

# Electric-field induced dipole blockade with Rydberg atoms

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High resolution laser Stark excitation of  $np$  ( $60 < n < 85$ ) Rydberg states of ultra-cold cesium atoms shows an efficient blockade of the excitation attributed to long-range dipole-dipole interaction. The dipole blockade effect is observed as a quenching of the Rydberg excitation depending on the value of the dipole moment induced by the external electric field. Effects of eventual ions which could match the dipole blockade effect are discussed in detail but are ruled out for our experimental conditions. Analytic and Monte-Carlo simulations of the excitation of an ensemble of interacting Rydberg atoms agree with the experiments indicates a major role of the nearest neighboring Rydberg atom.

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Long-range dipole-dipole interactions often play an important role in the properties of an assembly of cold atoms. One example is the efficiency of photoassociation of cold atoms and the formation of cold molecules [1]. In the case of a Rydberg atomic ensemble, the range of the dipole-dipole interactions can exceed several micrometers, leading to many-body effects [2, 3, 4]. An interesting application of the dipole-dipole interaction is the dipole blockade (DB) in Rydberg excitation. This effect offers exciting possibilities for quantum information [5] with the fascinating possibilities for manipulating quantum bits stored in a single collective excitation in mesoscopic ensembles, or for realizing scalable quantum logic gates [6]. The DB process for an ensemble of atoms is the result of shifting the Rydberg energy from its isolated atomic value due to the dipole-dipole interaction with the surrounding atoms. In a large volume, a partial or local blockade, corresponding to a limitation of the excitation is expected when the dipole-dipole energy shift exceeds the resolution of the laser excitation. In a zero electric field, Rydberg atoms present no permanent dipole and usually no DB is expected. Nevertheless, a van der Waals blockade, corresponding to a second order dipole-dipole interaction, has been observed through a limitation of the excitation of high Rydberg states  $np$  ( $n \sim 70 - 80$ ) of rubidium, using a pulsed amplified single mode laser [7]. CW excitations have also been performed [8, 9] showing the suppression of the excitation and affecting the atom counting statistics [8]. The DB phenomenon itself has been observed for the first time, in the case of cesium Rydberg atoms, for a so called Förster Resonance Energy Transfer (FRET) reaction,  $np + np \rightarrow ns + (n + 1)s$  [10]. The FRET configuration has several advantages: the dipole-dipole interaction can be tuned on and off by the Stark effect, the dipole-dipole interaction having its

maximum effect at the resonant field. Its main drawback comes from the fact that the resonance exists only for  $n \leq 41$  in the cesium case, which limited the observed DB to an efficiency of  $\sim 30\%$ .

In this letter, we report the experimental evidence for a DB with an efficiency larger than 60 %. In order to obtain higher couplings than the van der Waals or Förster ones, we apply an electric field to create a significant dipole moment. Indeed, the Stark Rydberg state,  $np$ , presents an electric dipole moment mostly due to their mixing with the  $(n - 1)d$  state. It is however experimentally challenging to observe the dipole blockade effect without any ambiguity. Ions could for instance lead to a similar effect as the DB one as shown by Fig 1 [discussed latter]. The letter is presented as following. First we discuss the role of ions and the importance of removing almost all ions in the experiment. We then report the DB results which are finally compared to an analytical model.

In every Rydberg DB experiment the role of ions needs to be studied carefully because the presence of a single ion creates a spurious electric field of 1.5 mV/cm at a distance of 100  $\mu\text{m}$ . The role of ions is often underestimated and can lead to a blockade similar to the Van der Waals blockade effect. Furthermore, due to the quadratic variation of the Rydberg  $np$  energy level as a function of the external electric field, the Stark effect created by ions is enhanced compared to the zero field configuration. In the Förster configuration, due to the low  $n$  value, it was clearly seen [10] that the presence of some ions did not affect the observed limitation of the excitation. However, for higher  $n$  value, the presence of a single ion during the excitation could easily, even for a moderate external electric field, shifts the Rydberg energy level by few MHz, which is on the order of the laser resolution. In other words, the appearance of a single ion can stop the excitation. To investigate in more detail how strong the effect of ions is, two kinds of numerical simulations have been performed. The result of the first model (see Fig.1 (a)), based on single atoms reduced density matrix evolution in the presence of the nearest neighbors dipole-dipole

