CHAPTER 3

Rydberg State Revisited for Deterministic Single-atom Source

This chapter presents our proposed mechanism for loading single rubidium atom in an optical dipole trap. The chapter begins, in section 3.1, with an illustrative picture of process for preparing trapped single atom by exploiting the long-range interaction of a Rydberg atom and a ground-state atom. It gives a qualitative understanding how the mechanism strongly induces one-body collisional loss that plays an important role in the single-atom loading achievement. Section 3.2 details the development of experimental conditions that the mechanism comes into operation. These conditions are needed for designing a single-atom trap experiment. In section 3.3, the model of light-assisted collision via two-photon excitation was used to evaluate the possibility in using a molecular Rydberg state of Rb_2 to load single rubidium atom in an optical dipole trap.

3.1 Single-atom loading via light-assisted Rydberg-ground collision

The mechanism for loading single atom is shown in Figure 3.1 and described as following. Suppose an atom is a three-level system; ground state, intermediate state, and Rydberg state. Let atoms be initially loaded into a far-off-resonance optical dipole trap. The interaction between a Rydberg atom and a ground-state atom is represented by a long-range adiabatic potential energy curves that includes both metastable molecule states and repulsive potential. The application of two-photon transition consisting of 780nm and 480nm photons, of which combined frequency is slightly higher than the resonance frequency between the ground state and preselected Rydberg state (measured on asymptotic lines in non-interaction regime), would give raise to the induced collision between ground and Rydberg atoms through the repulsive potential curve. The Condon point R_c is defined as the internuclear distance where the two-photon transition is on a resonance. Since the Rydberg atom only senses weakly repulsive dipole force [39] and

the ground-state atom is tightly pined by the trap, the Rydberg atom tends to slowly drift out from the trap region. Due to its long lifetime, the whole process of collision has completed before the spontaneous emission and hence the *one-body collisional loss* is significantly boosted. As time passes, such collisional loss events continue until there is only one atom left in the trap; hence the determinism of loading a single atom.



Figure 3.1: a) Potential energy curves of Rydberg-ground interaction and intermediateground interaction as function of internuclear distance (orange and black-gray curves respectively). The frequency of 780nm light is far blue-detuned from the intermediate transition by Δ to prevent real excitation to the intermediate state in which the two-photon excitation is influenced. The intermediate-ground state and Rydberg-ground state are coupled by 480nm light. The small blue detuning δ of total frequency is selected such that the excitation takes place whenever two colliding atoms come close at R_c distance. b) Energy level of an atom in optical dipole trap. Considering an atom of a colliding atomic pair excited to the Rydberg state (blue circle), it is not confined by the dipole force any longer and goes away. Hence the other atom (purple circle) remains. Many cycles of this one-body collisional lost process will end at the situation there is only one atom left.

3.2 Loading constraints

Before going further to the practical detail of a single-atom trap experiment, there are two parameters needed to be introduced. First, a parameter called *escape distance* D_{es} is defined as the minimum distance for a particular trap potential that a Rydberg atom with

total energy E needs to move from the center of trap to the point where after decaying to the ground state the residual kinetic energy is still high enough for exiting the trap. This distance can be calculated from

$$D_{es} = w_o \sqrt{\ln\left(\sqrt{\frac{2U_o}{mv_{mp}^2}}\right)},\tag{3.1}$$

where U_o and w_o are trap depth and 1/e radius of trap potential. v_{mp} is the most probable velocity given by Maxwell-Boltzmann statistics and it depends on temperature of atomic ensemble. Second, it is called *drift distance* D_f and defined as the maximum distance that a free Rydberg atom with lifetime τ_o in an atomic ensemble having temperature of Tcan move from a point to an another point. The analytic expression of this distance is

$$D_f = \sqrt{\frac{2k_BT}{m}}\tau_o,\tag{3.2}$$

where m is the rest mass of an atom. In order to reach the situation where the light assisted one-body collisional loss dominates over any loss processes, the drift distance must be longer than the escape distance,

$$D_f > D_{es}.\tag{3.3}$$

This condition ensures that when a ground-state atom is excited to the Rydberg state the atom will escape the trap after it decays to the ground state.

