# STIRAP Production of Rydberg Helium: Effects of Thermal Radiation and Level Multiplicity

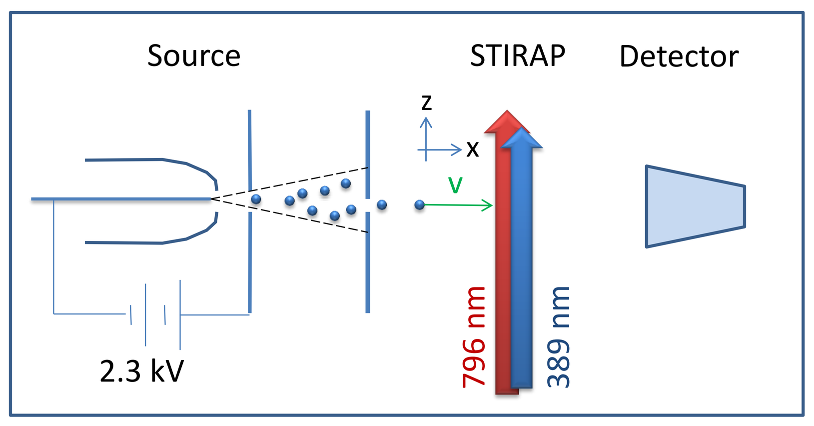
Petr M Anisimov, Yuan Sun, Harold Metcalf
*Stony Brook University, Stony Brook NY 11794-3800*

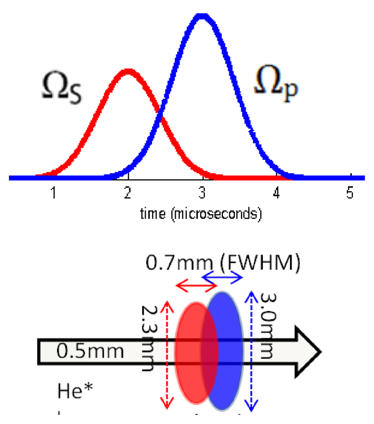
## Abstract

Stimulated Raman Adiabatic Passage (STIRAP) has been used in a series of experiments to excite Helium atoms to Rydberg states starting from the metastable $2^3S_1$ state. The usual picture of STIRAP in a three level system suggests that experimental efficiency should be nearly $100\%$, but our measured efficiency was limited to less than $70\%$. Here we report a detailed model of the STIRAP process in metastable Helium that accounts for the multilevel structure of the transition and effects of thermal radiation that lead to ionization as well as population redistribution among Rydberg states.

## Introduction

Helium atoms in a metastable state (He*) have high internal energy of 19.82 eV with a life time of 8000 s that could be exploited for neutral atom nanolithography [1] in order to create nanoscale structures through a resist assisted lithography mode [2]. Hence, a highly efficient production of Rydberg He* would allow for the use of inhomogeneous electrostatic fields to control atomic motion for the use in atom optics.
Here we consider the production of Rydberg Helium (He* with n=15 – 30) from a metastable $2^3S_1$ atomic beam that is produced from a reverse flow dc discharge source. Under steady running conditions, our source yields $\sim 0.5\times10^{14}$ He* atoms/(sr s) with an average longitudinal velocity about 1070 m/s and full width at half maximum of about 480 m/s.

