Controllable Interactions between Rydberg Atoms and Ultracold Plasmas

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Abstract. We discuss the control of dipole-dipole interactions in a frozen assembly of Rydberg atoms. We report the evidence of dipole blockade of the Rydberg excitation for two configurations: dipole blockade induced by electric field and dipole blockade in Förster resonance. We demonstrate that two individual atoms separated by ~ 4 µm can act as a collective dipole if their interaction is strong enough to be in the dipole blockade regime. This observation is crucial for the quantum entanglement of two or more atoms using dipole-dipole interaction. The dipole-dipole interactions between Rydberg atoms are also responsible for Penning ionization leading to the formation of an ultracold plasma. We have demonstrated that Penning ionization of np Rydberg cesium atoms can be prevented by considering repulsive dipole-dipole interactions.

1. Introduction

The atomic highly excited or Rydberg states were extensively studied in the eighties for their exaggerated properties [1]. Indeed they are so sensitive to weak perturbations that they offer the opportunity to explore phenomena inaccessible in other physical systems. Rydberg atoms are known to have huge polarizabilities leading to extreme collisional properties of room temperature atoms, in particular, large cross sections and long interaction times. The evidence of collisions with impact parameters of the order of the micrometer has for instance been demonstrated at room temperature [2]. The first experiments performed with cold Rydberg atoms demonstrated not only the long-range of the interatomic interactions, but their many-body nature as well [3,4]. On the time scale of the experiment, on the order of one microsecond, the behavior of the Rydberg ensemble corresponds to a frozen gas with more similarity to an amorphous solid than to a gas of colliding atoms. Moreover for longer times of a few microseconds, dense and cold Rydberg atomic samples have shown a very impressive behavior, their spontaneous evolution to a neutral ultracold plasma [5]. The ensemble of these first studies have clearly demonstrated that the physics of the cold Rydberg atoms is at the intersection of...
atomic physics, solid state physics, and plasma physics. If the initial conditions of the Rydberg ensemble fix its evolution and its behavior, the interactions between Rydberg atoms are the crucial elementary processes. To control the dipole-dipole interactions is therefore the key to control of the evolution of such ensembles. The possibility of controlling the strong long-range interactions between cold atoms has also been pointed out as being particularly exciting for quantum information applications [6,7]. One interesting process is the possibility of dipole blockade in the Rydberg excitation of atoms, due to the dipole-dipole interaction shifting the Rydberg energy from its isolated atomic value.

The paper is organized as following. In the section 2, we discuss the Rydberg-Rydberg interactions and more precisely the dipole-dipole ones in a frozen assembly of atoms [8]. The case of the Förster resonance is described carefully. In the section 3, we expose a first consequence of the dipole-dipole interaction, the dipole blockade of the Rydberg excitation of cold atoms. We discuss two examples: the dipole blockade induced by a static electric field [9], and the dipole blockade in Förster resonance [10], for which the collective excitation of a pair of individual atoms has also been demonstrated in the blockade regime [11]. In the section 4, we show the limit of the picture of the frozen Rydberg gas, with the role of the forces exerted between two Rydberg atoms due to the dipole-dipole interactions. We demonstrate that it is possible to control the attractive or repulsive character of these forces, for preventing the Penning ionization processes [12]. These effects are particularly important to control the evolution of cold Rydberg gases toward ultracold plasmas [13,14]. The section 5 is the conclusion.

2. Rydberg-Rydberg interactions

2.1. Van der Waals interaction between Rydberg atoms

Because of their large polarizabilities, Rydberg atoms are expected to be very sensitive to collisions. In zero electric field, Rydberg atoms present no permanent electric dipole momentum. For low angular momentum states, the \( C_6/R^6 \) van der Waals interaction is expected to dominate at long-range distances, \( R \), between two \( nl \) Rydberg atoms. The \( C_6 \) van der Waals coefficient is roughly given by

\[
C_6 \approx \sum_{n-n'=0,1,1} \sum_{l=l',1} \frac{|n|H_{dd}|n'l'|^2}{\Delta W_{nl,n'l'}},
\]

