabilities, $P(e_1, e_2)$ and $P(g_1, g_2)$ (open squares and diamonds, respectively), stay below the 10% level.

collision angle. The other probabilities $P(e_1, e_2)$ and $P(g_1, g_2)$ are due to erroneous detection counts and to three-atom collisions. They remain, on the average, at a low background level (about 10%). To get a "zero-collision angle" reference, we have changed *T* to 108 μ s, so that the atoms now crossed 37 mm downstream from the cavity axis and set $\delta/2\pi = 470$ kHz. We measured then $P(g_1, e_2) = 0, 01 \ (\pm 0.01)$ and $P(e_1, g_2) = 0.89 \ (\pm 0.01)$ instead of the ideal 1 value, due to detection errors. This demonstrates that the collision effect observed here is fully cavity-assisted as discussed above. The corresponding experimental points have been put in Fig. 2 at $\eta = 0$ (equivalent to "infinite" cavity detuning).

The coupling of A_1 and A_2 to each mode depends upon atomic positions. At cavity center $(t = 0)$, this coupling is characterized by the Rabi frequency $\Omega = 2D_{eg} \cdot E_0/\hbar$, where $D_{eg} = qa_0 n^2/2$ is the dipole matrix element between *e* and *g* ($\Omega/2\pi$ = 50 kHz deduced from atomic and cavity parameters, to be compared with the experimental value $\Omega/2\pi = 49 \pm 1$ kHz [18]). At time *t*, A_1 and *A*² have moved away from cavity center, and their coupling to each cavity mode is $\Omega \exp(-v_i^2 t^2/w^2)$ $(i = 1, 2)$. The energy exchange process involves the virtual emission by the first atom of one photon in one mode $[|e_1, g_2; 0_\mu\rangle \rightarrow$ $|g_1, g_2; 1_\mu\rangle; \mu = (a, b)$, combined to the photon absorption by the second atom $(|g_1, g_2; 1_\mu\rangle \rightarrow |g_1, e_2; 0_\mu\rangle)$. It is easy to compute the effect of such virtual transitions in the limit $\delta_a \gg \Omega$ (nonresonant cavity QED regime). The contribution of the intermediate state $|g_1, g_2; 1_u\rangle$ to the mixing angle involves the product of the atom's couplings divided by the frequency mismatch δ_a . Summing over the modes and averaging the variations of the atom-cavity coupling, we find the cavity-assisted collision mixing angle:

$$
\theta_c = \Omega^2 \bigg(\frac{1}{\delta_a} + \frac{1}{\delta_b} \bigg) \frac{\sqrt{\pi}}{4\sqrt{2}} \frac{w}{v_0},\tag{3}
$$

with $v_0 = \left[(v_1^2 + v_2^2)/2 \right]^{1/2}$.

Finally, replacing, in Eq. (3), Ω by its expression in terms of D_{eg} and E_0 and these quantities by their expressions in terms of cavity and atom parameters, we find

$$
\theta_c = \alpha \bigg(\frac{\omega}{\delta_a} + \frac{\omega}{\delta_b} \bigg) \frac{c}{v_0} \bigg(\frac{a_0 n^2}{b_c} \bigg)^2, \tag{4}
$$

with $b_c = (Lw)$ p $\sqrt{2\pi}$)^{1/2} = 0.81 cm.

While Eq. (2) is qualitative, Eq. (4) is exact for $\delta_a \gg \Omega$. Comparing Eqs. (4) and (2), we find that θ_c is obtained by multiplying by η the free space collision angle corresponding to an impact parameter $b_0 = b_c$. Since b_c is about 3 orders of magnitude larger than the $b₀$ value corresponding to $\theta_0 = \pi/4$ in free space, maximum entanglement in the cavity is achieved with η of the order of 10⁶. The solid lines in Fig. 2 are obtained by replacing, in Eq. (1), θ_0 by θ_c and by multiplying $P(e_1, g_2)$ and $P(g_1, e_2)$ by 0.89, in order to fit the data at $\eta = 0$ and thus account for detection errors. We note a good agreement between the experiment and this simple model for $\eta < 5 \times 10^5$, i.e., $\delta_a > 3 \Omega$.

