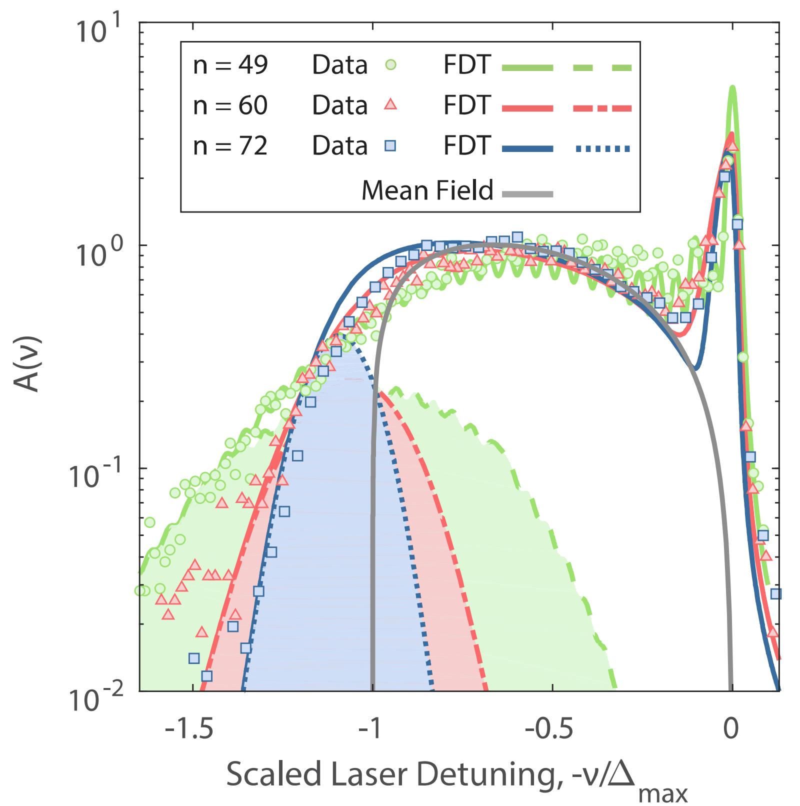
Lifetimes of ultralong-range Rydberg molecules in a dense Bose-Einstein condensate

Experiment: **Joseph D. Whalen**, F. Camargo, R. Ding, T. C. Killian, F. B. Dunning

Theory: J. Pérez-Ríos, S. Yoshida, J. Burgdörfer

Cold Rydberg gases

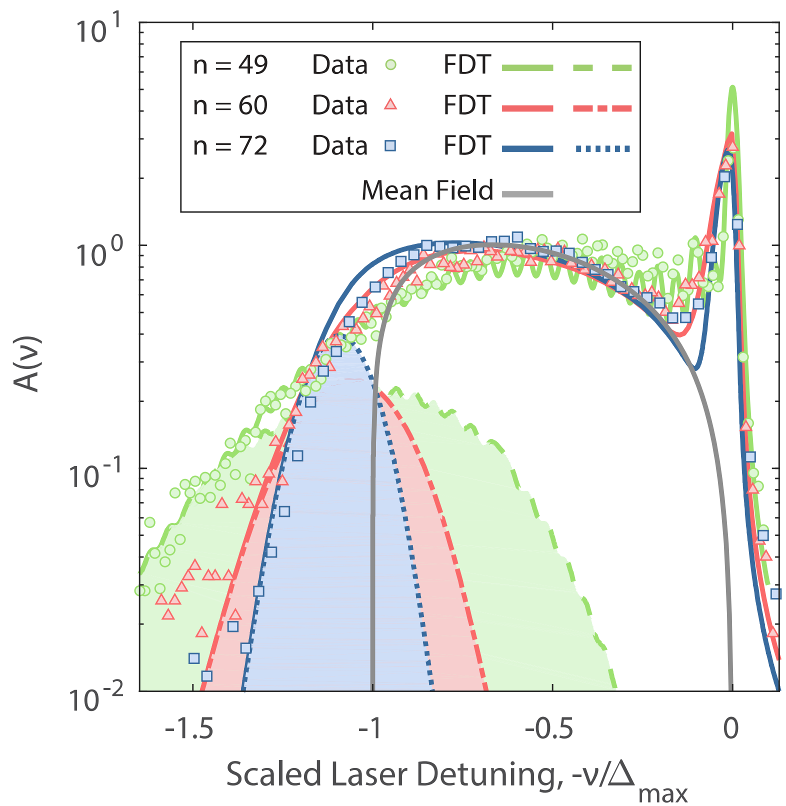
- Using highly excited atoms embedded in BEC or cold thermal gas we can study:
  - Tunable long-range dipolar interactions
  - Quantum Information
  - Electron-atom scattering
  - Impurity physics and polarons
  - Novel bound states



Camargo et al. https://arxiv.org/abs/1706.03717

Cold Rydberg gases

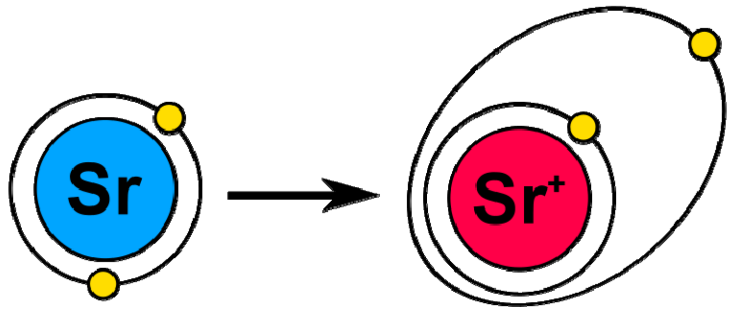
- Using highly excited atoms embedded in BEC or cold thermal gas we can study:
  - Novel bound states
- Question: How long do these excitations live in a dense gas?
- Motivated by studies in Rb
  - Low density – 2011 JPB **44** 184004
  - High density – PRX **6**, 031020
- Our work in Sr
  - Low density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



