Frozen Rydberg Gas:

A frozen Rydberg gas is a sample of atoms with highly excited electronic states and very low temperature. In our experiments, ^{85}Rb atoms are cooled and trapped in a magneto-optical trap (MOT) (fig.1&2), and excited to the Rydberg states (the principle quantum number n ~30) by pulsed dye lasers (fig.3). In such a system, the density of Rydberg atoms N_d is typically ~10^9atoms/cm^3, and the temperature T is ~300μK. The typical displacement $\Delta R = v\tau$~1μm of atoms moving at an average velocity v~38cm/s during the experiment time τ ~3μs is much less than the average interatomic distance $R_0 \sim (3/4\pi N_d)^{1/3} = 10\mu m$. So to first order approximation, they can be considered "frozen" in their position during the experimental process.

The exaggerated properties of Rydberg states, such as long lifetime, large size and large dipole moments etc. combined with extremely slow translational motion makes this system an ideal candidates for experiments involving novel collective phenomena. In fact, two of such many body effects have been observed in this lab, they are resonant dipole-dipole transfers [1][2], spontaneous evolution to an ultra-cold plasma[3]. Others to be expected in such a frozen Rydberg gas include dipole-dipole interactions, superradiance etc.

Laser Cooling and Trapping

Circularly polarized laser light and spatially varying magnetic field produces a position dependent force. For 1 dimension, as seen below, an atom on the left is closer to resonance with the $^{+}$(right circularly polarized) light due to the Zeeman shift, and will preferentially absorb it receiving a kick towards zero field. The same is true for an atom to the right of zero field. Moreover, with the red detuned lasers, the atoms will absorb more lights from the beam propagating along the direction opposite to their moving direction than the other beam due to the Doppler effect, therefore atoms always experience a viscous force which slow down their velocities. The motion of atoms with small displacements and velocities can approximated as a damped harmonic oscillator:

$$F = -\ x - a\dot{x}\ , \qquad \ddot{x} + 2\ \dot{x} + \ {}_0^2 x = 0$$

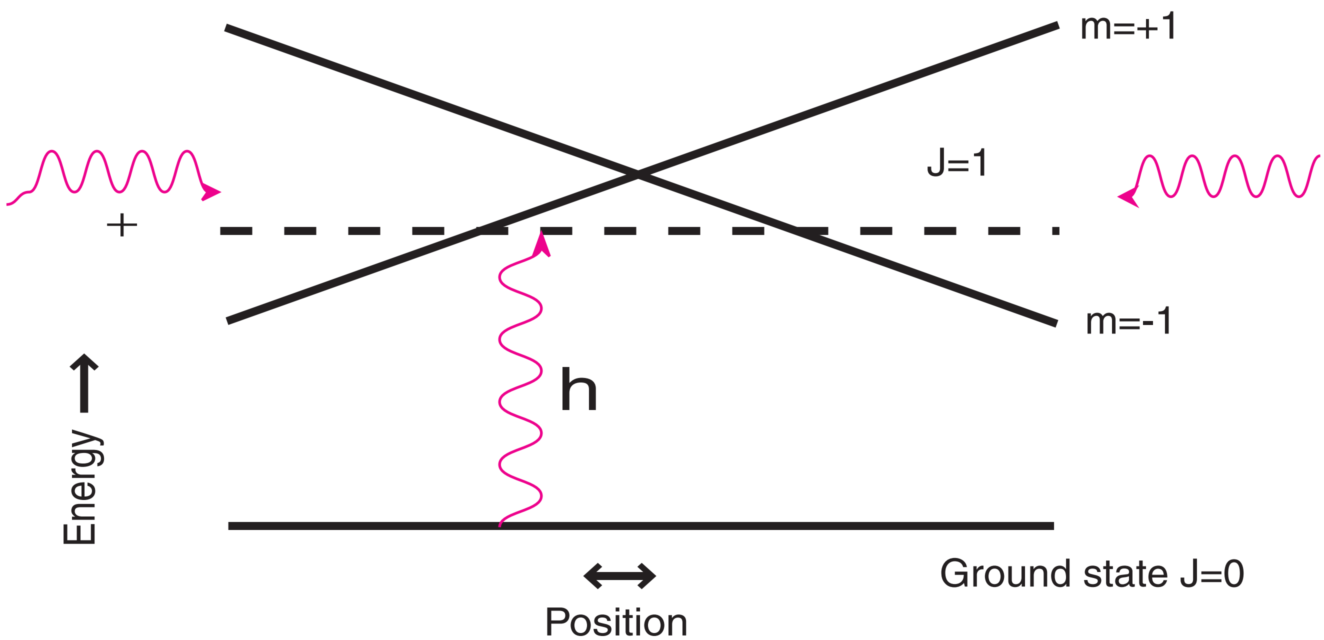
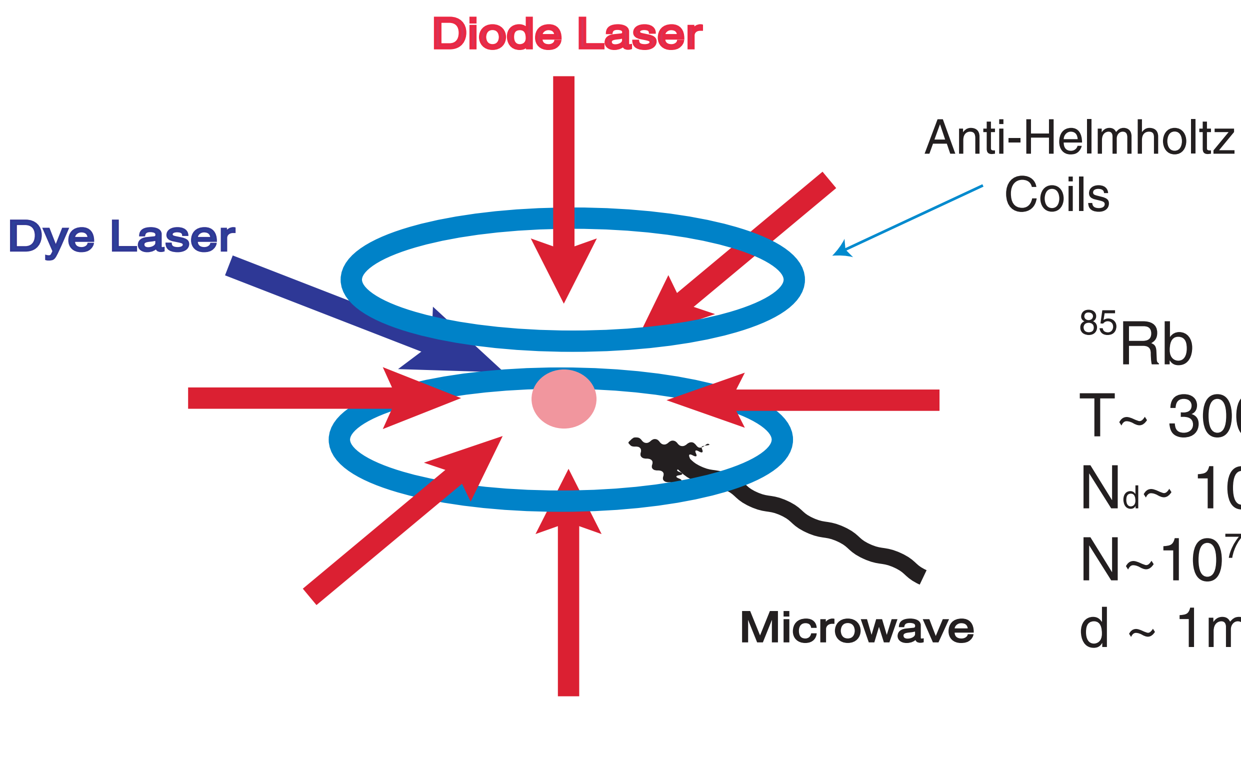


