Cold Rydberg gases

1. Basics of Rydberg atoms
2. Rydberg atoms in external fields
3. Rydberg-Rydberg interaction

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Outline – Rydberg-Rydberg interaction

• Long-range dipole – dipole interaction between Rydberg atoms.

• Some current research topics with interacting Rydberg atoms/ensembles
Long-range dipole –dipole interaction between Rydberg atoms
Interaction of two classical static dipoles

$$V_{dd} = \frac{\mu_1 \cdot \mu_2}{R^3} - \frac{3(\mu_1 \cdot \vec{R})(\mu_2 \cdot \vec{R})}{R^5}$$

Some special cases:

Attractive

\[ \uparrow \uparrow \uparrow \]

Repulsive

\[ \uparrow \uparrow \uparrow \]
Permanent Dipole of Rydberg Stark states in E-field

Permanent Dipoles in E fields

The resulting of linear Stark effects have permanent electric dipoles: \( \langle \psi_\pm | \vec{r} | \psi_\pm \rangle \neq 0 \) while \( \psi_\pm \) are superposition of different \( l \) states.

\[ \Rightarrow \text{Resonant dipole-dipole (almost like classical interaction between static dipoles)} \]
Interaction of two synchronously oscillating classical dipoles

\[ \vec{\mu}_1 = \hat{z} \mu_1 \cos \omega t \]

\[ \vec{\mu}_2 = \hat{z} \mu_2 \cos \omega t \]

\[ V_{dd} = \frac{\mu_1 \cdot \mu_2}{2R^3} - \frac{3(\mu_1 \cdot \vec{R})(\mu_2 \cdot \vec{R})}{2R^5} \]

The interaction is essentially the same but reduced by a factor of two due to the time averaging of the dipoles.

NOTE We always assume that we are in the near field regime.

\[ R < \lambda = \frac{2\pi c}{\omega} \]

and we ignore the radiation field.
Dipole – dipole interaction of two Rydberg atoms

The quantum version of oscillating dipoles is the interaction between transition dipole moments.

One example

\[ \langle ns, np | V_{dd} | np, ns \rangle \neq 0 \]

\[ \langle ns, np | \frac{\mu_1 \mu_2}{R^3} | np, ns \rangle = \frac{\langle ns | \mu_1 | np \rangle \langle np | \mu_2 | ns \rangle}{R^3} \neq 0 \]

and \[ E(|ns, np\rangle) = E(|np, ns\rangle) \]

\[ E_\pm = \pm \frac{\mu_1 \mu_2}{R^3} \]

States of two atoms
(Pair states or Diatomic states)

\[ |ns, np\rangle \quad \text{or} \quad |np, ns\rangle \]

Pair states

\[ |nsnp\rangle \quad |npns\rangle \]
Another example

Atomic states

\[ \langle ns, ns \mid V_{dd} \mid np, (n - 1)p \rangle \neq 0 \]

\[
\left\langle ns, ns \left| \frac{\mu_1 \mu_2}{R^3} \right| np, (n - 1)p \right\rangle = \frac{\langle ns, np \rangle |np, (n - 1)p \rangle}{R^3} \neq 0
\]

\[
E(|ns, ns\rangle) - E(|np, (n - 1)p \rangle) = \Delta
\]

Pair states

\[ H = \begin{pmatrix}
0 & \frac{\mu_1 \mu_2}{R^3} \\
\frac{\mu_1 \mu_2}{R^3} & -\Delta
\end{pmatrix} \]

\[ V_{dd} = \frac{\mu_1 \mu_2}{R^3} \propto n^4, \]

\[ \Delta \sim 1 / n^3 \]

|ns, ns\rangle or |np, (n - 1)p\rangle or |(n - 1)p, np\rangle
Resonance d-d interaction vs Van der Waals interaction

- resonance dipole-dipole interactions
- van der Waals (off-resonance d-d)

20 S + 20 S → 19 P + 20 P

Safinya et al., PRL 47, 2405 (1981)

Huge interaction, ~ GHz (n ~ 40, R ~ 1 μm)
Easily tuned by: external E field (Δ), n (V_{dd}, Δ), or R (V_{dd})
Resonance d-d interaction vs Van der Waals interaction

- resonance dipole-dipole interactions
- van der Waals (off-resonance d-d)

Hamiltonian

\[
H = \begin{pmatrix}
|20s20s\rangle & |19p20p\rangle \\
0 & \frac{\mu_1\mu_2}{R^3} \\
\frac{\mu_1\mu_2}{R^3} & \Delta
\end{pmatrix}
\]

\[V_{dd} = \frac{\mu_1\mu_2}{R^3} \propto n^4,\]
\[\Delta \sim 1 / n^3\]