Camargo et al. https://arxiv.org/abs/1706.03717

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

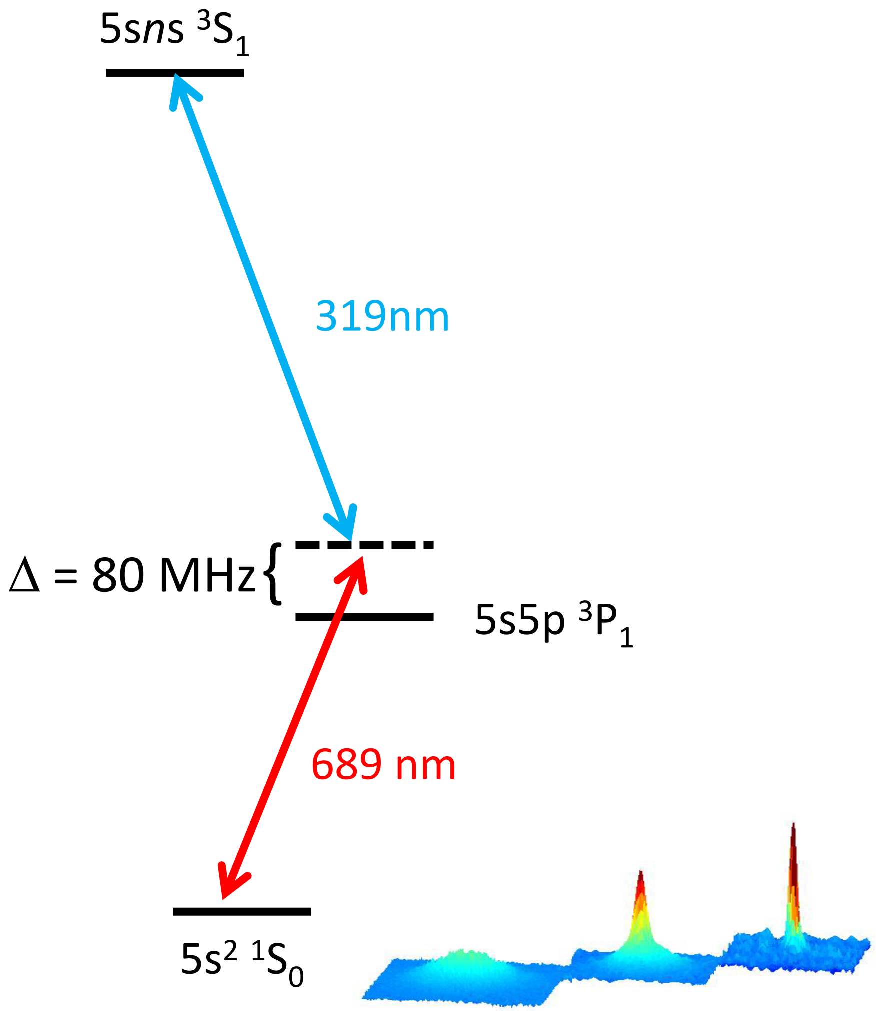


$5s^2\ ^1S_0$ $5sns\ ^3S_1$

Property	n Dependence	Sr $5s50s\ ^3S_1$
Binding Energy	n^{-2}	50 cm^{-1}
Radiative Lifetime	n^3	38 μs
Rydberg Radius $\langle r \rangle$	n^2	200 nm
Ionization Field	n^{-4}	65 V/cm
Polarizability	n^7	56 MHz cm^2/V^2

Experimental method

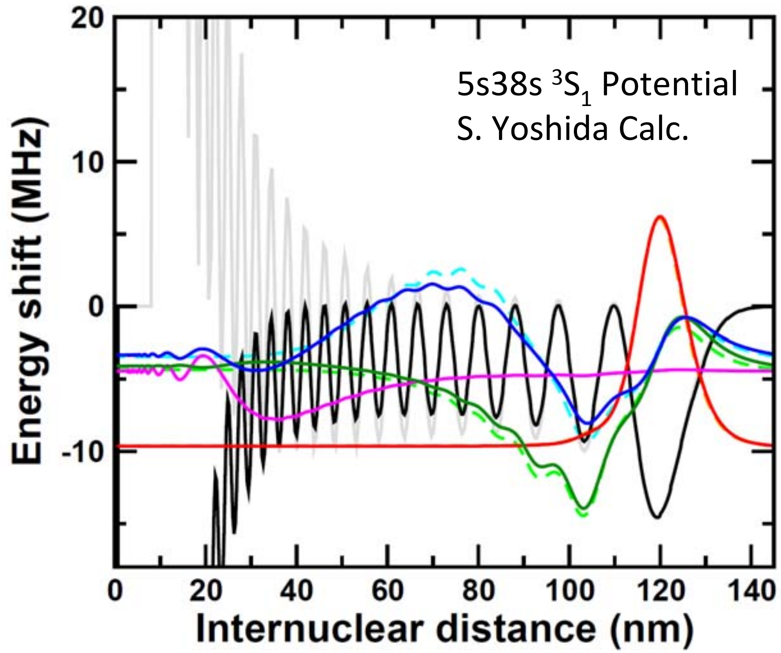
- Prepare BEC of 4 x 10^5 ^{84}Sr atoms
  - 1064nm ODT, harmonic potential
  - Peak density = 4 x 10^{14} cm^{-3}
- Excite to triplet states via $^3P_1$ intercombination line
- Obtain spectrum by scanning UV photon energy
- Hold excited Rydberg atoms for 1-100 µs
- Detect Rydberg population on MCP with selective field ionization (SFI)