Figure 1

Magneto-Optical Trap

Generalize the principles of laser cooling and trapping in Fig.1 to 3 dimensions , using 6 laser beams and a pair of anti-Helmholtz coils to create a magneto-optical trap(MOT)[4]. Fig.2 shows the key elements of the trap. The trap is vapor loaded, while ^{85}Rb atoms are emitted into vacuum by heating cathode getters of Rubidium. The typical trap parameters are showed below. Dye laser and microwave are used to excite and transfer Rydberg populations.



^{85}Rb

T~ 300 k

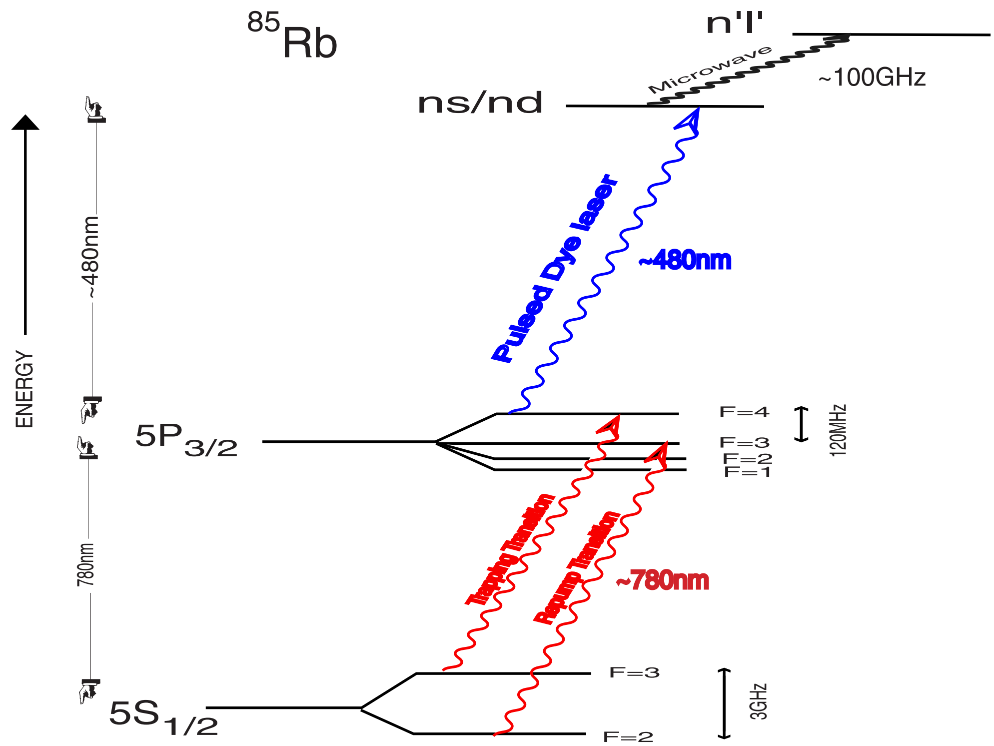
N_d~ 10^{10} cm^{-3}

N~10^7 atoms

d ~ 1mm

Figure 2

Excitation Scheme



Experimental Timing Sequence

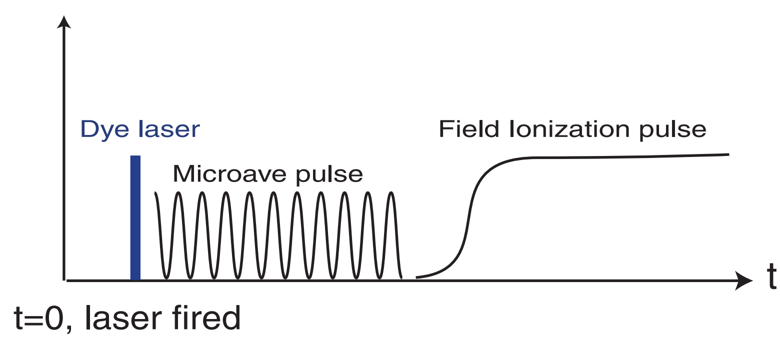


Figure 3

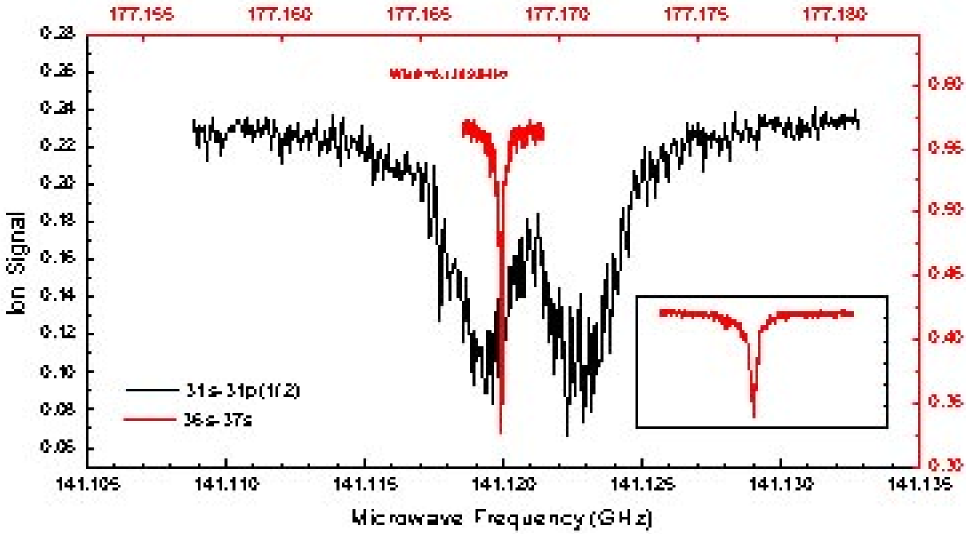
Microwave Spectroscopy

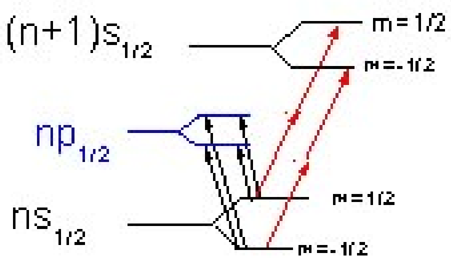
The basic idea of this research is to use microwave spectroscopy to probe collective phenomena of frozen Rydberg gas, more specifically is to look at how those many body interactions affect the linewidth and amplitude of the spectra of microwave transitions.

The basic excitation scheme and experimental timing are shown in fig.3. A microwave pulse is sent into the atomic sample to transfer atoms to some nearby Rydberg states after laser excitation. We get a microwave spectrum by detecting the final state of this microwave transition while scanning microwave frequency.