Potential Curves

If \(V_{dd} \ll \Delta\) (at large \(R\))

\[W = -\frac{V_{dd}^2}{\Delta}, \Delta + \frac{V_{dd}^2}{\Delta}\]
\[\Delta W = \pm \frac{V_{dd}^2}{\Delta} \sim \frac{n^{11}}{R^6}\]

So van der Waals interaction,

\[
\frac{C_6}{R^6}
\]

Where \(C_6 \propto n^{11}\)

If \(V_{dd} \gg \Delta\) (at small \(R\))

\[W = \pm V_{dd} \sim \frac{n^4}{R^3}\]

Dipole-dipole interaction
Tuning the dipole-dipole interaction with E-field

\[ 2 E_{np}(F_0) = E_{ns}(F_0) + E_{(n+1)s}(F_0) \]

**Energy Diagram**

- \( E_{np}(F) \)
- \( E_{ns}(F) \)
- \( E_{(n+1)s}(F) \)
- \( F_0 \)

**Energy Formulas**

- \[ E_{np}(F) = \frac{\sqrt{2} \mu_1 \mu_2}{R^3} \Delta(F) \]
- \[ E_{ns}(F) = \frac{\sqrt{2} \mu_1 \mu_2}{R^3} \Delta(F) \]
- \[ V_{dd} = \frac{\sqrt{2} \mu_1 \mu_2}{R^3} \propto n^4 \]

**States**

- \( |+\rangle = \frac{|pp\rangle + |ss'\rangle}{\sqrt{2}} \)
- \( |-\rangle = \frac{|pp\rangle - |ss'\rangle}{\sqrt{2}} \)

**Energy Expression**

- \[ E_\pm = \frac{\Delta \pm \sqrt{\Delta^2 + 4V_{dd}^2}}{2} \]
Some details of dipole-dipole interaction

Consider the nsnp npns pair states, which are coupled by the dipole-dipole coupling

$$\langle n_{\text{sn}}, n_{\text{p}} | V_{dd} | n_{\text{p}}, n_{\text{s}} \rangle =$$

$$E(|n_{\text{ns}}, n_{\text{p}}\rangle) = E(|n_{\text{p}}, n_{\text{s}}\rangle)$$

$$\langle n_{\text{sn}}, n_{\text{p}} | V_{dd} | n_{\text{pns}} \rangle = \langle n_{\text{sn}}, n_{\text{p}} | \frac{\vec{\mu}_1 \cdot \vec{\mu}_2 - 3(\vec{\mu}_1 \cdot \vec{R})(\vec{\mu}_2 \cdot \vec{R})}{R^3} \rangle |n_{\text{pns}}\rangle$$

There are two natural choices for an axis of quantization

- The internuclear axis
- The direction suggested by a field, which might produce the dipoles, messy but possible.
The internuclear axis as the quantization axis

The nsnp npns pair states with the internuclear axis chosen as the quantization axis, in the z direction.

The dipole dipole matrix element becomes

\[
\langle \text{nsnp} | V_{dd} | \text{npns} \rangle = \langle \text{nsnp} | \frac{x_1 x_2 + y_1 y_2 - 2 z_1 z_2}{R^3} | \text{npns} \rangle \\
\langle \text{nsnp} | V_{dd} | \text{npns} \rangle = \langle \text{ns} | \langle \text{np} | V_{dd} | \text{np} \rangle | \text{ns} \rangle \\
= \frac{\langle \text{ns} | x | \text{np} \rangle \langle \text{np} | x | \text{ns} \rangle + \langle \text{ns} | y | \text{np} \rangle \langle \text{np} | y | \text{ns} \rangle - 2 \langle \text{ns} | z | \text{np} \rangle \langle \text{np} | z | \text{ns} \rangle}{R^3}
\]

Angular momentum is conserved, so we construct states of total angular momentum \( M \) along the quantization axis using

\[
M = m_1 + m_2
\]
Using basis states of the form \( n_{snp} \) there are six states, two each of \( M=0, 1, \) and \(-1\).

