Lifetimes of ultralong-range Rydberg molecules in a dense Bose-Einstein condensate

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Cold Rydberg gases

- Using highly excited atoms embedded in BEC or cold thermal gas we can study:
  - Tunable long-range dipolar interactions
  - Quantum Information
  - Electron-atom scattering
  - Impurity physics and polarons
  - Novel bound states

Cold Rydberg gases

- Using highly excited atoms embedded in BEC or cold thermal gas we can study:
  - Novel bound states
- **Question:** How long do these excitations live in a dense gas?
- Motivated by studies in Rb
  - Low density – 2011 JPB 44 184004
  - High density – PRX 6, 031020
- Our work in Sr
  - Low density – PRA 93, 022702, 2015
  - High density (this talk) – arXiv:1707.02354

Rydberg atoms

- Atomic state of high principal quantum number $n$
- Electron orbits far away from ionic core
- Highly exaggerated properties that scale with $n$
  - Massive size
  - Long lifetime
  - Huge dipole moment

<table>
<thead>
<tr>
<th>Property</th>
<th>$n$ Dependence</th>
<th>Sr $5s5p^3S_1$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Binding Energy</td>
<td>$n^{-2}$</td>
<td>50 cm$^{-1}$</td>
</tr>
<tr>
<td>Radiative Lifetime</td>
<td>$n^3$</td>
<td>38 μs</td>
</tr>
<tr>
<td>Rydberg Radius $(r)$</td>
<td>$n^2$</td>
<td>200 nm</td>
</tr>
<tr>
<td>Ionization Field</td>
<td>$n^{-4}$</td>
<td>65 V/cm</td>
</tr>
<tr>
<td>Polarizability</td>
<td>$n^7$</td>
<td>56 MHz cm$^2$/V$^2$</td>
</tr>
</tbody>
</table>
Experimental method

• Prepare BEC of $4 \times 10^5 \ ^{84}\text{Sr}$ atoms
  • 1064nm ODT, harmonic potential
  • Peak density $= 4 \times 10^{14} \text{ cm}^{-3}$
• Excite to triplet states via $^3\text{P}_1$ intercombination line
• Obtain spectrum by scanning UV photon energy
• Hold excited Rydberg atoms for 1-100 $\mu$s
• Detect Rydberg population on MCP with selective field ionization (SFI)
Rydberg molecules

\[ V(\vec{R}) = \frac{2n\hbar^2 a_s(k)}{m_e} |\psi_{nl}(\vec{R})|^2 + \frac{6n\hbar^2 a_p(k)^3}{m_e} |\nabla\psi_{nl}(\vec{R})|^2 \]

- Exciting a Rydberg atom in a cold dense gas
- Scattering between Rydberg electron and ground-state atoms can form bound states
- Predicted by Greene et. al - Phys. Rev. Lett. 85, 2458, 2000
- First observed in 2009 by Pfau group - Nature 458, 1005-1008, 2009
Rydberg molecules

![Graph showing MCP signal dependence on detuning and internuclear distance. The graph includes peaks labeled for n=38, Atomic, Dimers, Trimers, and Tetramers.](image)
Increasing principal quantum number

\[ \rho = 3 \times 10^{14} \text{ cm}^{-3} \]

Atoms per Rydberg Volume

Principal Quantum Number

- \( \rho = 3 \times 10^{14} \text{ cm}^{-3} \)
- \( n=38 \)
- \( n=49 \)
- \( n=60 \)

MCP Signal (arb)

Detuning (MHz)
Increasing principal quantum number

Binding Energy $\sim 1/n^6$
Neutral atoms inside Rydberg $\sim n^6$

$\rho = 3 \times 10^{14} \text{ cm}^{-3}$

MCP Signal (arb)

Atoms per Rydberg Volume

-30 -25 -20 -15 -10 -5 0

Detuning (MHz)
Laser detuning selects local density

• Mean field predicts shift in resonance proportional to local density

\[ \Delta E = \frac{2\pi \hbar^2}{m_e} A_S(k) \rho \]

• Large red laser detuning implies excitation at higher density.