To be able to use a microwave spectrum to probe an interaction, we have to find transitions, which has much narrower linewidth than the interaction we are interested in, in spite of the 10G/cm magnetic field gradients in the trap (fig.2). Figure 4-7 show several such spectra and how they can be use to study broadening effect.

s-p Transition vs. s-s Transition





s-s two photon transitions

$\Delta m = 0$ Allowed

$\Delta m = \pm 1$ Not allowed

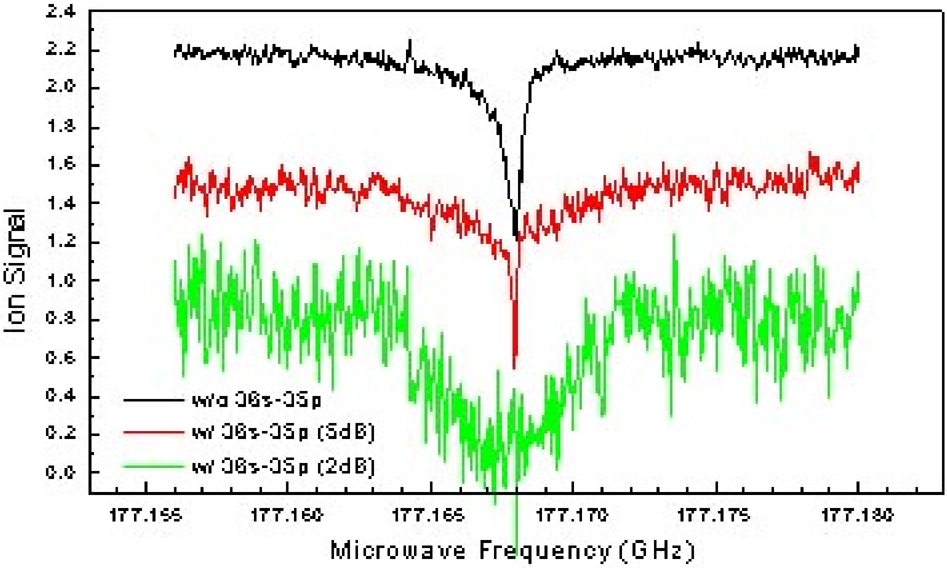
In single photon s-p transitions, $\Delta m = 0, \pm 1$ transitions are all allowed. 5MHz is the narrowest linewidth we can ever get due to the presence of strong inhomogeneous trapping magnetic field(fig.2). Comparing to 100MHz widths of some dipole-dipole resonant transfer[2][5], it will still work as a sensitive probe for such phenomena.

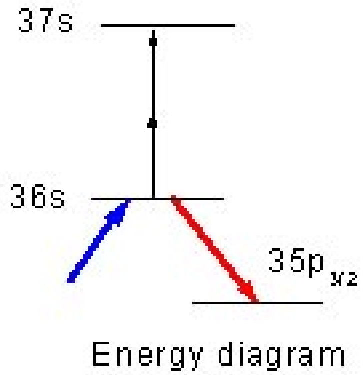
Two photon s-s transitions, with seletion rule $\Delta m = 0$ and the same g factors for both s states, the linewidth is free of the effect of magnetic field, can be reduced to transform limit of ~ 100kHz linewidth, this will be sensitive enough for most of many body effects.

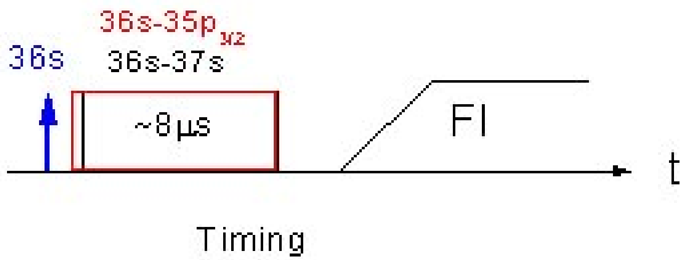
Here s-p and s-s transitions are plot on the same scale.