For example the \( M=0 \) states are \( n_{snp_0} \) and \( n_{p_0 ns} \), with the Hamiltonian matrix

\[
H = \begin{pmatrix}
0 & V_{dd} \\
V_{dd} & 0
\end{pmatrix}
\]

Where the matrix element is given explicitly by

\[
V_{dd} = -\frac{2\langle ns|z|np\rangle\langle np|z|ns\rangle}{R^3}
\]

The two \( M=0 \) eigenvalues and eigenstates are

\[
W = \pm V_{dd} = \mp \frac{2\mu}{R^3}
\]

\[
|+\rangle = \frac{|nsnp\rangle + |npns\rangle}{\sqrt{2}}, \quad W(|+\rangle) = +V_{dd} = -\frac{2\mu}{R^3}
\]

\[
|-\rangle = \frac{|nsnp\rangle - |npns\rangle}{\sqrt{2}}, \quad W(|-\rangle) = -V_{dd} = +\frac{2\mu}{R^3}
\]

One of the eigenstates is symmetric in the interchange of the two atoms, and one is antisymmetric,
There are analogous 2x2 matrices for the M=1 and M=-1 states, which also give symmetric and antisymmetric states.

Solid lines are M=0 states.
Broken lines are M=1 and -1 states.
There are two M=1 and two M=-1 states.
Return to the case of the nsns np(n-1)p pairs M=0 for example

\[ \Delta \text{ nsns np(n-1)p (n-1)pnp} \]

The Hamiltonian matrix of the three M=0 states where

\[
H = \begin{pmatrix}
0 & V_{dd} & V_{dd} \\
V_{dd} & \Delta & 0 \\
V_{dd} & 0 & \Delta \\
\end{pmatrix}
\]

The three eigen energies are

\[
W = \Delta \\
W = \frac{\Delta \pm \sqrt{\Delta^2 + 4V_{dd}^2}}{2}
\]

\[
V_{dd} = -\frac{2\langle ns | z | np \rangle \langle np | z | ns \rangle}{R^3}
\]
The energy levels

\[ \frac{1}{R^3} \quad \frac{1}{R^6} \]

There is a state with no shift—degeneracy of the p pairs

The shift changes from \( \frac{1}{R^6} \) to \( \frac{1}{R^3} \) when \( V \sim \Delta \), from a van der Waals to a dipole-dipole interaction.
Fine, but quite real details

The spin orbit splittings must be taken into account

Often only one spin orbit coupled state is important. In Rb and Cs $np_{3/2}np_{3/2}$ is resonant with $ns(n+1)s$ but $np_{1/2}np_{1/2}$ is not.

There are smaller matrix elements.
There are more states.

The Zeeman degeneracy leads to states with no shift.
-Kiffner, Walker and Saffman

We have already seen an example of this.
Calculated van der Waals shifts from Walker and Saffman (2007)

Note these are calculated with the internuclear axis as the quantization axis.
Fig. 1. Rydberg atom interaction potentials calculated as described in the text for Cs and Rb Rydberg atom pairs around the 37D + 37D asymptote. The calculation is done for the case of no background electric field. From Cabral et al. (2011).
Dipole Blockade and Collective Excitation

Blockade: a deterministic way to make entanglement

\[ |\psi_+\rangle = \frac{1}{\sqrt{2}} (|g,r\rangle + |r,g\rangle) \]

Rabi Oscillation

Interatomic Spacing (\(\mu m\))

Energy (GHz)

\(n \approx 40\)
Dipole blockade in mesoscopic ensembles (principle)

Only one Rydberg atom can be excited inside the blockade sphere.

\[ V = C_6 / R_{\text{max}}^6 \sim \Delta \nu_{\text{laser}} \]

Blockade radius up to tens of microns for our parameters.
Dipole blockade in mesoscopic ensembles (possible applications)

If the radius is smaller than the blockade radius, the only accessible state is the symmetric Dicke state with 1 Rydberg excitation.

Generalization of the 2 atoms case to $N$ atoms

Collective excitation with Rabi frequency:

$$\Omega_{\text{coll}} = \sqrt{N}\Omega$$

$$|\Psi_{\text{coll}}\rangle = \frac{1}{\sqrt{N}} (e^{ikr_1}|rgg...g\rangle + e^{ikr_2}|grg...g\rangle + \ldots + e^{ikr_N}|ggg...r\rangle) \equiv |r^{\{1\}}g^{\{N-1\}}\rangle$$

All the $N$-1 other states with one excitation are uncoupled to ground state because of symmetry properties
Fast Quantum Gate


- two q-bit states, $|0\rangle$ and $|1\rangle$
- coupled differently to Rydberg state $|r\rangle$
- two atoms individually addressible

All pulses: $|0\rangle \rightarrow |r\rangle$

No excitation Due to blockade

Phase Gate
- $|1,1\rangle \rightarrow |1,1\rangle$
- $|0,1\rangle \rightarrow -|0,1\rangle$
- $|1,0\rangle \rightarrow -|1,0\rangle$
- $|0,0\rangle \rightarrow -|0,0\rangle$
Frozen Rydberg gases and dipole-dipole energy transfer
Many-body effects in the frozen Rydberg gas