with \( H_{dd} = \frac{\mu_1 \mu_2}{R^3} - 2 \left( \frac{\mu_1 R}{R^3} \right) \left( \frac{\mu_2 R}{R^3} \right) \). \( H_{dd} \) is the long-range dipole-dipole coupling between \( nl \) and \( n'l' \) Rydberg states (\( l'=l \pm 1 \)), and \( \Delta W_{nl,n'l'} \) their energy difference. The dipole-dipole interaction between two atoms labeled 1 and 2 is written in atomic units (a.u.). \( \mu_{1,2} \) corresponds for each atom, to the electric dipole transition matrix element between the states \( nl \) and \( n'l' \). For \( n \sim n' \), these latter acquire huge values, \( \sim n^2 \) a.u., where the atomic unit of electric dipole moment is \( ea_0 = 8.48 \times 10^{-30} \) C.m = 2.54 Debye (\( e \) is the charge of the electron). The dipole matrix elements reach values of a few thousand Debye for \( n \sim 30 \), to compare to values of a few Debye for electric dipole moments of even the most polar molecules. As \( \Delta W_{nl,n'l'} \) scales as \( n^3 \), the \( C_6 \) coefficient should present a rapid \( n^{11} \) scaling. However for configurations, where \( \Delta W_{nl,n'l'} \) is small enough, the van der Waals behavior of the potential is modified. A dipole-dipole interaction, \( C_3/R^3 \sim H_{dd} \), is then expected to dominate at long-range distances, \( R \), which corresponds for instance to the case of the Förster resonance.

2.2. Dipole-dipole interactions induced by a static electric field and at Förster resonance

In the presence of an electric field, Rydberg atoms can have a significant permanent dipole moment and higher \( n \) excitation leads to higher dipole-dipole couplings. Adding a small electric field is therefore a very simple way to control the dipole-dipole interaction. Another way to control the dipole-dipole coupling through Stark effect is the case of the resonant dipole-dipole energy transfer [2-4], so-called Förster resonance. We consider for instance the case of the cesium atom (the other alkali atoms present similar properties), for which the \( np_{3/2} \) state is located nearly midway in energy between the
two neighboring s states, $ns$ and $(n+1)s$ (labeled $s$ and $s'$). For $n < 42$, exact resonance can be reached by adding a small electric field shifting the $p$ states such that the collisional reaction

$$np_{3/2} + np_{3/2} \rightarrow ns + (n+1)s$$

occurs and leads to an important transfer of population to the $s$ states by exchange of internal energy between the two Rydberg atoms. The efficiency of the transfer is due to the long-range interaction of the dipole-dipole interaction, $H_{dd}$. $\mu_1$ ($\mu_2$ respectively) corresponds here to the electric dipole transition matrix element between the states $np$ and $ns$ ($np$ and $(n+1)s$ resp.). The order of magnitude of the coupling is given by $\frac{C_3}{R^3} \sim \frac{n^4}{R^3}$ a.u., which corresponds to several gigahertz for $n \sim 60$ and $R \sim 1 \mu m$. When the two atoms are in state $|np, np\rangle$, they interact strongly which leads to symmetric energy shifts $\Delta E = \pm \frac{C_3}{R^3}$, corresponding to the symmetric and antisymmetric state $\left(|np, np\rangle \pm |ns, ns\rangle\right)/\sqrt{2}$.

3. Dipole blockade of the Rydberg laser excitation

3.1. Principle of the dipole blockade

The use of the dipole blockade of the excitation has been proposed as a very efficient realization of a scalable quantum logic gate [7]. The principle of the dipole blockade of the laser excitation of cold Rydberg atoms is described on the figure 1 for the case of two atoms for a Förster resonance. The ground state $|g\rangle$ and the Rydberg state $|r\rangle$ of each atom are coupled by a laser with a Rabi frequency $\Omega$. When one atom is excited in a Rydberg state, the dipole-dipole interaction shifts the resonance and prevent the laser excitation of the second one (figure 1(a) conditional excitation).