Figure 4

Probe Atom-Photon Interaction with s-s Transition







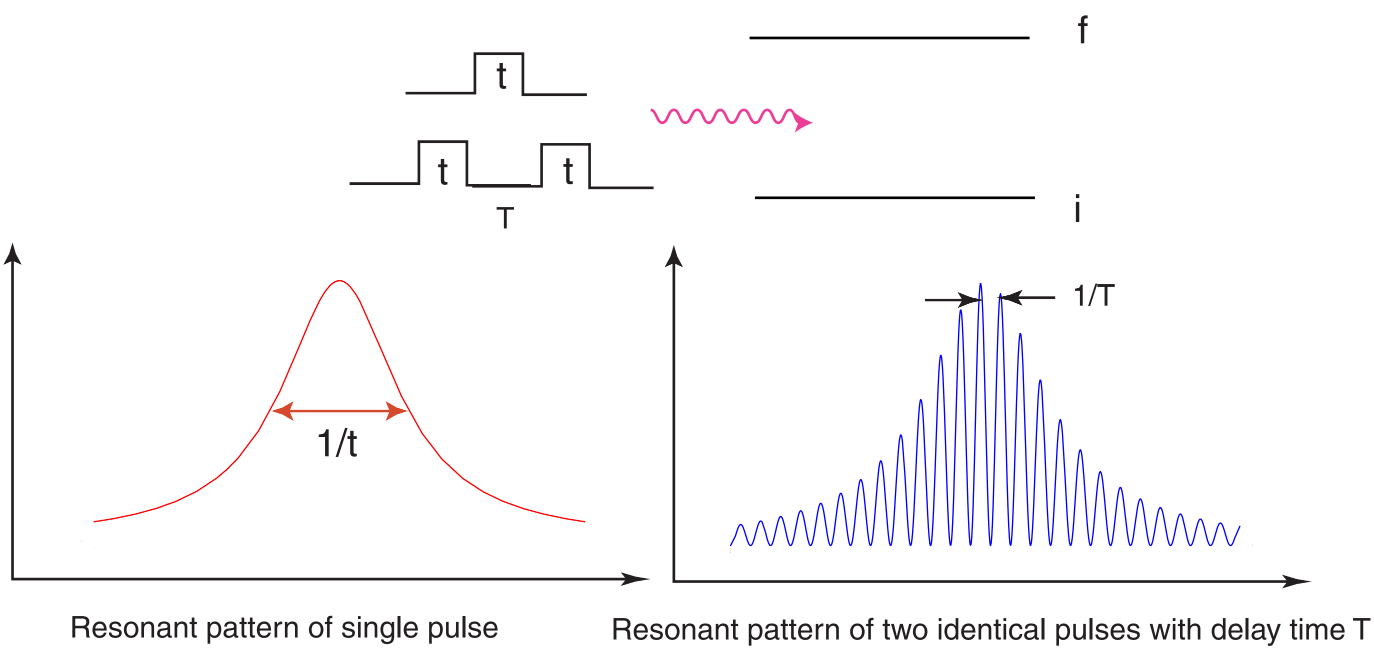
These spectra show how atom-photon interaction broaden the s-s transition spectra. After laser excitation, two microwave pulse are sent into system at the same time. Fix one of microwave frequency on the resonance of 36s-35p$_{3/2}$, scan another microwave frequency through the resonance of 36s-37s transtion.

Without the s-p pulse, the linewidth of s-s transition is narrower than 1MHz (black trace). With the presence of s-p pulse, s-s transtion is partly broadened(red trace) since 36s state has been modified by atom-photon interaction. Increasing microwave power, the transition are completely broadened to 5MHz(green trace)

Figure 5

Ramsey Interference Method

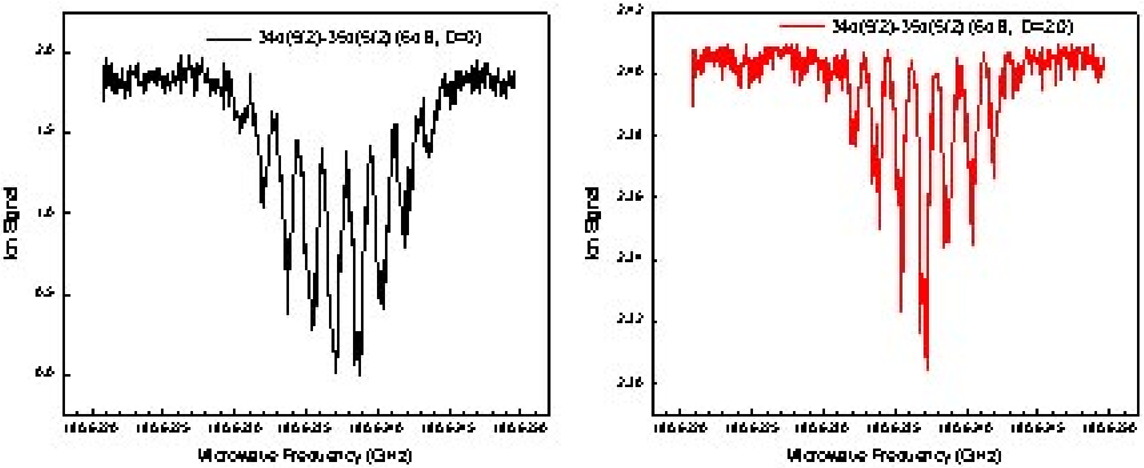
Driving a two-level system with microwave pulses



With two microwave pulses seperated by T, the spectrum has an envelope that is identical to the single pulse and is modulated due to the interference. This Ramsey fringes enable one to increase the spectral resolution without using very long pulses.

Figure 6

Probe the Effect of Ions with Ramsey Fringes



$34d_{5/2}$-$35d_{5/2}$ Ramsey Fringes (D = Optical Density)

These spectra show how the presence of ions wash out the Ramsey fringes. After atoms are laser excited to 34d state, two 1 μs microwave pulses, seperated by 6μs are sent into the sample, scan the microwave frequency through the resonance of $34d_{5/2}$ - $35d_{5/2}$ transition, we then get Ramsey fringes.

In the black trace, the laser intensity is higher, it gives more ions (photon ions and/or plasma ions), these ions dephase the coherent evolution of the atomic sample, the Ramsey fringes are partly washed out. In the red trace, the laser intensity has been reduced by a factor of 100, consequently, the density of ions is much smaller, there are almost no dephasing from ions, the contrast of fringes is about 1.

Figure 7

Improve the Quantum Defects for ^{85}Rb

With these high spectral resolution microwave spectroscopy, we have measured resonant frequencies of ns-np, ns-(n+1)s, and nd-(n+1)d transition of cold Rydberg atoms in MOT for n ~ 27-37. We are able to improve the quantum defects for the s,p and d states of ^{85}Rb by an order of magnitude. The previous and improved quantum defects are listed below.