From the practical point of view, the strength of repulsive interaction between Rydberg atom and ground-state atom fundamentally decreases as principle quantum number n increase. This relation limits the range of available blue detuning δ that can be selected to determine a Condon point R_c . Moreover, smaller δ means the total frequency of excitation fields is closer to single atom resonance. Hence the probability of one-body loss induced by one-body excitation, instead of by induced collision, is higher. This causes a major problem when there is only one atom left in the trap in the presence of excitation laser field. The atom has a chance to be excited to Rydberg state and then drifts out from the trap, hence single-atom loading efficiency will strongly decrease. Hence there are two crucial conditions that must be satisfied in order to prevent such problem. First, the principle quantum number n must be small in the way that corresponding barrier potential is strong enough in which the detuning δ can be chosen significantly larger than the one-body scattering rate of two-photon transition. Second, to satisfy Eq.(3.3), the trap dimension, w_o , must be smaller than the drift distance determined by the lifetime of the Rydberg state of interest. However, the latter condition is automatically satisfied in a standard 3D optical lattice experiment since the dimension of single lattice site is in order of sub-micron while typical values of drift distance of cold Rydberg atom, having principle quantum number between 30 to 50, lie between 1 μ m to few tens μ m.



Figure 3.2: Escape distance D_{es} is the minimum distance that the Rydberg atom needs to move for escaping the trap.

In addition to the two presented condition, the density is an important factor needed to be concerned. If the Condon point R_c is larger than averaged separation distance between two adjacent atoms in the trap, at a particular high density, it has a chance that a colliding pair will be excited to a metastable bound state (shaded area in Fig.(3.1)) and then the whole process gets out of control. To fix this problem, the frequency of 480nm light must be precisely tuned to a preselected Rydberg state, of which interaction with a ground state atom has the same Condon point. Therefore an additional required condition is that the density of atomic ensemble must be prepared in which the average separation distance between atoms is larger than the selected Condon point.

3.3 Analysis of single-atom loading probability

The possibility of single-atom loading is evaluated through the probability of occurrence of one-body collisional loss compared to other processes including elastic collision, no collision, two-body collisional loss, and one-body loss via one-body excitation. In principle, to fully characterize the dynamic of cold collision between atoms in light field, one need to treat the problem quantum mechanically using the scattering theory. To simplify the problem, we treat the motion of two colliding atoms in an optical potential classically but excitation semi-classically using the Landau-Zener model presented in Chapter 2. Let define five possible scattering processes relevant in the determination of single-atom loading efficiency as follow:

- 1. One-body collisional loss: collision takes place and one of two colliding atoms escapes the trap after collision finished. If this scattering process dominate, higher probability of loading single atom. This channel is denoted by D(2|1).
- 2. *Two-body collisional loss*: collision takes place and both of two colliding atoms escape the trap after collision finished. This scattering process reduces the efficiency of single-atom loading. This channel is denoted by D(2|2).
- 3. *Elastic collision*: collision takes place and both of two colliding atoms still are in the trap after collision because the kinetic energy of each atom dose not change. This channel is denoted by D(2|0).
- 4. *No collision*: collision does not take place, denoted by D(2|N).
- 5. One-body loss via one-body excitation: an atom can gain additional kinetic energy by scattering with near or on resonance photons. This process heats atomic ensemble and cause trap loss, hence it reduces efficiency of single-atom loading. This process is denoted by D(1|1).

Clearly that the process D(2|1) needs to strongly dominate over the other processes especially D(2|2) and D(1|1) in order to achieve the determinism of single-atom loading. To look for a possibility under practical conditions, let consider the light-assisted collision between a rubidium Rydberg atom in $35D_{5/2}$ and a ground-state rubidium atom in $5S_{1/2}$ in an optical dipole trap with 10MHz trap depth. Assuming atomic ensemble in the trap is in thermal equilibrium and has temperature of 65 μ K. The approximated repulsive adiabatic interaction potential of $5S_{1/2} + 35D_{5/2}$ is shown in Fig.(3.3). This potential was calculated by using *e*-*B* scattering length presented in Chapter 2. The state $5P_{3/2}$ is the intermediate level in this case. Throughout the discussion, the condition Eq.(3.3) is assumed to be satisfied since it can be achieved in typical dipole trap experiment without special effort.