Dipole-dipole allowed

\[ pp \rightarrow ss' \]
\[ ps' \rightarrow s'p \]
\[ ps \rightarrow sp \]

Frozen Rydberg gas: atoms move only a few percent of internuclear distance in a MOT
Resonant Dipole-Dipole Energy Transfer in a Nearly Frozen Rydberg Gas

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Department of Physics, University of Virginia, Charlottesville, Virginia 22901
(Received 4 August 1997)

\[ \text{Rb}^{25}s_{1/2} + \text{Rb}^{33}s_{1/2} \rightarrow \text{Rb}^{24}p_{1/2} + \text{Rb}^{34}p_{3/2} \]
Many-Body Effects in a Frozen Rydberg Gas

I. Mourachko, D. Comparat, F. de Tomasi, A. Fioretti, P. Nosbaum,* V. M. Akulin,† and P. Pillet

*Laboratoire Aimé Cotton, CNRS II, Bât. 505, Campus d’Orsay, 91405 Orsay Cedex, France
(Received 4 August 1997)

\[
\begin{align*}
Cs_A[np_{3/2}] + Cs_B[np_{3/2}] &\rightarrow Cs_A[ns] + Cs_B[(n + 1)s], \\
Cs_C[np_{3/2}] + Cs_A[ns] &\rightarrow Cs_C[ns] + Cs_A[np_{3/2}]
\end{align*}
\]

\[
\begin{align*}
Cs_C[np_{3/2}] + Cs_B[(n + 1)s] &\rightarrow Cs_C[(n + 1)s] + Cs_B[np_{3/2}]
\end{align*}
\] (2)

Stark Field $\mathcal{E}$ [V/cm]

Ion yield from 24s Rydberg state

Detuning [MHz]
Observing the Dynamics of Dipole-Mediated Energy Transport by Interaction-Enhanced Imaging


Principle: Absorption imaging in presence of coupling light

- Rydberg atoms
- Ground state atoms
Observing the Dynamics of Dipole-Mediated Energy Transport by Interaction-Enhanced Imaging


For $n = 37$

- $|ns\rangle \langle n+1s|$  
- $|np_{3/2}\rangle \langle np_{3/2}|$

For $n > 42$

- $|F_0\rangle$  
- $|F\rangle$

\begin{align*}
1) & (n^{ns} + 3/2^{np}) \\
5/2 & (n-1)^{d_{5/2}}
\end{align*}
The light-harvesting apparatus of green sulphur bacteria and the Fenna-Matthews-Olson (FMO) protein. The excitations from absorbed light are transported through the FMO protein with detailed shown on the right. b) The pictorial representation of Förster energy transfer in the FMO protein.


Excitation blockade and collective excitations
Early experimental evidence

When the volume size is larger than the blockade radius, several atoms can be excited to the Rydberg states in the atomic cloud. However, there is rapid saturation of the excitation. The number of excited atoms is

$$\text{Number of excited atoms} \sim \frac{V_{\text{sample}}}{V_{\text{blockade}}}$$

Proper definition of the dipole blockade radius $R_b$ is:

$$\Omega \sqrt{N_b} = \left| \frac{c_p}{R_b^2} \right| \text{ where } N_b \text{ is the number of atoms contained in the blockade sphere.}$$
Dipole Blockade at Förster Resonances in High Resolution Laser Excitation of Rydberg States of Cesium Atoms

Thibault Vogt, * Matthieu Viteau, Jianming Zhao, ‡ Amodsen Chotia, Daniel Comparat, and Pierre Pillet

Laboratoire Aimé Cotton, CNRS, Bâtiment 505, Campus d’Orsay, 91405 Orsay, France

(Received 5 March 2006; published 24 August 2006)
Observation of collective excitation of two individual atoms in the Rydberg blockade regime

Alpha Gaëtan1, Yevhen Miroshnychenko1, Tatjana Wilk1, Amodsen Chotia2, Matthieu Viteau2, Daniel Comparat2, Pierre Pillet2, Antoine Browaeys1* and Philippe Grangier1
Observation of collective excitation of two individual atoms in the Rydberg blockade regime

Alpha Gaëtan¹, Yevhen Miroshnychenko¹, Tatjana Wilk¹, Amodsen Chotia², Matthieu Viteau², Daniel Comparat², Pierre Pillet², Antoine Browaeys¹* and Philippe Grangier¹
Dipole blockade in mesoscopic ensembles

Observation of coherent many-body Rabi oscillations

Y. O. Dudin, L. Li, F. Bariani and A. Kuzmich*

Hundreds of atoms!