The energy of (n,l) state from ionization limit is

$$E(n,l) = -Ry/(n-\delta)^2$$
$$\delta(n,l) = \delta_0 + \delta_2/(n-\delta_0)^2$$

		Previous Value	Improved Value
$ns_{1/2}$	δ_0	3.13109(2)	3.1311793(18)
	δ_2	0.204(8)	0.1785(16)
$np_{1/2}$	δ_0	2.65456(15)	2.6548832(18)
	δ_2	0.388(60)	0.2902(16)
$np_{3/2}$	δ_0	2.64145(20)	2.6416721(18)
	δ_2	0.33(18)	0.2952(20)
$nd_{3/2}$	δ_0	1.347157(80)	1.34809171(86)
	δ_2	-0.59553	-0.60286(52)
$nd_{5/2}$	δ_0	1.347157(80)	1.34646572(75)
	δ_2	-0.59553	-0.59600(80)

Dipole-Dipole interactions

The dipole-dipole interactions between ultra-cold Rydberg atoms were first observed through resonant dipole-dipole energy transfer experiments by Anderson[1] and Mourachko[2].

In such an atomic system, referred as a "frozen Rydberg gas", the radius of Rydberg atoms is so large and the velocity of atoms is rather small, the atoms interact with a large number of other atoms without changing their position, much like the situation of interactions between atoms in solid. Such a system is really in the intersection of atomic physics and condensed matter physics.

In current research, we have showed some further evidence of the dipole-dipole interactions, which reinforce the interpretation to the experimental observation done by Anderson[1] and Lowell[5] in this lab. Moreover, we are trying to probe the dipole-dipole interaction directly by microwave spectroscopy.

Besides the importance in the study of ultra-cold collisions, this "frozen Rydberg gas" with strong dipole-dipole interactions has also been proposed for manipulating and implementing quantum process.[7][8]

Resonant Dipole-Dipole Energy Transfer[1]

At room temperature, resonant energy transfer occurs by a well-understood binary collision process, the linewidth depends on the collision velocity of the atoms but not on their density. But it's qualitatively different at 300μK, where the dipole-dipole interaction of the atoms approaches their thermal energy. In latter case, it is clearly not a binary process but a many-atom process, which has some similarities to the formation of energy bands in a solid.

The specific process has been studied is, for two atoms, the process

$$\text{Rb } 25s_{1/2} + \text{Rb } 33s_{1/2} \longrightarrow \text{Rb } 24p_{1/2} + \text{Rb } 34p_{3/2} \qquad (1)$$

As shown in fig.a below, it is resonant at the electric fields E = 3.0 and 3.4V/cm, where the $25s_{1/2}$-$24p_{1/2}$ and $33s_{1/2}$-$34p_{3/2}$ intervals are equal. Because of the splitting of $|m_j|=1/2$, and 3/2, levels of the $34p_{3/2}$ state, there are two resonances. Fig.b shows how the linewidth of resonances change as the number density varies. (Fig.a &b are taken from Ref.[1])

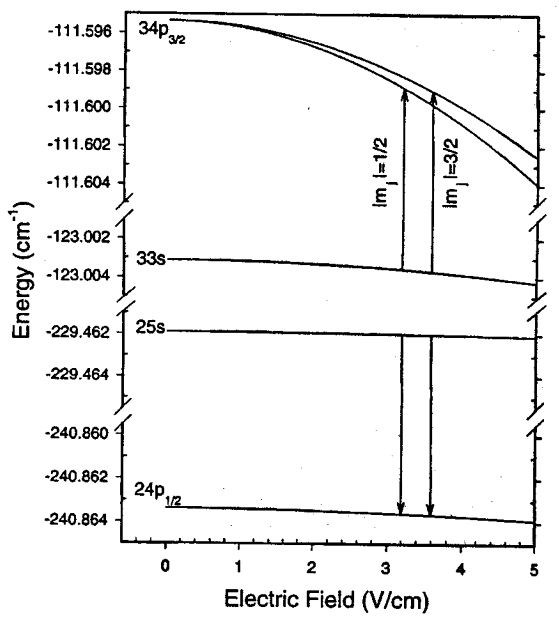


FIG. a Energy levels of Rb in an electric field showing the two energy transfer resonances of Eq. (1).

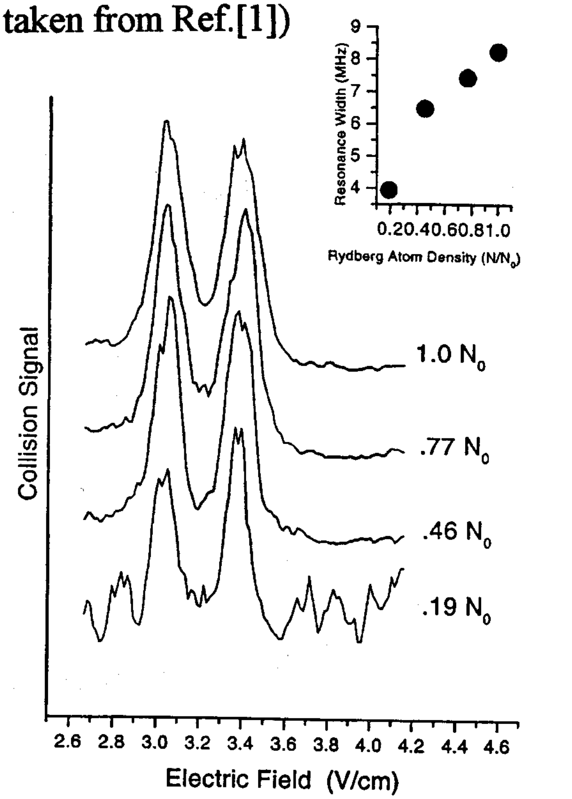


FIG. b The $25s_{1/2} + 33s_{1/2} \rightarrow 24p_{1/2} + 34p_{3/2}$ resonances at 300 μK observed in the MOT at four densities, $0.19N_0$, $0.46N_0$, $0.77N_0$, and N_0. The inset shows the width of the observed resonances vs relative density.