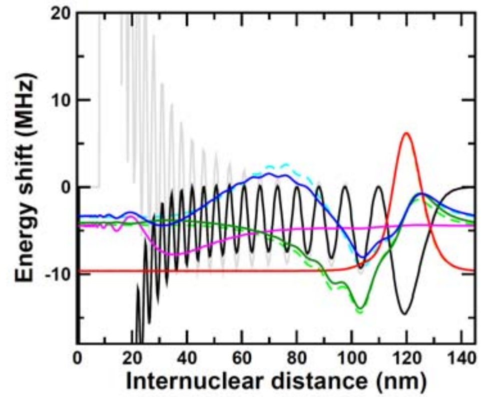
Rydberg molecules

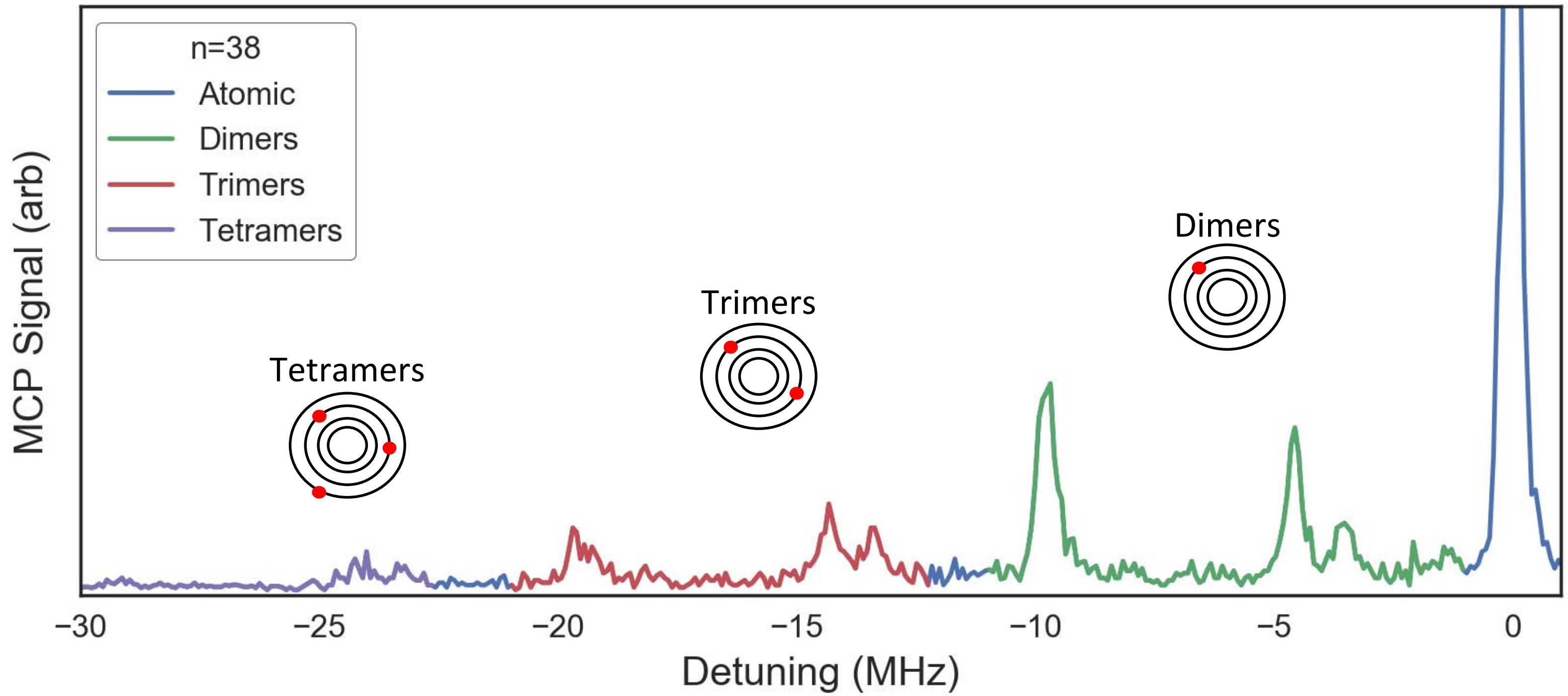
$$V(\vec{R}) = \frac{2\pi\hbar^2 a_s(k)}{m_e}\left|\psi_{nl}(\vec{R})\right|^2 + \frac{6\pi\hbar^2 a_p(k)^3}{m_e}\left|\vec{\nabla}\psi_{nl}(\vec{R})\right|^2$$