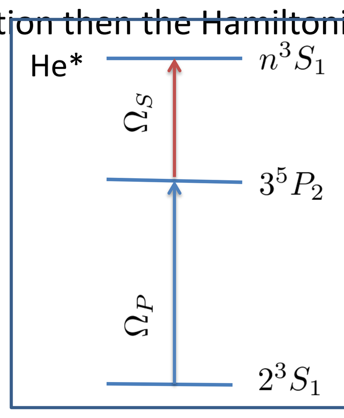



The excitation of He* atoms to Rydberg state is based on a two-photon mechanism known as Stimulated Raman Adiabatic Passage (STIRAP) in a three-level Ladder configuration. This approach allows one to use Ti:Sapphire laser at its fundamental "red" (780 – 830 nm) and doubled "blue" (389 nm) frequencies operated in a continuous mode. The atoms in the beam cross two laser beams and see the following electric field: $E=\sqrt{\frac{P}{\pi\epsilon_0 c\sigma_x\sigma_y}}\exp\left(-\frac{(x-x_0)^2}{4\sigma_x^2}-\frac{y^2}{4\sigma_y^2}+ikz\right)$,
where $P$ is laser power, $x_0$ is a relative position of the laser beam along the atomic beam with $x=v_xt$ and $z=v_zt$, and $\sigma=0.5w/\sqrt{2\ln2}$ in terms of the geometrical size of the beam measured full width at half maximum , $w$ . The characteristic parameters of our experimental realization are: $w_x=$ 0.5 mm, $w_y=$2 mm, and $P=$700 mW (50 mW) for red (blue) laser beams that correspond to Stokes and pump fields, which are arranged in counter intuitive order.

## Internal structure of He*

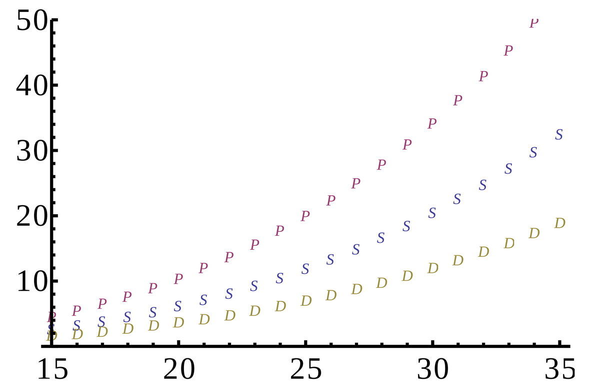
The Ladder system considered here starts at a metastable $2^3S_1$ state connected by the pump field to an intermediate state $3^5P_2$ which in turn is connected by the Stokes field to a Rydberg state $n^3S_1$. When, the pump field is frequency locked to the transition then the Hamiltonian is



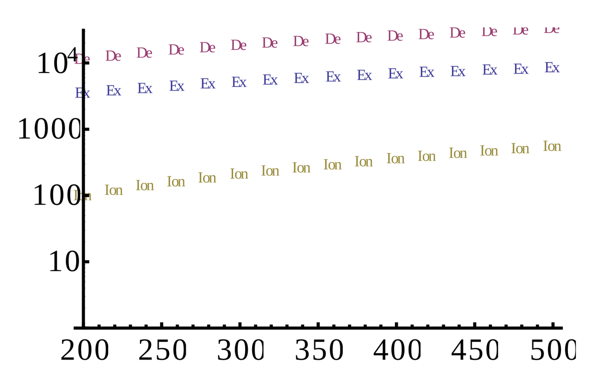

$$\hat{H}=\frac{\hbar}{2}\begin{pmatrix}0 & -\Omega_P^* & 0\\ -\Omega_P & 0 & -\Omega_S^*\\ 0 & -\Omega_S & 2\Delta\end{pmatrix}$$

The lifetime of the intermediate level is 94.8 ns due to the combination of direct and indirect radiative decays to the $2^3S_1$ state. The direct decay channel has Einstein coefficient $A=9.4746\times10^6$ s-1 distributed among magnetic sublevels according to the selection rules. Due to much faster decay rates on the subsequent transitions, the indirect decay channel $3^5P_2\to3^3S_1\to2^{2j+1}P_j\to2^3S_1$ could be effectively described by the decay rate of the first transition $A=1.0736\times10^6$ s-1 evenly distributed among magnetic sublevels regardless of selection rules. The radiative lifetime of Rydberg states as well as transition rates due to black body radiation for $24^3S_1$ Rydberg state are calculated based on results of Ref. [3].

Lifetime in $\mu s$ vs n for S, P and D states



Transition rates to adjacent Rydberg states vs T



## Two models

In order to describe STIRAP process in this system we developed two models. The first model ignores level multiplicity resulting in an effective three level system. The second model takes into account a multilevel nature of an actual He* atom that account for selection rules and allows to study effects of polarizaiton. In either case, the system evolution is governed by the following equation for the density matrix

$$\frac{d}{dt}\hat{\rho}=-\frac{i}{\hbar}[\hat{H},\hat{\rho}]+\sum_m\hat{L}(\hat{\sigma}_m)\hat{\rho}$$