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interaction (detailed hereafter), illustrates the role of a single ion present at the beginning of the excitation towards the  $70p$  state. Such ion would affect drastically the results because it has almost the same effect as the dipole-dipole interaction without any ion present. However, in our experiment the ions do not appear at the beginning of the excitation, but only when Rydberg atoms are present. Indeed, we have experimentally studied the temporal evolution of the number of ions and found a constant rate of ionization and a linear dependence on the number of Rydberg atoms. The origins of the ions are mainly thermal blackbody radiation and collisional processes between Rydberg atoms and surrounding hot  $6s$  or cold  $6p, 7s$  atoms (in preparation). Rydberg atom-Rydberg atom collisions, such as Penning ionization occurring from the pair dynamics under the influence of the attractive long-range forces [11, 12], occur only after one microsecond. We have then derived a second model taking into account a constant ionization rate of the Rydberg atoms. The result of the second model, based on a kinetic Monte Carlo simulation, illustrates (see Fig.1 (b)) the role of such a Rydberg ionization, but here with a rate 20 times larger than the experimental one, if present during the excitation. This would also affect drastically the results because few ions have almost the same effect than the dipole-dipole interaction when no ions are present. The kinetic Monte-Carlo algorithm has been chosen because it gives the exact time evolution of rate equations system [13]. Here the rate equations describe the (2-level) atoms excitation, the sum of all possible two-body dipole-dipole interactions and the external and ionic electric fields effects. In conclusion, the main result of these two types of simulations is that ions can lift any Rydberg density effect such as the DB one.

Therefore, the number of ions has to be minimized experimentally, for instance by avoiding the excitation of too many Rydberg atoms. The details of the experimental setup have been described in reference [10]. The Rydberg atoms are excited from a cloud of  $5 \times 10^7$  cesium atoms (characteristic radius  $\sim 300 \mu\text{m}$ , peak density  $1.2 \times 10^{11} \text{ cm}^{-3}$ ) produced in a standard vapor-loaded magneto-optical trap (MOT). The first step of the excitation,  $6s, F = 4 \rightarrow 6p_{3/2}, F = 5$ , is provided either by the trapping lasers (wavelength:  $\lambda_1 = 852 \text{ nm}$ ) or by an independent laser. The density of excited,  $6p_{3/2}$ , atoms can be modified by switching off the repumping lasers before the excitation sequence. The second step,  $6p_{3/2}, F = 5 \rightarrow 7s, F = 4$ , is provided by an infrared diode laser in an extended cavity device from TOPTICA (wavelength:  $\lambda_2 = 1.47 \mu\text{m}$ , bandwidth: 100 kHz). The experimental average intensity is  $\sim 3 \text{ mW/cm}^2$  twice the saturation one. The last step of the excitation,  $7s, F = 4 \rightarrow np_{1/2,3/2}$  (with  $n = 25 - 140$ ), is provided by a Titanium:Sapphire laser (Ti:Sa), wavelength  $\lambda_3 = 770 - 800 \text{ nm}$ , bandwidth: 1 MHz. The Ti:Sa laser is switched on with a fixed optical frequency during a

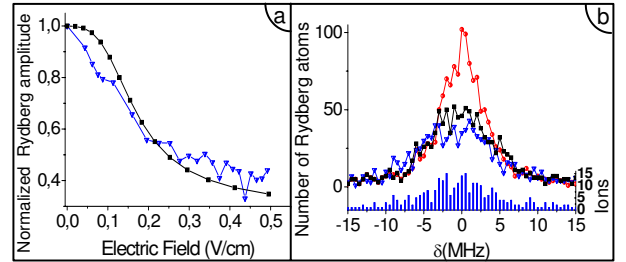


FIG. 1: (Color online) Comparison of the DB with the ion blockade effect. Number of atoms excited from ground state to the  $70p_{3/2}$  Rydberg state after a 300ns laser excitation. (a) Reduced density matrix model (see text) for different external electric field values. Down triangles: a single ion is randomly present in the sample before the laser excitation but no dipole-dipole interaction is taken into account. Squares: calculation taking into account only the dipole-dipole interaction with no ions. (b) Kinetic Monte Carlo simulation of the laser excitation, versus the detuning from resonance, of 8500 ground state atoms, in a spherical gaussian cloud of  $\sigma = 30 \mu\text{m}$  ( $1/e^{1/2}$ ) radius at a given electric field ( $F=0.4\text{V/cm}$ ). No dipole interactions (circles), with no dipole interactions but with ions appearing from Rydberg ionization (down triangles), dipole interactions but no ions (squares).