Interpretation

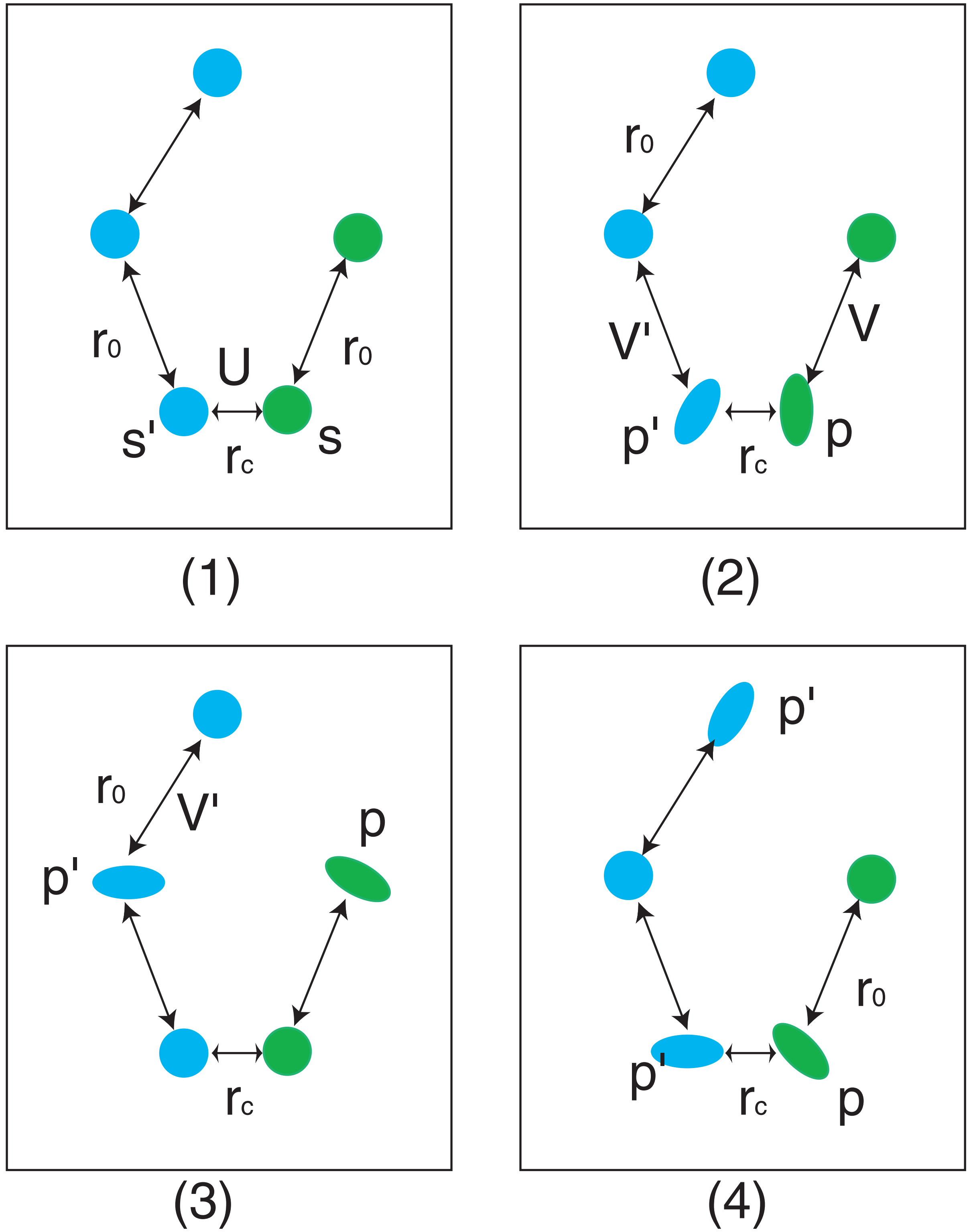
In the resonant dipole-dipole transfer process, there are three pairs dipole-dipole coupling.

$$\text{Rb } 25s_{1/2} + \text{Rb } 33s_{1/2} \longrightarrow \text{Rb } 24p_{1/2} + \text{Rb } 34p_{3/2} \qquad \text{(a)}$$
$$\text{Rb } 25s_{1/2} + \text{Rb } 24p_{1/2} \longrightarrow \text{Rb } 24p_{1/2} + \text{Rb } 25s_{1/2} \qquad \text{(b)}$$
$$\text{Rb } 33s_{1/2} + \text{Rb } 34p_{3/2} \longrightarrow \text{Rb } 34p_{3/2} + \text{Rb } 33s_{1/2} \qquad \text{(c)}$$

Eqation (a), (which is identical to Eq.(1) in page 12), is the dipole-dipole coupling responsible for the observed resonant energy transfer from the two s states to the two p states. It is only resonant at the fields E = 3.0 and 3.4 V/cm, and has the strength $\mu_A\mu_B/r^3$ (μ_A is dipole matrix element between $25s_{1/2}$ and $24p_{1/2}$, μ_B is dipole matrix element between $33s_{1/2}$ and $34p_{3/2}$), where r is the spacing between the two atoms. The dipole-dipole coupling of Eqs. (b) and (c) are resonant at all electric fields, have magnitudes $\mu_A{}^2/r^3$ and $\mu_B{}^2/r^3$.

If we only consider the dipole-dipole coupling from Eq.(a) for an average interatomic spacing, $r=r_0$, the typical dipole-dipole interaction $\mu_A\mu_B/r_0{}^3$ = 0.24MHz, which is much smaller than observed resonance width ~ 5MHz.

A qualitative description of the process, which reproduces the major features of the observations, starts with the assumption that the atoms are frozen in place. The cartoon in figure 8 is a simplified picture of this process. Some pairs of atoms are closely spaced by r_c, where $r_c < r_0$, have strong couplings (fig.8 (1)(2)), and are able to undergo the transition of Eq.(a) far off resonance, by up to $\mu_A\mu_B/r_c{}^3$, leading to the large observed linewidths. There are too few of these atoms, though, to give the magnitude of the signal observed. However, while in the p states, resonant energy transfer can occur to other 33s and 25s atoms, which are the mean distance r_0 away, by the process of Eqs. (b) and (c) (fig.8 (3),(4)), respectively. This transfer occurs at rates given approximately by $\mu_A{}^2/r_0{}^3$ and $\mu_B{}^2/r_0{}^3$, and slowly expands p state population outward from the close atoms, this can be considered as a diffusion process. The linewidth is determined by the coupling of the close atoms, and the time scale for the development of the signal and its magnitude are determined by the couplings due to the average spacing.



r_c : close pair, r_0 : average distance

s: 25s, p : 24p(1/2); s' : 33s, p' : 34p(3/2)

circles represent atoms in s states, while ovals in p states

U: A $/r^3$ V: A^2/r^3 V': $^2/r^3$

Figure 8

Broadening
From ns-np Dipole-Dipole Interactions

Following the above interpretation, we would expect to see the line broadening if we put some atoms in a state, which has a strong dipole-dipole coupling to one of the four states involved in the previous resonant energy transfer experiments, into the system.