![Figure 1. (a) Principle of the Rydberg blockade between two atoms, in the regime of conditional excitation. (b) When both atoms are simultaneously excited in the blockade regime, the symmetrical state $|\Psi_+\rangle$, described in the text, is only coupled to the ground state $|g,g\rangle$ with a strength $\sqrt{2}\Omega$ while the state $|\Psi_-\rangle$ is not coupled by the laser to the states $|g,g\rangle$ and $|r,r\rangle$.](image)

More precisely, when the two atoms are in state $|r,r\rangle$, they interact strongly which leads to symmetric energy shifts $\Delta E = \pm \frac{C_3}{R^3}$ (see previous paragraph). When the interaction energy becomes larger than both the Rabi frequency and the laser resolution, the laser is out of resonance with the transition coupling the singly with doubly excited state, and only one atom at a time can be transferred to the Rydberg state. When both atoms are simultaneously excited, a fundamental consequence of the blockade is that any of the two atoms can carry the excitation. They end up in the entangled state $\Psi_+ = \frac{1}{\sqrt{2}}(|gr\rangle e^{ikr} + |rg\rangle e^{iks})$, where $r_a$ and $r_b$ are the position of the two atoms, and $k$ the wavevector of the excitation. The coupling from the two-atom state $|g,g\rangle$ toward the state $|\Psi_+\rangle$ is
While the state $|\Psi_+\rangle = \frac{1}{\sqrt{2}} \left( |g, r\rangle e^{i\delta_x} - |g, r\rangle e^{i\delta_y} \right)$ is not coupled with the ground state. In the blockade regime, the two atoms are therefore described by an effective two-level system involving collective states $|g, g\rangle$ toward $|\Psi_+\rangle$.

The dipole blockade effect can be generalized to an assembly of N atoms. If the volume of the laser excitation is small enough, no second atom can be excited after the Rydberg excitation of a first one, producing an atomic ensemble in a singly excited collective state. If we consider the high-resolution laser excitation of a large ensemble of atoms from the ground state or from a low excited state towards a Rydberg state, the dipole-dipole interaction between Rydberg atoms should lead to a limitation of the number of excited atoms versus the initial density of atoms. A partial, or local, blockade of the excitation is thus expected. The first excited Rydberg atoms shift the resonance for their non-excited neighbors and prevent their excitation with a narrow-bandwidth laser.

### 3.2. Dipole blockade induced by electric field

The first observation of a blockade regime of the Rydberg laser $np$ excitation of an assembly of cold Rb atoms is due to the van der Waals interaction between Rydberg atoms [15]. In order to obtain higher couplings, an electric field can be applied to create a significant dipole moment for each atom. The figure 2 of the reference [9] shows the evidence for the dipole blockade induced by electric field. It constitutes the first observation of the dipole blockade effect.

![Figure 2.](image)

(a) Excitation of the spectral lines of the 75$p_{3/2}$ level for different electric fields. The shift of the lines is due to the Stark effect. (b) Comparison of spectral lines of the 70 $p_{3/2}$ for two different electric fields. Number of detected spurious ions is also shown (inset, log scale).

The Rydberg atoms are excited from a cloud produced in a cesium vapour loaded magneto-optical trap. The laser excitation is a three-step one, $6s \rightarrow 6p_{3/2} \rightarrow 7s \rightarrow np_{3/2}$, provided by three cw resonant lasers simultaneously applied during a time of 300 ns with a repetition rate of 80 Hz. The dipole blockade controlled through Stark effect presents an efficiency of 60% limited by the resolution of the excitation ($\sim 5$ MHz). One difficulty of the experiment is the eventual presence of spurious ions, which can be responsible for a Coulomb blockade very similar to the dipole one. We have verified that no ion (less than one every 50 shots) was present during the laser excitation.

A simple analytical model of the excitation including the dipole-dipole interaction of each atom with its closest Rydberg neighbor gives a good agreement with the dipole blockade data. This preferential role of the closest Rydberg neighbor is confirmed by Monte Carlo simulations [16].