time,  $\tau = 0.3 \mu\text{s}$ , by means of an acousto-optic modulator. Due to the short  $7s$  lifetime and the short excitation time the spectral resolution of the excitation  $\Delta_L$  is only on the order of 5 MHz (when no Ti:Sa optical saturation is present). The Rydberg atoms are selectively field ionized by applying, at a chosen time (between 0 and  $1 \mu\text{s}$ ) after the end of the Ti:Sa laser pulse, a high field pulse with a rise time of 700 ns. We use a time of flight technique and a Micro Channel Plate (MCP) detector to know the total number of detected ions at the MCP. From the  $\sim 30\%$  quantum detection efficiency of the MCP we estimate the number of Rydberg atoms present in the experimental area. We define the density as  $n = N/(2\pi\sigma^2)^{3/2}$ . The beams of the infrared diode laser and Ti:Sa laser are focused into the atomic cloud with a  $(1/e^2)$  radius  $2\sigma = 70 - 125 \mu\text{m}$ . The repetition rate of the experiment is 80 Hz. The Ti:Sa laser polarization is linear and parallel to the direction of the applied electric field, leading to the excitation of the magnetic sublevel,  $np_{1/2}$  or  $np_{3/2}$   $|m| = 1/2$ . For clarity we will present here only results obtained with  $np_{3/2}$  atoms, but similar results have been obtained when using the  $np_{1/2}$  state [14].

Fig.2 (a) displays an ensemble of spectra of the excitation of the level,  $75p_{3/2}$ , for different values of the static electric field,  $F$ , indicating the limitation of the excitation induced by the electric field. Fig.2 (b) displays two spectra of the resonance line for the excitation of the level,  $70p_{3/2}$ , at zero field and at  $F = 0.25 \text{ V/cm}$ . A clear effect appears at the center of the resonance line with a 60% decreasing of the intensity of the line. At

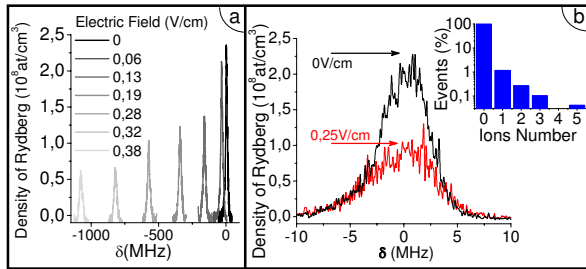


FIG. 2: (Color online) (a) Excitation spectral lines of the  $75p_{3/2}$  level recorded for different electric fields with  $\sigma = 120\mu\text{m}$ . (b) Excitation spectral lines of the  $70p_{3/2}$  level for two different fields, as a function of the detuning from the resonance frequency of the transition  $7s \rightarrow 70p_{3/2}$  with  $\sigma = 35\mu\text{m}$ . Events of MCP detected ions during the scan are also presented (inset, log scale).

this point, it is very important to know if this reduction of the excitation can be attributed to DB or due to ion blockade. In the present experiment, for evacuating the role of any ions, we have performed the experiment with conditions where the observed number of ions is on average well below unity as shown in inset of Fig. 2 (b). This absence of ions is confirmed by the fact that the appearance of ions would be correlated with the Rydberg signal. Indeed, in the simulation, but not in the experiment, we observe a correlation between the ions and the Rydberg signal, leading to the limitation or blockade of the excitation when ions appear (see Fig 1 (b), down triangles).

We then attribute the limitation of the excitation to a DB and not to an ion blockade. We stress here that the wings of the resonance should (and are) not affected by the DB because the number of excited Rydberg atoms is reduced. However, in our experiment an apparent increasing of the linewidth (FWHM) is observed because the line has no more a Lorentzian shape.

In Fig. 2 (a) we have observed a quite efficient decrease of the number of excited atoms with  $F$  for  $n = 75$  due to the DB effect coming from the existence of a non zero permanent dipole, aligned with the static electric field  $\vec{F}$ ,  $\langle np, F | q_e \vec{r} | np, F \rangle = \vec{\mu}$ . Similarly, Fig.3 (a) demonstrates the DB effect for the excitation of different levels,  $np_{3/2}$ , with  $n$  equal to 60, 70 and 85. In this study, the intensity of the Ti:Sa laser is set to be inversely proportional to the oscillator strength of the transition,  $7s \rightarrow np$ , meaning a  $n^3$  dependence. To compare the curves obtained for different principal quantum numbers  $n$ , it is convenient to introduce the scale parameter  $\theta$  characterizing the dipole coupling for each level,  $np$  defined by  $\tan \theta = \frac{|W_n|}{h\Delta_n/2}$ , where  $h\Delta_n$  is the zero field energy difference between the  $(n-1)d$  and  $np$  levels and  $W_n$  the Stark coupling  $W_n = \langle (n-1)d_j, m | -q_e \vec{r} \cdot \vec{F} | np_j m \rangle$ . Fig. 3 (b) shows the decreasing of the Rydberg excitation for  $np_{3/2}$  states, with  $n$  equal to 60 and 85, versus  $\tan^2 \theta$ .