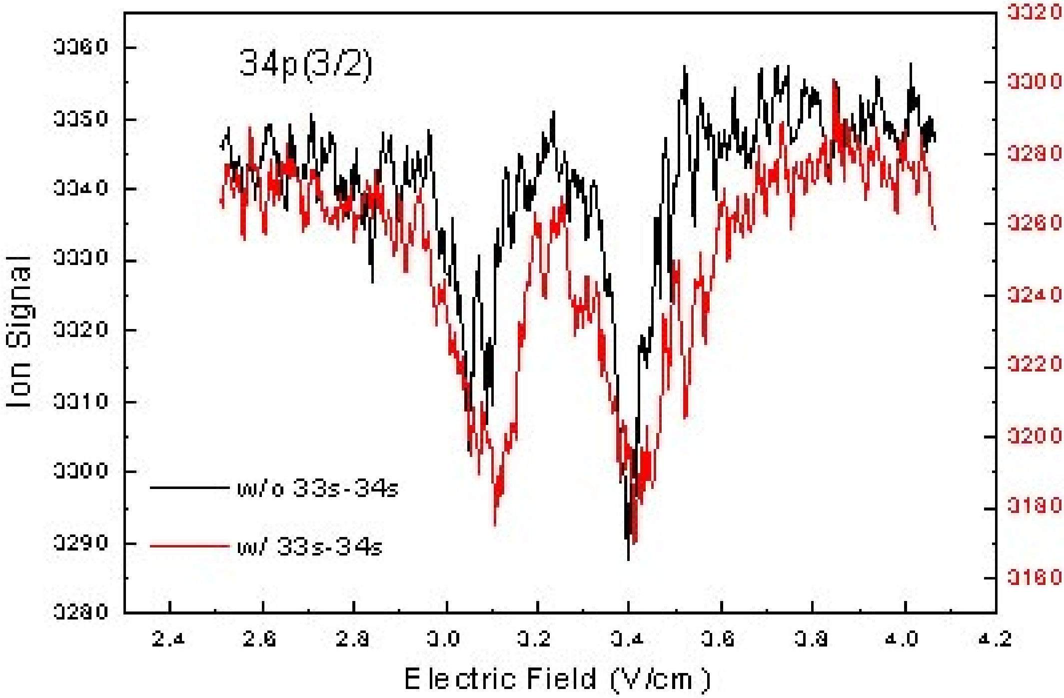
Figure 9 shows the experimental schemes and result for this experiment. 33s and 25s atoms are laser excited as before, though the two excitations are delayed by about 2μs. After atoms are excited into 33s states, a microwave pulse with duration ~1.5μs is sent into system to transfer some 33s population into 34s, which has very strong dipole-dipole interaction with $34p_{3/2}$, μ_C^2/r^3, (μ_C is the dipole matrix element between 34s and $34p_{3/2}$, which is about 8 times bigger than μ_B).

$$\text{Rb } 34s_{1/2} + \text{Rb } 34p_{3/2} \longrightarrow \text{Rb } 34p_{3/2} + \text{Rb } 34s_{1/2} \qquad \text{(d)}$$

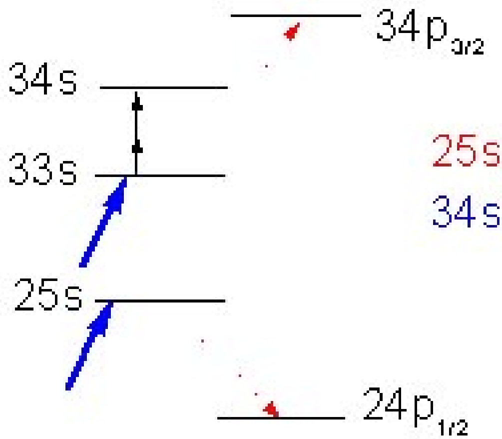
Populating 34s with microwave, instead of laser, greatly reduces the number of ions, which may cause some extra broadening, the signal is much less noisy as well. The microwave pulse comes before 25s population, so it wouldn't cause any power broadening. With the microwave power we have, maximum 10% 33s population can be transferred to 34s. An electric field pulse turn the system to resonance while 25s laser is fired.

The spectrum shows the broadening effects caused by introducing the 34s population into the system, even through the 33s population has been reduced by the s-s microwave transition. The cartoon in figure 10 demonstrates the ns-np dipole-dipole interaction process in the same way as seen in figure 8. In this process, the diffusion of $34p_{3/2}$ states from the close pair (fig.10(1)&(2)) is accelerated due to the presence of 34s and the strong coupling between them (fig.10(3)&(4)), more p state atoms will be produced even at off-resonance electric field, therefore we observed the broadened resonance. The observation agreed with the interpretation of dipole-dipole interaction.