Figure 3.3: Approximated repulsive semi-molecular potential of $5S_{1/2} + 35D_{5/2}$.

One-body excitations of both $5P_{3/2}$ and $35D_{5/2}$ contribute to the occurrence of D(1|1) process. At first glance minimizing the occurrence of D(1|1) caused by excitation of $5P_{3/2}$ can be done by setting far detune Δ from D2 line resonance. However, Eq.() implies that if Δ is too large, the two-photon excitation rate will be significantly reduced, hence occurrence of D(2|1). To find an optimum range of Δ , it is set to be an independent variable that all possible occurrences are plotted as function depending on it. For the contribution from one-body excitation of $35D_{5/2}$, the two-photon detuning δ is thus chosen to be 3 MHz above asymptotic line as shown in Fig.(3.3). This is a reasonable value because the power broadening of two-photon transition of isolated atom is only around 770 kHz, hence low one-body loss via one-body excitation can be expected. In addition, the

detuning δ of 3 MHz is less than the trap depth and hence excitation at the Condon point does not induce two-body collisional loss and also whenever a colliding pair is excited to the repulsive region, one can ensure D(2|1) process is forced to take place.

The source of D(2|2) arises from the light-assisted collision between an atom in $5S_{1/2}$ and its colliding pair in $5P_{3/2}$ state. The collision can be induced by 780 nm light whose detuning Δ is blued from resonance, in this case it is 100 MHz. The dipoledipole interaction of $5S_{1/2} + 5P_{3/2}$ can cause energy shift in which the frequency of detuned 780 nm becomes on resonance of semi-molecular potential. Since the detuning of 100 MHz is much larger than the trap depth, this type of collision causes D(2|2). However both D(2|1) and D(2|2) take place at different Condon points, R_{C1} and R_{C2} respectively. Since R_{C1} is normally longer than R_{C2} , the occurrence of D(2|2) can be strongly suppressed by preparing the atomic ensemble in a low density. Fig.(3.4) shows the probability distribution function of separation distance of the two adjacent atoms at density of 3×10^{11} cm⁻³. The proportion of a colliding pair having the separation distance in which the light-assisted collision of $5S_{1/2} + 35D_{5/2}$ takes place is higher than the proportion of $5S_{1/2} + 5P_{3/2}$. Therefore D(2|1) collision through $5S_{1/2} + 35D_{5/2}$ interaction dominates over D(2|2).



Figure 3.4: Distribution of inter-particle distance

Fig.(3.5) shows the approximated strength in arbitrary unit of occurrences of each scattering process as function of intermediate detuning Δ . This plot was obtained from

applying Landau-Zener model and taking into account that D(2|1) and D(2|2) occur at different Condon points. At detuning of 100 MHz, the occurrence of D(2|1) dominates over D(2|0), D(1|1), and D(2|2). D(2|N) process has the maximum strength but fundamentally it does not affect the efficiency of single-atom loading except the time used to switch on excitation lasers.



Figure 3.5: Occurrence strength of scattering processes as function of intermediate detuning. The gray shaded area covers the range of the detuning from 0 MHz to 80 MHz in order to indicate the safe range from one-body excitation that induces D(1|1).

3.4 Summary

Apart from all crucial conditions presented in this chapter, there however are advantages of our proposed mechanism over the process implemented in [14]. First, it does not need the large blue detuning, that may induces two-body collisional loss event. Hence the high probability of one-body collisional loss can be expected. Second, although the finetuning on the two-photon excitation is needed, the overall performance does not strongly depend on trap depth and all uncontrollable thermal parameters, e.g. energy shared between collision pair, do not determine the one-body loss event or single-atom loading efficiency. Therefore this mechanism can be applied to the preparation of optical lattices where each well is filled with just single atom.