In this perturbative regime, the collision is to first order insensitive to thermal photons. If there are N_{μ} photons in one mode, the virtual process in which an additional photon is emitted $(|e_1, g_2; N_\mu\rangle \rightarrow |g_1, g_2; N_\mu + 1\rangle \rightarrow$ $|g_1, e_2; N_\mu\rangle$ interferes destructively with the one in which a photon is absorbed $(|e_1, g_2; N_\mu\rangle \rightarrow |e_1, e_2; N_\mu - 1\rangle \rightarrow$ $|g_1, e_2; N_\mu\rangle$, since the corresponding amplitudes have opposite signs. The net result is N_{μ} independent and identical to the one obtained for a cavity in its vacuum state. This is verified by observing that the solid line theoretical curve, computed by assuming $N_{\mu} = 0$, fits well the results of the experiment, in which the probability to have $N_{\mu} = 1$ is 0.25. A similar insensitivity to thermal excitations of atom-atom interaction mediated by virtual coupling to a vibration mode occurs in ion traps [14,15].

For larger η values, the condition $\delta_a/\Omega \gg 1$ is no longer valid and the collision angle departs from the perturbative expression [Eq. (4)], even for cavity modes at zero temperature. In addition, the effect of thermal field excitations cannot then be neglected. Thus, we numerically solve the equations of motion of the two atoms in the cavity, taking into account exact atom-cavity coupling as well as the 0.25 thermal photons per mode (but neglecting cavity relaxation during interaction time). The theory is normalized to fit the data at $\eta = 0$. The results are given by the dotted curves in Fig. 2, which reproduce qualitatively well the variations of $P(e_1, g_2)$ and $P(g_1, e_2)$ in the whole range of η values up to a 2π collision angle. Note, however, that, for large η values, the contrast of the experimental oscillations is smaller than the theoretical one.

FIG. 3. "Transverse" correlations: Bell signal $\langle \sigma_{1x} \sigma_{2,\phi} \rangle$ versus relative phase ϕ . The modulation reveals the coherence of the cavity-assisted collision process.

Part of this contrast reduction originates in three-atom collision processes. For small η 's, up to the point $\theta_c = \pi/4$ of maximum entanglement, the two-atom collision model is quite satisfactory.

The coherence of the cavity-assisted collision is checked in a second experiment. We choose a different set of parameters: $T = 115 \mu s$, $v_1 = 500 \text{ m/s}$, $v_2 = 319 \text{ m/s}$. Fixing η to realize $\theta_c = \pi/4$, we apply independently to A_1 and A_2 a $\pi/2$ pulse *R* (with a frequency ω_r close to ω), realizing a basis change in the *e*-*g* subspace. The delay between the pulses is $\tau = 22 \mu s$. In the Bloch vector representation, the $|e\rangle$ and $|g\rangle$ states correspond to a "pseudospin" along the "*Oz* axis." Detecting the energy of *A*¹ after the *R* pulse amounts to a "transverse" detection of the corresponding pseudospin. Finding A_1 in e (respectively, *g*) is then equivalent to measuring it along the "*Ox* axis" [eigenstate $|+_x\rangle$ of the Pauli matrix σ_x (respectively, $|-\mathbf{x}\rangle$]. For A_2 , we detect in the same way the states $|\pm_{\Phi}\rangle$ which are the eigenstates of $\sigma_{\phi} = \cos \phi \sigma_{x} +$ $\sin \phi \sigma_y$, with $\phi = (\omega - \omega_r)\tau$.