- 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

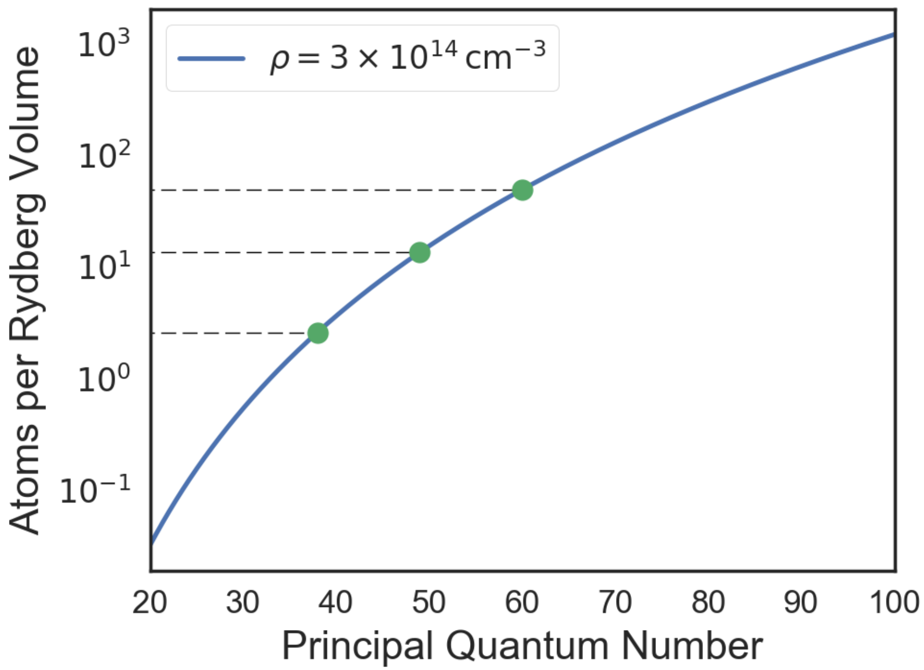


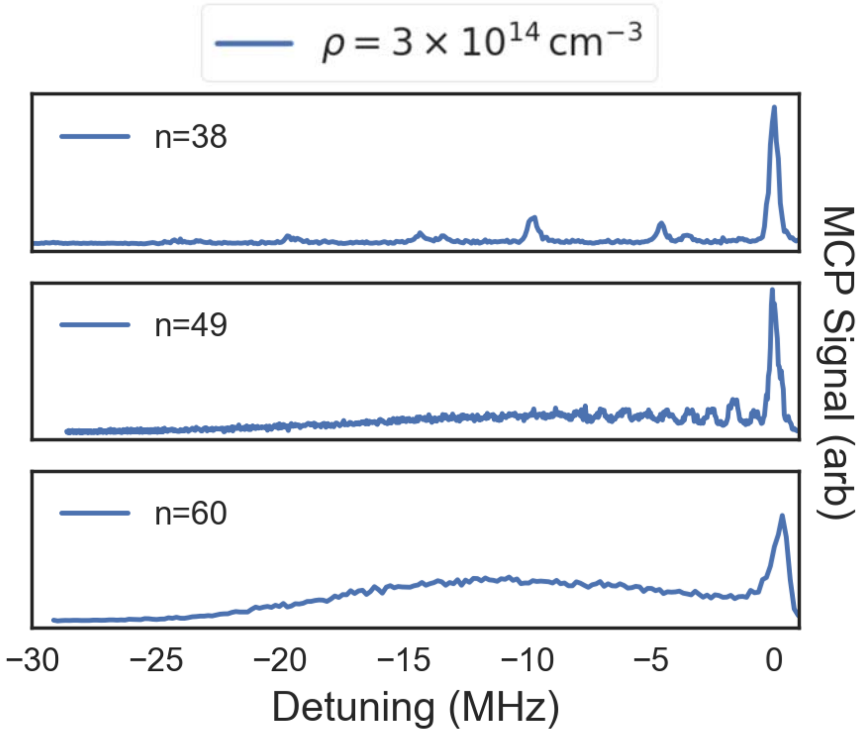
Rydberg molecules



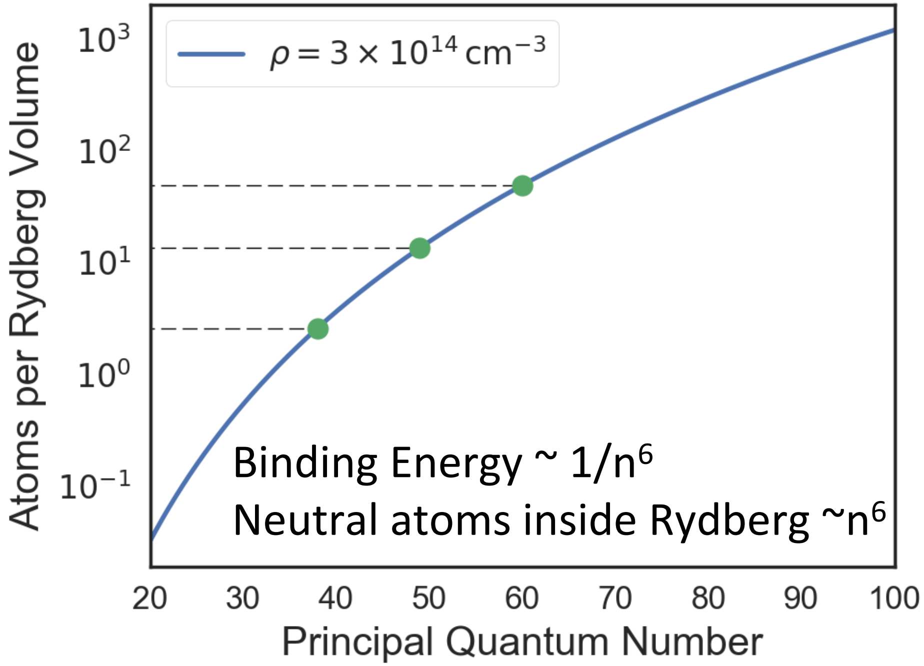


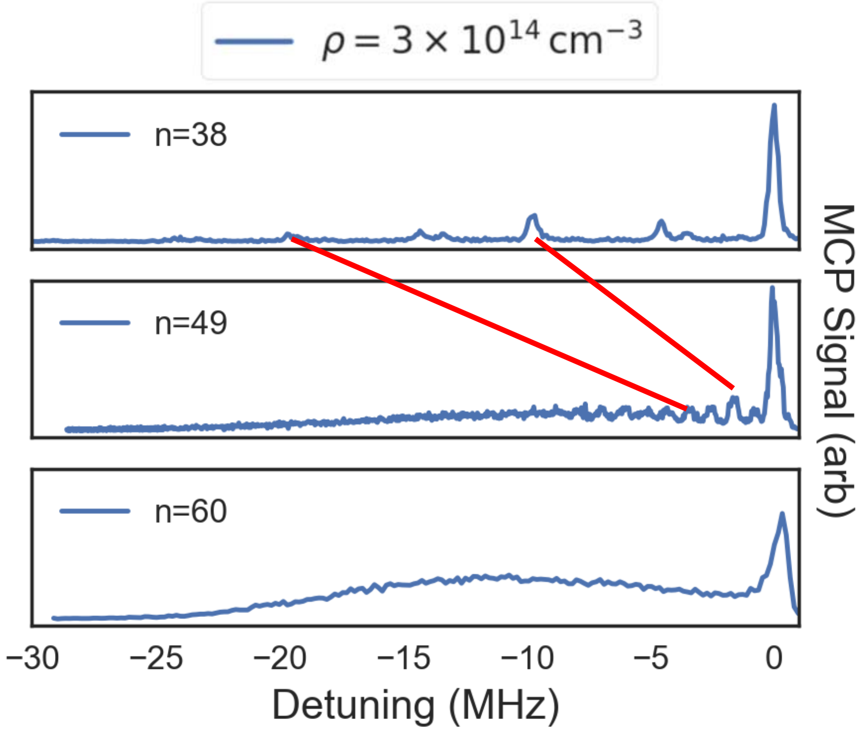
Increasing principal quantum number





Increasing principal quantum number



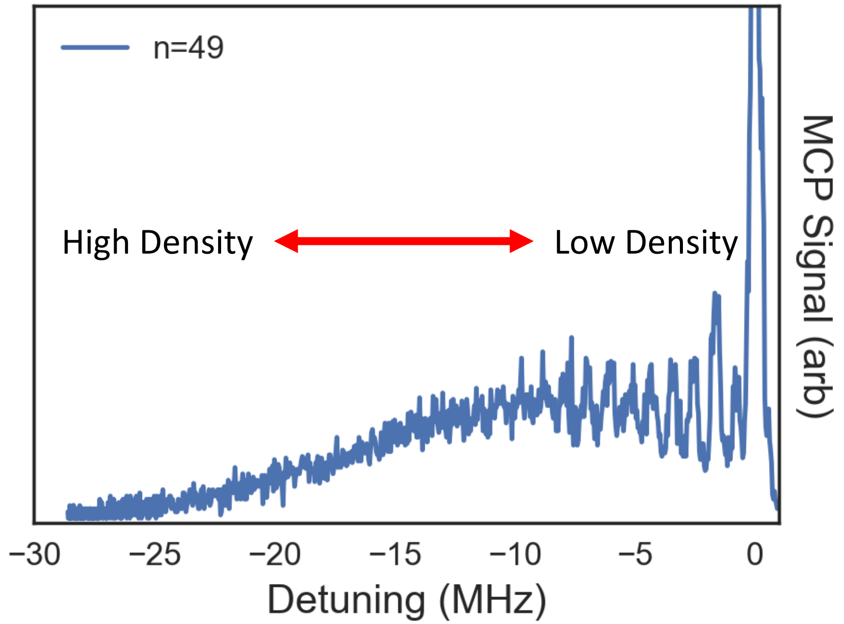