### 3.3. Dipole blockade in Förster resonance

The dipole blockade can also be observed at a Förster resonance as $np_{3/2} + np_{3/2} \rightarrow ns + (n+1)s$ (see figure 3) [10]. The experiment is similar to the previous one, except that the electric field value necessary to reach the Förster resonance are weak. The efficiency of the process is characterized by the
minimum of the excitation in figure 3a. It is limited to 30%, essentially due to the relatively low \( n \) considered \((n < 42)\). We notice that the excitation at the Förster resonance corresponds to an efficient transfer towards the levels \( ns \) an \((n+1)s \) levels (figure 2b). We detect (figure 3c) the presence of a few ions mostly produced after the laser excitation. This ion background is mostly due to blackbody ionization. It is interesting to notice that the number of ions is greater for higher electric field than the field at the Förster resonance. We interpret this result as a Penning ionization process because of the attractive force due to dipole-dipole interaction. A clear evidence of such effect will be exposed in section 4.

### 3.4. Collective excitation of two individual trapped atoms in the blockade regime

The dipole blockade of the laser excitation has been observed for a pair of individually trapped atoms [11,17]. Moreover the collective two-atom behavior has been experimentally demonstrated with the excitation of an entangled state between the ground and Rydberg levels [11]. In this experiment, two individual Rubidium 87 atoms are excited to the Rydberg state \( |r\rangle = |58d_{3/2}\rangle \) in a two-photon process. Both atoms are simultaneously illuminated by the excitation laser beams \((< 500 \text{ ns})\). The experimental details are given in the reference [11]. The state \( 58d_{3/2} \) is chosen because of the existence of a quasi-resonance of Förster from the near energy degeneracy between the two-two atom states \((58d_{3/2}, 58d_{3/2})\) and \((60p_{3/2}, 56f_{5/2})\). The two atoms are confined in two independent optical dipole traps, which are turned off during the excitation to avoid an extra light-shift [18]. A successful excitation of an atom to the Rydberg state is detected through the loss of the atom when the dipole trap is turned back on, as atoms in the Rydberg state are not trapped in the tweezers.

![Figure 3. Dipole blockade at Förster resonance for the 38p3/2 level versus the applied electric field. (a) Total number of Rydberg atoms, (b) number of 38s atoms, and (c) number of formed ions.](image)

![Figure 4. Collective excitation of the two atoms, labeled a and b, separated by 3.6 \( \mu m \) (condition of blockade regime). The circles represent the probability to excite one atom a when the atom b is absent. A fit to the data yields a Rabi oscillation frequency of \( \Omega/2\pi = 7.0 \pm 0.2 \text{ MHz} \). The squares represent the probability to excite only one atom when the two atoms are trapped and are exposed to the same excitation pulse.](image)
The distance between the two traps can be varied between 3 and 20 µm. The energy interaction between the two atoms due to the dipole-dipole coupling is 50 MHz for an interatomic distance of 4 µm. The figure 4 shows the result of two experiments, where we apply the Rydberg excitation either to a single atom or to two neighboring atoms at a distance of 3.6 µm. The probability to excite both atoms in the Rydberg state is suppressed, as it is expected in the blockade regime. In the figure 4, the probability to excite only one of the two atoms as a function of the duration of the excitation, can be compared with the probability to excite one atom inside a given trap when the other trap is empty. The two probabilities oscillate with different frequencies, whose the ratio of which 1.38 ± 0.03 is close to the expected value of $\sqrt{2}$, which is the signature of the collective behavior of the two atoms. This result will be used to create the entanglement of two atoms in two hyperfine ground states, by laser inducing the decay of the Rydberg state.

4. Control of Penning ionization through attractive or repulsive potentials

A cold Rydberg gas can spontaneously evolve into a plasma. If there is even a very slow ionization process, cold ions are produced, and at some point their macroscopic space charge traps all subsequently produced electrons. The trapped electrons lead to a collisional avalanche which rapidly redistributes the population initially put into a single Rydberg state. Typically two-thirds at least of the atoms are ionized, while the other atoms are driven to lower states to provide the requisite energy [5,13,14]. The origin of the initial ions can be due to different processes such as for instance blackbody radiation. One other mechanism for this ionization is that pairs of atoms excited to attractive diatomic potential curves collide resulting in the ionization of one the atoms, the second atom being driven to a lower state (Penning ionization process). In an effort to isolate the effect of attractive and repulsive potentials from other effects, we have examined the ionization of cold Cs $np$ atoms excited with narrow-bandwidth excitation. The interesting features of the Cs $np$ states are shown in figure 5.