Broadening From ns-np Interactions

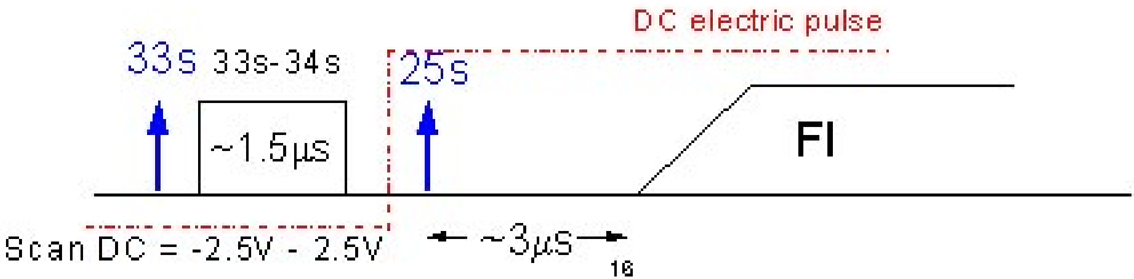


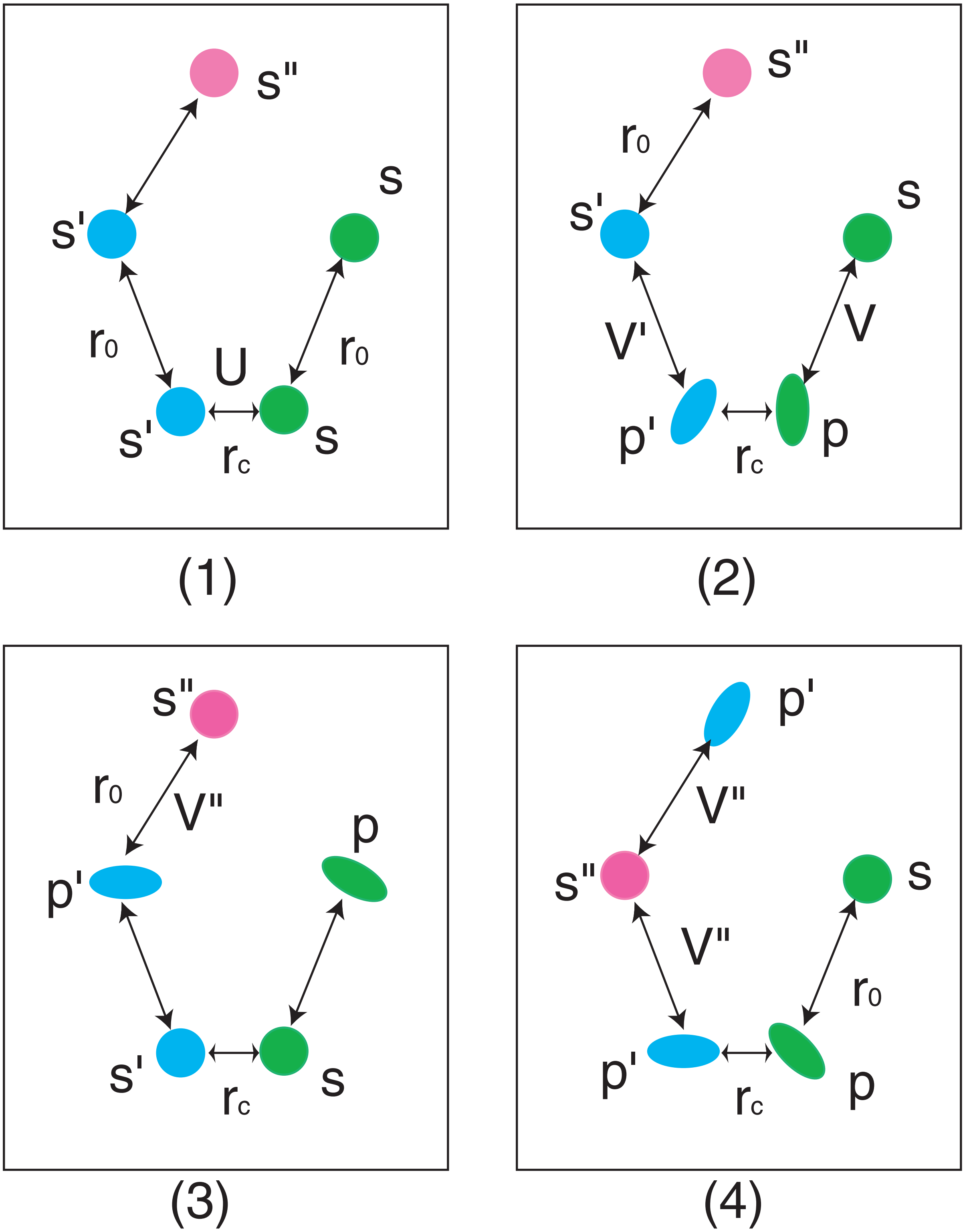
(at E=3.4V/cm , 1V/cm = 53MHz)



$25s_{1/2} + 33s_{1/2} - 24p_{1/2} + 34p_{1/2}$

$34s_{1/2} + 34p_{3/2} - 34p_{3/2} + 34s_{1/2}$





r_c : close pair, r_0 : average distance

s: 25s, p : 24p(1/2); s' : 33s, p' : 34p(3/2), s":34s

circles represent atoms in s states, while ovals in p states

U: $_A$ $/r^3$ V: $_A{}^2/r^3$ V': $^2/r^3$ V": $_c{}^2/r^3$

Figure 10

Conclusions & Future work:

1. We have measured resonant frequencies with high precision and improved quantum defects by an order of magnitude.
2. We have done experiments, which strongly support the dipole-dipole interaction picture in resonant energy transfer process in a “frozen” Rydberg sample. Several other schemes have been brought up for further studying the dipole-dipole interactions in such systems.
3. We have explored the possibility to use microwave as a probe to detect many body effects. The study of dipole-dipole interactions and ultra-cold plasma by directly using microwave spectroscopy is underway.

Reference:

[1] W.R.Anderson, J.R.Veale, and T.F.Gallagher, *Phys.Rev.Lett*. **80**, 249 (1998)

[2] I.Mourachko, D.Comparat, F.de Tomasi, A.Fioretti, P.Nosbaum, V.M.Akulin, and P.Pillet, *Phys.Rev.Lett.* **80**, 253 (1998)

[3] M.P.Robison, B.Laburthe Tolra, Michael W. Noel, T.F.Gallagher, and P.Pillet, *Phys.Rev.Lett.* **85**,4466 (2000)

[4] E.Rabb, M.Prentiss, A.Cable, S.Chu, and D.Prichard, *Phys.Rev.Lett.* **59**, 2631 (1987)

[5] J.R.Lowell, *Dipole-Dipole Interactions in a Frozen Rydberg Gas*, PhD thesis, University of Virginia,1998

[6] T.F.Gallagher, *Rydberg Atoms*, Cambridge University Press, 1994

[7] D.Jaksch, J.I.Cirac, and P.Zoller; S.L.Rolston; R.Cote and M.D.Lukin, *Phys.Rev.Lett.* **85**, 2208 (2000)

[8] M.D.Lukin, M.Fleischhauer, and R.Cote; L.M.Duan, D.Jaksch, J.I.Cirac, and P.Zoller *Phys.Rev.Lett.* **87**, 037901 (2001)