• With a narrow laser we can choose excitation density precisely
Population dynamics

- Measure decay of Rydberg population at a fixed laser detuning (fixed density)
- At high densities we observe fast initial loss and slow radiative decay at later times
- Indicative of molecular loss channels other than spontaneous emission
- Possible mechanisms
  - Sr$_2^+$ production
  - L-changing collisions
Loss mechanisms – $\text{Sr}_2^+$ production

- Reaction initiated by core-ion + nearest ground-state atom polarization potential
- Electron takes away binding energy and escapes the trap
- Observed as loss in molecular signal
- $\text{Rb}_2^+$ directly observed in other experiments

$$V_{\text{ion}}(r) \propto -\frac{C_4}{r^4}$$

Dominant at short range

$$\text{Sr}^* + \text{Sr} \rightarrow \text{Sr}_2^+ + e^- + \Delta E$$
Loss mechanisms – $L$ changing collisions

- Reaction initiated by core-ion + nearest ground-state atom polarization potential
- Rydberg atom in high $L$ is ejected from the trap
- Ejected Ryds. undergo no more collisions, $1/e$ lifetime scales as $n^3$
- Observed as residual signal and characteristic shift in SFI profile

\[
\text{Sr}^* (5sns) + \text{Sr} \rightarrow \text{Sr}^* (5sn'\ell') + \text{Sr} + \Delta E
\]
Model

- Results consistent with simple rate equation model
- Three component fit with rate constants
  - $\Gamma_{\text{AI}}$ – Associative ionization rate
  - $\Gamma_L$ – L-changing collision rate
  - $\Gamma_R$ – Radiative decay rate
- Fit sum of parent atoms, $N_p$, and L-changed atoms, $N_L$, to total population

\[
N_p(t) = N_0 e^{-(\Gamma_{\text{AI}} + \Gamma_L + \Gamma_R)t}
\]
\[
N_L(t) = N_0 \frac{\Gamma_L}{\Gamma_{\text{AI}} + \Gamma_L} e^{-\Gamma_R t} \left[ 1 - e^{-(\Gamma_{\text{AI}} + \Gamma_L)t} \right]
\]
$n$ dependence of rates

- Destruction rates largely independent of $n$ when many atoms present in Ryd. orbital.
- Indicates presence of one ground-state atom near the core-ion leads to destruction of Ryd. molecule.
- Radiative lifetime negligible compared to collision rates
Density dependence of rates

Collision time $t$ scales inversely with density in the limit $E_{\text{coll}} \rightarrow 0$

$$t = \int_{r_i}^{r_f} \frac{dr}{v(r)} = \int_{r_i}^{r_f} \frac{dr}{\sqrt{\frac{2}{\mu} \left( E_{\text{coll}} + \frac{C_4}{r^4} \right)}}$$

$$t \propto -\int_{r_i}^{r_f} r^2 dr \propto r_i^3$$

$$r_i \propto \rho^{-1/3} \quad \Rightarrow \quad t \propto 1/\rho$$
Density dependence of rates

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$$t = \int_{r_i}^{r_f} \frac{dr}{v(r)} = \int_{r_i}^{r_f} \frac{dr}{\sqrt{\frac{2}{\mu} \left( F_{\text{coll}} + C \frac{A}{r^4} \right)}}$$

$$t \propto -\int_{r_i}^{r_f} r^2 dr \propto r_i^3$$

$$r_i \propto \rho^{-1/3} \implies t \propto 1/\rho$$
Conclusions and outlook

• Lifetime of Rydberg excitation in dense gases is limited by collision time with nearest neighbor
• Two likely decay mechanisms
  • $\text{Sr}_2^+$ production
  • L-changing collisions
• Described by simple kinematic model

• Should rate decrease at higher $n$?
  • Electron spends less time near the nucleus
• Do particle statistics affect molecular formation / lifetimes?
  • Experiments with $^{87}\text{Sr}$ underway
Thank You

Killian Group (Rice)
Rydberg:
- Francisco Camargo
- Roger Ding
- Joe Whalen
- Soumya Kanungu
Neutral/Lattice:
- Jim Aman
- Josh Hill
Plasma:
- Thomas Langin
- Grant Gorman
- Zhitao Chen

Theory Collaborators
- Jesús Pérez-Ríos
  Purdue
  Universidad Del Turabo
- Joachim Burgdörfer
  Vienna University of Technology
- Shuhei Yoshida

July 26, 2017

Purdue University
TU Wien
NSF
The Welch Foundation
Ultracold Atoms and Plasmas
Signature of L-changing collisions

State Changing Collisions at n=72

Hold Time
- 0.0us
- 10.1us
- 100.0us

MCP Counts

Adiabatic Ionization

Diabatic Ionization

Electric Field (V/cm)

0 10 20 30 40 50 60 70 80 90 100

Relative Population

Time Delay (μs)

0 20 40 60 80 100

\[ \Delta = -21.2 \text{ MHz} = 2.7 \times 10^{14} \text{ cm}^{-3} \]