![Figure 5](image-url)

**Figure 5.** (a) Energy difference at zero electric field, between a pair of atoms in state $np_{n+1}s$ ($ss'$) for different $n$. The curve crosses zero at $n \sim 42$. (b) and (c) Schematic potential curves of a pair of atoms as a function of internuclear distance. For $n > 42$ the $pp$ state lies below the $ss'$ one, while for $n < 42$ the reverse is true. We excite the $np3/2$ state, or pairs of atoms to the molecular $pp$ state. The repulsive potential is excited for $n > 42$, the attractive potential for $n < 42$. For $n = 42$, both potentials are excited.

We consider again the experimental approach in the Cs vapour-loaded MOT. The Cs Rydberg atoms $np_{3/2}$ are now excited in zero electric field. The cold Rydberg gas is allowed to evolve from 450 ns to 50 µs. At a given evolution time, the population is analyzed with a field ionization. Both time-resolved ion and Rydberg atom signals are recorded. The figures 6(a) and 6(b) of the reference [12], shows typical recordings after an evolution time of 10 µs, obtained by scanning the frequency of the...
laser excitation \((7snp3/2)\), for \(n = 40\) and \(43\), respectively, and for two different laser powers corresponding to two different densities of Rydberg atoms. The results show unambiguous difference between repulsive \((n = 40)\) and attractive \((n = 43)\) potentials. At low power (low density), we observe in both cases a small number of ions. At high power (higher density), for \(n = 40\) the number of ions is still small, but at \(n = 43\) we observe at resonance the complete ionization of the Rydberg sample, which is the signature of the formation of an ultracold plasma.

![Figure 6](image)

**Figure 6.** Rydberg and Ion signal with two different laser intensity (42mW and 278mW) for (a) 40p state (repulsive) and (b) 43p (attractive) Data are taken after 10μs of free evolution.

![Figure 7](image)

**Figure 7.** Ion density as function of initial Rydberg density for different \(n\) states and three delay time (a) 0.45 (b) 5 and (c) 10μs. at 10μs the "40p BB" is without 6p cold and hot atoms.

The figure 7 shows the density of ions versus the density of Rydberg atoms initially excited, for 0.45, 5 and 10 μs delay times. For 0.45 μs delay the ionization yields are essentially the same, but there is clear difference for 5 and 10 μs delays, between \(n < 42\) and \(n > 42\) due to the dipole-dipole-induced collisions in the case of the attractive potential. For \(n < 42\), the ionization is mainly due to blackbody radiation. For the 10 μs delay the ion production starts to become very non linear, which is
attributed to the trapping of the electrons by the ions and the subsequent ionization collisions with the Rydberg atoms.

5. Conclusion
The reported results underscore the notion that the dipole-dipole interaction is the dominant feature of the cold Rydberg gas. They demonstrate the role of the dipole-dipole coupling and more precisely of the Förster resonances, for the control of the excitation and of the evolution of an ensemble of a frozen Rydberg atomic gas. The Rydberg dipole blockade offers an efficient quantum engineering for the entanglement of pairs of atoms at a macroscopic distance and for the realization of quantum gates. The control of the dipole-dipole forces exerted between two Rydberg atoms opens interesting prospects for the control of the ionization in a cold Rydberg gas. Several directions could be explored: the Rydberg atomic ensemble as a quantum simulators and the preparation of correlated plasmas.

Acknowledgments
This work is performed in the frame of the Institut Francilien pour la Recherche sur les Atomes Froids (IFRAF), of the Réseau Thématique de Recherche Avancée “Triangle de la Physique”, and of the CORYMOL experiment supported by an ANR grant (N) NT05-2 41884).

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