Laser detuning selects local density

- Mean field predicts shift in resonance proportional to local density

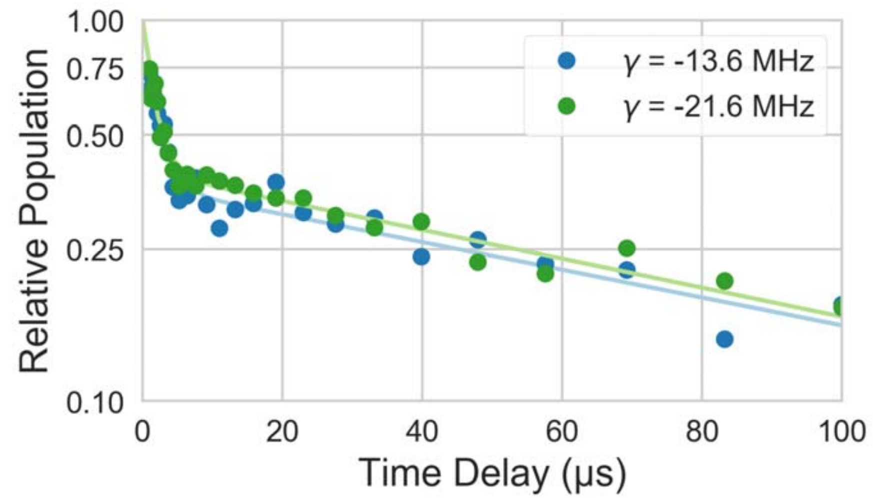
$$\Delta E = \frac{2\pi\hbar^2}{m_e} A_s(k)\rho$$

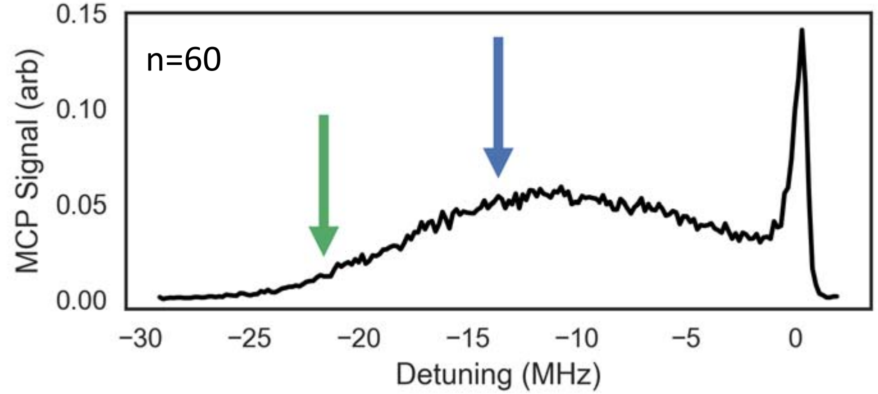
- Large red laser detuning implies excitation at higher density.
- With a narrow laser we can choose excitation density precisely