$\Omega \Rightarrow \sqrt{N} \Omega$

- several Rabi cycles observed
- collective enhancement of Rabi frequency
Coupling a single electron to a Bose–Einstein condensate

Jonathan B. Balewski¹, Alexander T. Krupp¹, Anita Gaj¹, David Peter², Hans Peter Büchler², Robert Löw¹, Sebastian Ioffersberth¹ & Tilman Pfau¹
Only one photon can be present at a time in the atomic sample ⇒ polaritons propagate one by one! The time difference between output single photons is determined by the time traveled by each polariton inside the medium.

Transmission of the probe beam versus probe beam frequency for different incoming photon rates

(Attenuation length $l_a = 1/(n_a \sigma_0)$ of a few microns when one polariton is already excited)
Blockade condition is $R_b > l_a$, $\tau_d = 300$ ns between output pulses
Dimensions $\sigma_r \times \sigma_{ax} = 10 \times 34 \, \mu m^2$
Long range molecules
Ultralong-range Rydberg Molecules

Covalently bond molecules
Macrodimers

\[ \text{H} \cdot \text{H} \]

\[ \text{H} - \text{H} \]

\[ \delta^- \quad \delta^+ \]

Rb\((n\ell)\) + Rb\((5s)\)
Trilobite-like
Trilobite-like Rydberg Molecules

Non-polar molecule: $\text{Rb}(n) + \text{Rb}(5s) \ (l \leq 2)$

Polar molecule: $\text{Rb}(n) + \text{Rb}(5s) \ (l > 2)$

Dipole moment $\sim 1\text{K}D$

$U(R) = E_{nd} + 2\pi A_T[k(R)]|\psi_{nd0}(\vec{R})|^2.$

$U_n(R) = -\frac{1}{2n^2} + 2\pi A_T[k(R)] \sum_{l=l_{\min}}^{n-1} \frac{2l + 1}{4\pi} R_{nl}(R)^2.$

Trilobite-like Rydberg Molecules


M.A. Bellos et al. PRL 111, 053001 (2013)
Dipole-dipole-coupled double-Rydberg molecules

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(Received 21 May 2012; published 10 September 2012)
Observation of Rydberg-Atom Macrodimers: Micrometer-Sized Diatomic Molecules

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Laboratory of Physical Chemistry, ETH Vladimir-Prelog-Weg 2, 8093 Zurich, Switzerland
(Received 19 June 2016; published 19 August 2016)
Three-Body Bound States in Dipole-Dipole Interacting Rydberg Atoms

Martin Kiffner,1,2 Wenhui Li,1,3 and Dieter Jaksch2,1

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(Received 24 July 2013; published 4 December 2013)
Spontaneous evolution of a cold Rydberg gas into a cold plasma
Dipolar forces

Attractive

<table>
<thead>
<tr>
<th>F</th>
<th>Attractive</th>
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<tbody>
<tr>
<td><em>μ₁</em></td>
<td>A</td>
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<tr>
<td><em>μ₂</em></td>
<td>B</td>
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Repulsive

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<th>Repulsive</th>
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<tbody>
<tr>
<td><em>μ₁</em></td>
<td>A</td>
</tr>
<tr>
<td><em>μ₂</em></td>
<td>B</td>
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</tbody>
</table>

Energy plot:

- Vertical axis: Energy
- Horizontal axis: Distance (R)
- Green arrow: Energy increase
- Black arrow: Energy decrease
Spontaneous formation of Ultra-cold plasmas

Spontaneous ionization

\[ \gamma_{BB} \rightarrow e^- \]

Potential energy

Black-body
Collisions with background gas
Dipole-dipole collisions

Trapping of electrons in space charge

Expansion

Direct photoionization?

\[ \text{Ionization threshold} \]
\[ \text{Rydberg} \quad n \approx 30 \]

511 nm

6s\text{1/2}

6p\text{3/2}

k_B T_{\text{electron}}

Trapping laser
Ionization due to dipolar forces

$$Cs(np_{3/2}) + Cs(np_{3/2}) \rightarrow Cs^+ + Cs(n' l')$$

$$n' \ll n$$
Ultracold neutral plasmas

Direct photoionization of a gas of cold atoms
Strongly-coupled plasmas?

Gam = Coulomb Energy/Thermal Energy

\[ \Gamma = \frac{e^2}{4\pi \varepsilon_0 a / k_B T} > 1 \]

Wigner Crystallization of neutral plasma?

Killian, Science 316 705-708

Hamaguchi et al., 1997, Pohl et al., 2004