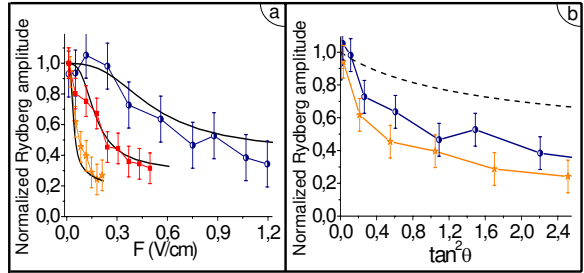


FIG. 3: (Color online) (a) Fraction of the number of Rydberg atoms excited at different electric fields compared to the number of Rydberg atoms excited at zero field, for different  $np_{3/2}$  states, with  $n$  equal to 60 (circles), 70 (squares) and 85 (stars). The TiSa laser intensity is given by  $(n/85)^3 \times 560 \text{ W/cm}^2$ . Solid lines are given by the reduced density matrix model. (b) Data for  $n=60$  and  $n=85$  [same symbols as (a)], characterized by  $\tan^2 \theta$  (see text), the theoretical curve (dashed line) is obtained without the DB.

We observe a more efficient DB effect for  $n = 85$  than for  $n = 60$  when compared with the calculated (dashed) curve without the dipole-dipole interaction.

To model our data, we have developed a model based on the interaction of each atom  $i$ , with its Rydberg neighbors  $j$  (more details can be found in [14]). If atom  $i$  could be excited to the Rydberg state its energy would be shifted by an amount of

$$W_{ij} = \frac{\vec{\mu}_i \cdot \vec{\mu}_j - (\vec{\mu}_i \cdot \vec{n}_{ij})(\vec{\mu}_j \cdot \vec{n}_{ij})}{4\pi\epsilon_0 R_{ij}^3}$$

where  $\vec{n}_{ij} = \vec{R}_{ij}/R_{ij}$  with  $R_{ij}$  the interatomic distance between the Rydberg atoms  $i$  and  $j$ . Due to the  $1 - 3\cos^2 \psi$  variation of the  $W_{ij}$ , where  $\psi$  is the angle between the internuclear axis and the dipoles (i.e. the external field), the angular averaging of  $W_{ij}$  is zero. Therefore, we need to discretized the averaging. We found by the Monte Carlo simulation that the nearest neighbor interaction dominates. We then model the dipole-dipole interaction using only the nearest neighbor atom  $j$  and neglect the mean-field interaction. This is the opposite in the van de Waals interaction where we neglect the nearest neighbor interaction.  $W_{ij}$  is significant only when both atoms are in the Rydberg state. We find

$$Tr_{\neq i} [W_{ij}, \rho] \simeq \frac{\mu^2 n_{Ry}}{4\pi\epsilon_0} (\vec{r}_i) [|np\rangle_{ii} \langle np|, \rho_i]$$

which corresponds for the atom  $i$  to a shift of the Rydberg level  $np$  proportional to the local Rydberg density  $n_{Ry}(\vec{r}_i)$ . Here  $\rho$  is the density matrix for the ensemble of the atoms, and  $\rho_i = Tr_{\neq i} \rho$  is the reduced density matrix for the single atom  $i$ . For a given atom  $i$ , we solve the evolution equation of the partial density matrix

$$i\hbar \frac{d\rho_i}{dt} = [H_i, \rho_i] + Tr_{\neq i} [W_{ij}, \rho]$$

$H_i$  is the (Bloch eq. type) Hamiltonian describing the 3-step excitation. The spontaneous emission relaxation terms are taken into account but not written here for sake of simplicity.