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

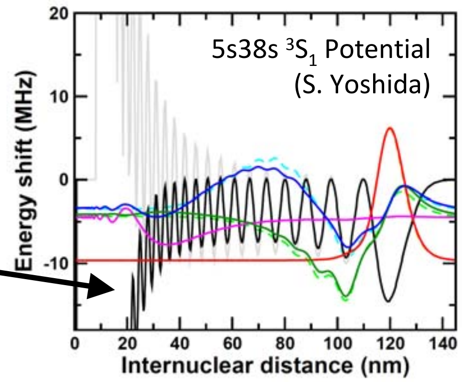




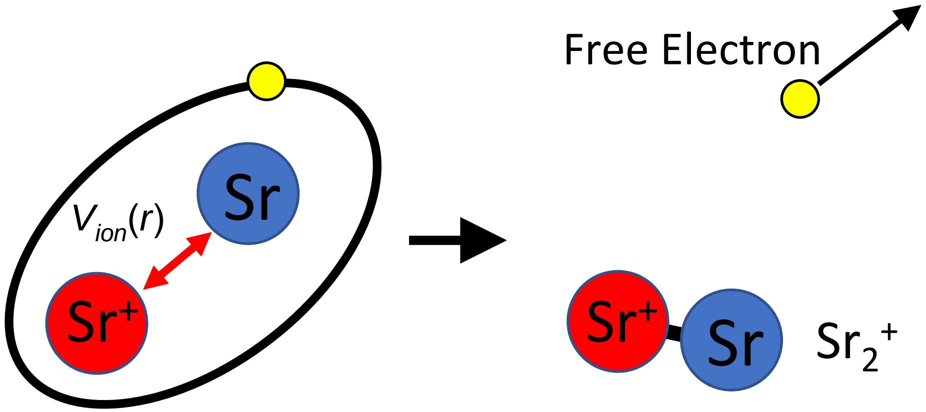
Loss mechanisms – Sr_2^+ production

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

Dominant at short range



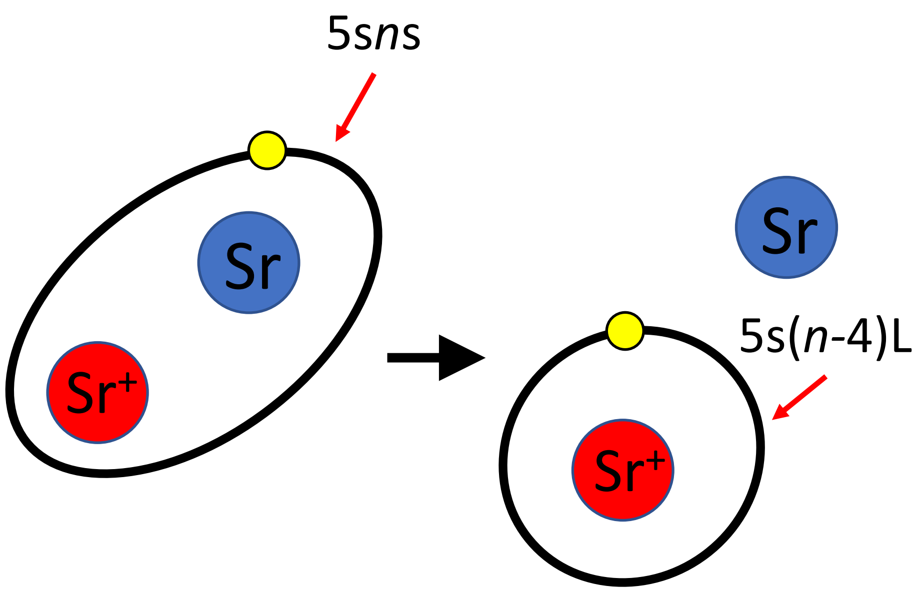
- Reaction initiated by core-ion + nearest ground-state atom polarization potential
- Electron takes away binding energy and escapes the trap
- Observed as loss in molecular signal
- Rb_2^+ directly observed in other experiments



$$\mathrm{Sr}^* + \mathrm{Sr} \rightarrow \mathrm{Sr}_2^+ + e^- + \Delta E$$

Loss mechanisms – *L* changing collisions

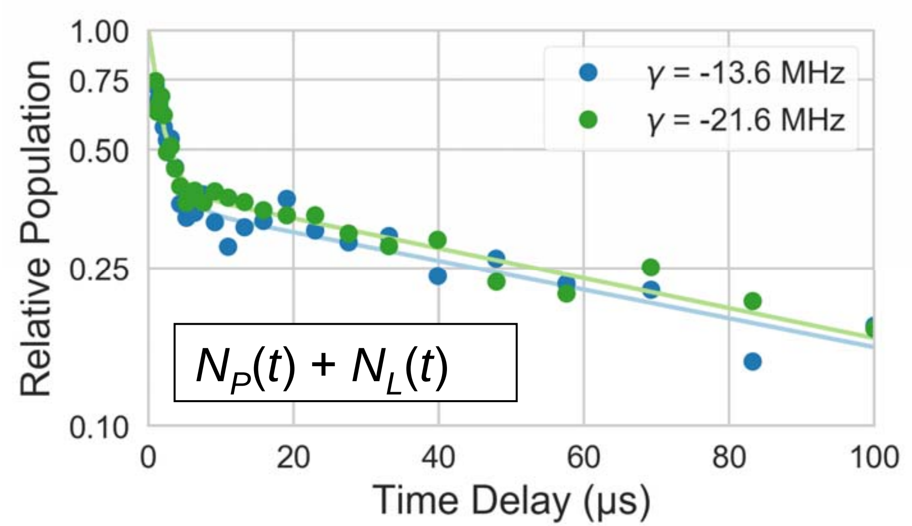
- Reaction initiated by core-ion + nearest ground-state atom polarization potential
- Rydberg atom in high L is ejected from the trap
- Ejected Ryds. undergo no more collisions, 1/e lifetime scales as n^3
- Observed as residual signal and characteristic shift in SFI profile



$$\mathrm{Sr}^*(5sns) + \mathrm{Sr} \rightarrow \mathrm{Sr}^*(5sn'\ell') + \mathrm{Sr} + \Delta E$$

Model

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



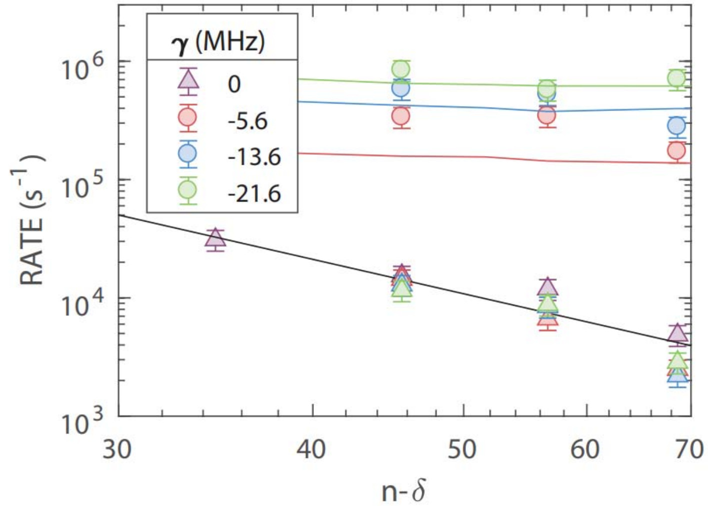
$$N_p(t) = N_0 e^{-(\Gamma_{AI}+\Gamma_L+\Gamma_R)t}$$