By repeating the experiment while sweeping ω_r (thus ϕ), we reconstruct the combination of joint probabilities $P(+_{1,x}; +_{2,\phi}) + P(-_{1,x}; -_{2,\phi}) - P(+_{1,x}; -_{2,\phi}) P(-_{1,x}; +_{2,\phi}) = \langle \sigma_{1,x} \sigma_{2,\phi} \rangle$. This "Bell signal," shown in Fig. 3 versus ϕ , measures the angular correlations between the transverse spin components associated to the two atoms. Ideally, the process should prepare a pair of maximally entangled EPR particles, with a signal oscillating between $+1$ and -1 . The reduced contrast of the observed modulation, about 50%, is due to the already mentioned defects in the entangled state preparation, as well as imperfections in the *R* pulses.

After we have improved our setup (notably by preparing the atoms via a deterministic and not a Poissonian process), many promising experiments generalizing the present study will become possible. By combining a twoatom cavity-assisted collision with single atom unitary operations, robust quantum gates directly coupling atomic qubits could be realized [12] and new tests of Bell's inequalities with atoms performed. Situations where three atoms at a time cross the cavity and interact with its field via real or virtual photon processes could lead to the realization of useful three-bit logical gates.

We acknowledge support of the European Community and of the Japan Science and Technology Corporation (International Cooperative Research Project: Quantum Entanglement). Laboratoire Kastler Brossel, Université Pierre et Marie Curie and ENS, is associated with CNRS (UMR 8552).

- [1] D. P. DiVincenzo, Science **270**, 255 (1995); A. Ekert and R. Josza, Rev. Mod. Phys. **68**, 3733 (1997).
- [2] *The Physics of Quantum Information,* edited by D. Bouwmeester, A. Ekert, and A. Zeilinger (Springer-Verlag, Berlin, 2000).
- [3] W. H. Zurek, Phys. Rev. D **24**, 1516 (1981); **26**, 1862 (1982); A. O. Caldeira and A. J. Leggett, Physica (Amsterdam) **121A**, 587 (1983); E. Joos and H. D. Zeh, Z. Phys. B **59**, 223 (1985).
- [4] C. Monroe *et al.,* Phys. Rev. Lett. **75**, 4714 (1995).
- [5] E. Hagley *et al.,* Phys. Rev. Lett. **79**, 1 (1997).
- [6] A. Rauschenbeutel *et al.,* Phys. Rev. Lett. **83**, 5166 (1999).
- [7] A. Rauschenbeutel *et al.,* Science **288**, 2024 (2000).
- [8] G. K. Brennen *et al.,* Phys. Rev. Lett. **82**, 1060 (1999).
- [9] D. Jaksch *et al.,* Phys. Rev. Lett. **85**, 2208 (2000).
- [10] J. M. Raimond, G. Vitrant, and S. Haroche, J. Phys. B **14**, L655 (1981).
- [11] M. P. Robinson *et al.,* Phys. Rev. Lett. **85**, 4466 (2000).
- [12] S. B. Zheng and G. C. Guo, Phys. Rev. Lett. **85**, 2392 (2000).
- [13] P. S. Julienne, A. M. Smith, and K. Burnett, Adv. At. Mol. Opt. Phys. **30**, 141 (1993).
- [14] K. Mölmer and A. Sorensen, Phys. Rev. Lett. **82**, 1835 (1999).
- [15] E. Solano, R.L. de Matos Filho, and N. Zagury, Phys. Rev. A **59**, R2539 (1999); **61**, 029903(E) (2000).
- [16] C. A. Sackett *et al.,* Nature (London) **404**, 256 (2000).
- [17] G. Nogues *et al.,* Nature (London) **400**, 239 (1999).
- [18] The latter value, obtained by a direct recording of the Rabi oscillation for $\delta_a = 0$, is slightly larger than the 47 kHz value reported in M. Brune *et al.,* Phys. Rev. Lett. **76**, 1800 (1996). The increase results from a better alignment of the atomic beam which now crosses the cavity mode closer to its central antinode.