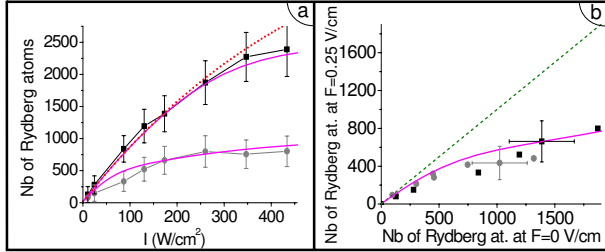


FIG. 4: (Color online) Number of Rydberg atoms excited versus the Ti:Sa laser intensity, in the case of the  $70p_{3/2}$  state, for (a)  $7s$ -atom density  $D \sim 4 \pm 2 \times 10^9 \text{ cm}^{-3}$  and in the presence of two different electric fields, 0 V/cm (squares) and  $F_1 = 0.25$  V/cm (circles); (b) Number of Rydberg atoms excited in the presence of the electric field  $F_1$  is plotted versus the number of Rydberg atoms excited at zero field, for two  $6p$   $7s$ -atom densities,  $D$  (squares) and  $D/2.7$  (circles). The DB efficiency is the ratio between by the gap between the experimental points and the straight dashed line of slope 1. In (a) and (b): solid lines are theoretical calculations taking into account the van der Waals blockade at zero field and the DB in the presence of the electric field  $F_1$ . In (a), for comparison, we have also presented the curve with no van der Waals coupling (dotted curve).

Fig. 3 (a) shows the comparison between the calculated curves and the experimental ones with a good agreement. All curves in this letter, are calculated using the exact dipole moments calculated by considering the ensemble of the levels of the Stark diagram [15]. However, most of the results can be understood using a simpler two-level approach, valid for small electric fields ( $F \ll 1/3n^5$ , atomic units), assuming only mixing of the  $np$  state with the  $(n-1)d$  one. In this model the  $np$  Rydberg permanent dipole is  $\mu \sim q_e a_0 n^2 \sin \theta$  and the excitation (dashed) curve of Fig. 3 (b) would have a value of  $\cos^2(\theta/2)$ . The DB condition:  $W_{ij} \sim \frac{\mu^2}{4\pi\epsilon_0 R_{ij}^3} < h\Delta_L$  can then be simply evaluated. This DB condition gives a limitation to the excitation corresponding to a density

$$n_{Ry} \sim h\Delta_L 4\pi\epsilon_0 / \mu^2.$$

Such a limitation is illustrated by Fig. 4(a), showing the evolution of the DB versus the Ti:Sa laser intensity in the case of the  $70p_{3/2}$  Rydberg level, where the DB condition gives  $N \sim 800$  Rydberg atoms and fits well to the experiment ( $\sigma \sim 50 \mu\text{m}$ ). At the electric field  $F_1 = 0.25$  V/cm (corresponding to  $\tan^2 \theta = 1$ ) the appearance of the DB occurs at intensities higher than  $100 \text{ W/cm}^2$ . The DB efficiency is evaluated by comparison with the zero field case. We observe a power saturation for intensities larger than  $250 \text{ W/cm}^2$  explaining why the maximum

DB efficiency is reached when the intensity is close to  $250 \text{ W/cm}^2$ . For higher intensity an optical broadening occurs so  $\Delta_L$  increases and the DB efficiency decreases. For the zero field case, taking into account the van der Waals coupling between the atoms modifies the theoretical curve, giving a better agreement with the experimental data. The DB efficiency, evaluated by comparison with the zero field case, is also presented in Fig.4(b) from two data sets taken at two different  $6p$  (and so two different  $7s$ )-atomic densities. The DB appears not to be dependent on the  $6p$ ,  $7s$  densities but only on the number of excited Rydberg atoms. This result is intrinsic to the DB and cannot be due to an ion blockade effect coming from Rydberg atoms ionization created by collisions with  $6p$  or  $7s$  atoms.

To conclude, we have presented the evidence for an efficient DB controlled via the Stark effect, of the Rydberg excitation in a cold cesium atomic sample. The observation of the DB is challenging because no ions should be present during the short excitation pulse. An analytical model based on the preferential role of the nearest neighboring Rydberg atoms, confirmed by M.C. simulations, has been derived and provides a good account of the data. This result is really promising for quantum information. A microwave field instead of Stark effect or Förster may be used to produce the dipole moments and to control the DB [16, 17]. A particularly promising direction of research is now the observation of the total DB, meaning the collective excitation of a single atom. Prospect for quantum gate devices and control of ultra-cold Rydberg atoms in this quantum regime is still a challenge.

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