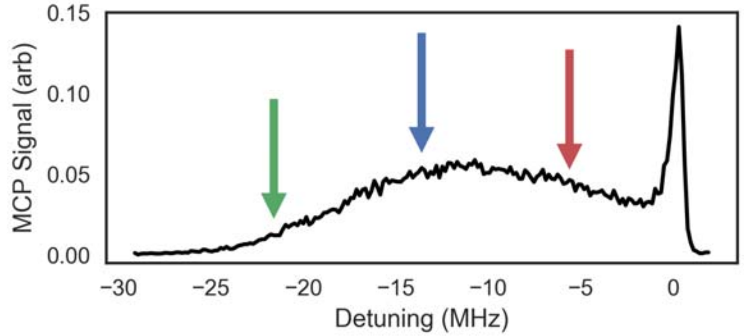
$$N_L(t) = N_0 \frac{\Gamma_L}{\Gamma_{AI}+\Gamma_L} e^{-\Gamma_R t}\left[1 - e^{-(\Gamma_{AI}+\Gamma_L)t}\right]$$

n dependence of rates

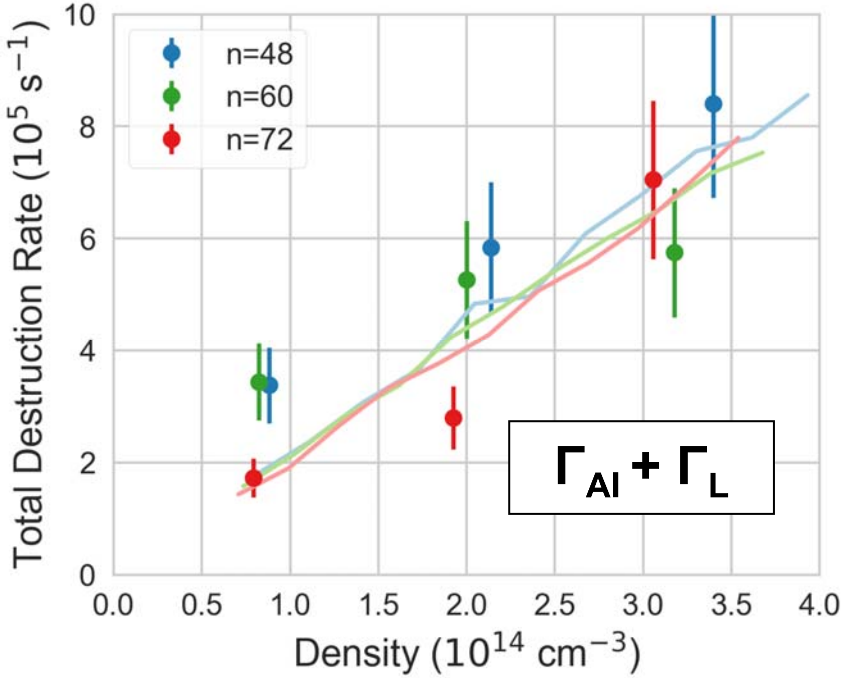
$\Gamma_{AI} + \Gamma_{L}$

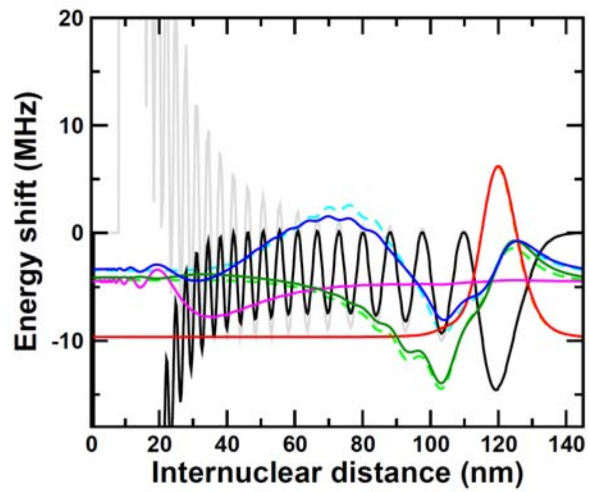
- Destruction rates largely independent of *n* when many atoms present in Ryd. orbital.
- Indicates presence of one ground-state atom near the core-ion leads to destruction of Ryd. molecule.
- Radiative lifetime negligible compared to collision rates





Density dependence of rates





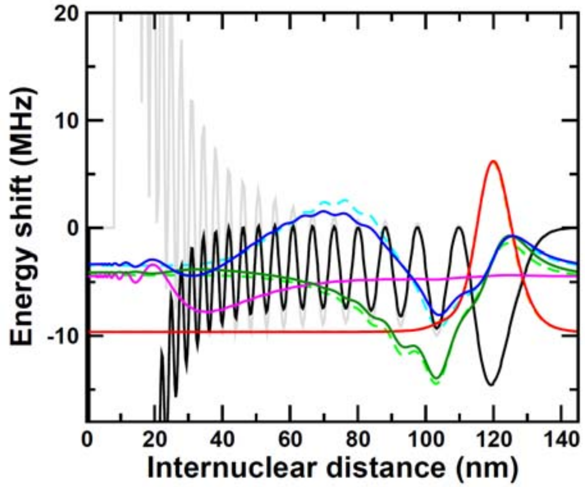
Collision time t scales inversely with density in the limit $E_{coll} \rightarrow 0$

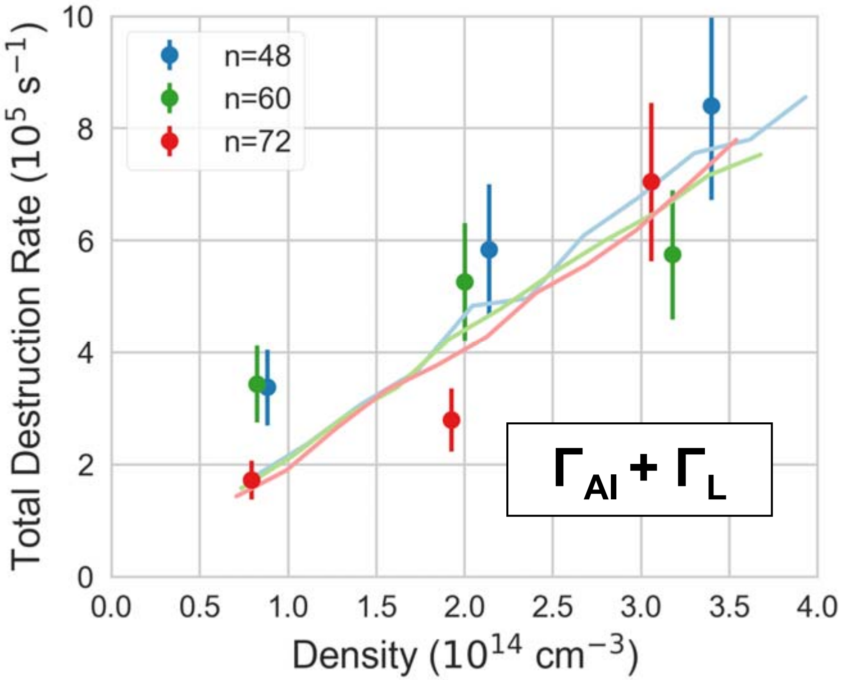
$$t = \int_{r_i}^{r_f} \frac{\mathrm{d}r}{v(r)} = \int_{r_i}^{r_f} \frac{\mathrm{d}r}{\sqrt{\frac{2}{\mu}\left(E_{coll} + \frac{C_4}{r^4}\right)}}$$

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

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

Density dependence of rates





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

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

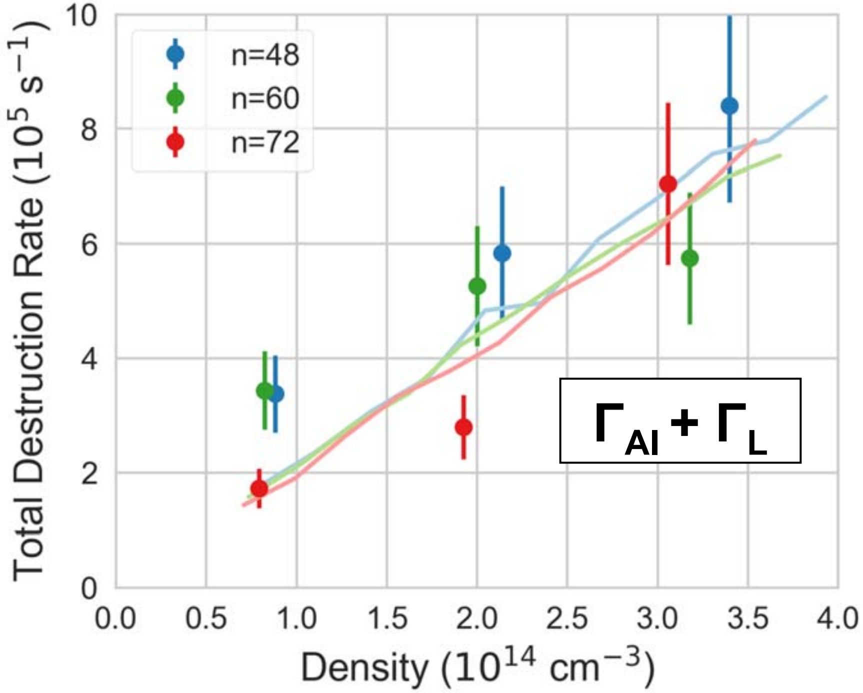
$$t \propto -\int_{r_i}^{r_f} r^2 \mathrm{d}r \propto r_i^3$$

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

Conclusions and outlook

- Lifetime of Rydberg excitation in dense gases is limited by collision time with nearest neighbor
- Two likely decay mechanisms
  - 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}Sr underway



Thank You

Killian Group (Rice)

Rydberg:

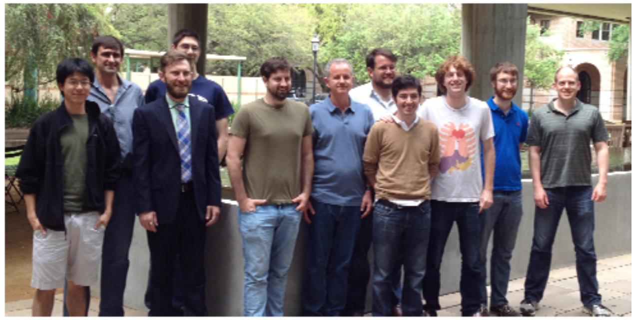
- Francisco Camargo
- Roger Ding
- Joe Whalen
- Soumya Kanunga

Neutral/Lattice:

- Jim Aman
- Josh Hill

Plasma:

- Thomas Langin
- Grant Gorman
- Zhitao Chen



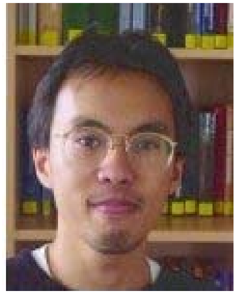
Barry Dunning

Tom Killian

Theory Collaborators

Jesús Pérez-Ríos
Purdue
Universidad Del Turabo

Joachim Burgdörfer



Shuhei Yoshida

Vienna University of Technology

Signature of L-changing collisions