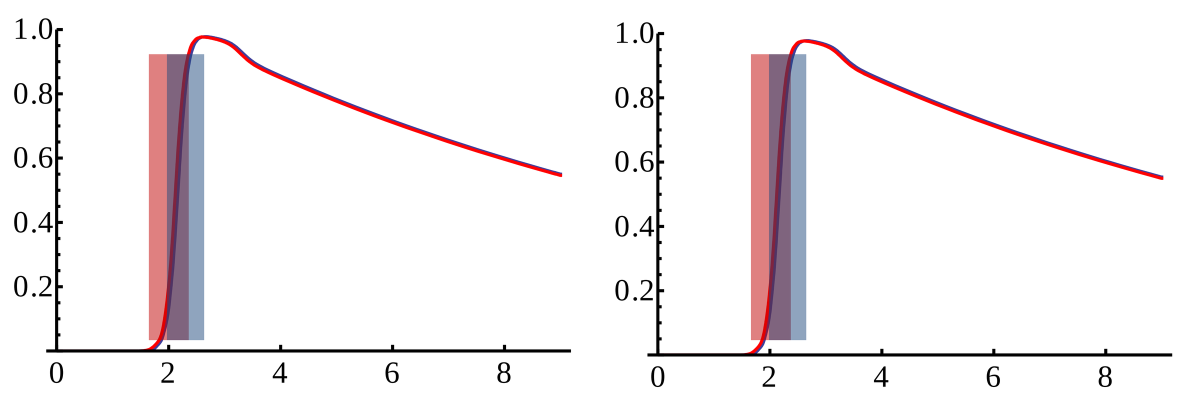
where the Hamiltonian depends on the Rabi frequencies of the corresponding electric fields, $E_{P,S}:\Omega_{P,S}=d_{i\to j}E_{P,S}/\hbar$, where reduced dipole moments of corresponding transitions are used, $d_{i\to j}=\sqrt{3\pi\epsilon_0\hbar(\frac{\lambda_{ij}}{2\pi})^3A_{i\to j}\frac{2J_j+1}{2J_i+1}}$, expressed through the Einstein coefficients for spontaneous decays $A_{i\to j}$ and transition wavelength $\lambda_{ij}$; where $\hat{L}(\hat{\sigma}_m)\hat{\rho}=\hat{\sigma}_m\hat{\rho}\hat{\sigma}_m^\dagger-0.5(\hat{\rho}\hat{\sigma}_m^\dagger\hat{\sigma}_m+\hat{\sigma}_m^\dagger\hat{\sigma}_m\rho)$ is a Lindblad operator for the m$^{th}$ decay channel described by the lowering operator $\hat{\sigma}_m=\sqrt{A_{i\to j}}\,|j\rangle\langle i|$.
Assuming linearly polarized light and quantization axis along the propagation vector, the Hamiltonian for multi level description has the following coupling matrixes:

$$\Omega_P\to\Omega_P\begin{pmatrix}\frac{i}{\sqrt{2}} & 0 & 0\\ 0 & \frac{i}{2} & 0\\ \frac{i}{2\sqrt{3}} & 0 & \frac{i}{2\sqrt{3}}\\ 0 & \frac{i}{2} & 0\\ 0 & 0 & \frac{i}{\sqrt{2}}\end{pmatrix},\Omega_S\to\Omega_S\begin{pmatrix}i\sqrt{\frac{3}{10}} & 0 & 0\\ 0 & \frac{1}{2}i\sqrt{\frac{3}{5}} & 0\\ \frac{i}{2\sqrt{5}} & 0 & \frac{i}{2\sqrt{5}}\\ 0 & \frac{1}{2}i\sqrt{\frac{3}{5}} & 0\\ 0 & 0 & i\sqrt{\frac{3}{10}}\end{pmatrix}^T$$

Energy separation between Rydberg states is small so that black body radiation is sufficient to redistribute population from the target Rydberg state. These additional states are combined as an effective level called "dump" that is populated through excitation and deexcitation rates. This level has dominant P character and decays to the ground state slower than S state.
There are seven decay channels in our system. Two decay channels for $3^5P_2$ have been already discussed. The total radiative decay rate for Rydberg state given in Ref. [3] has two main contributions: 1. direct decay to $3^5P_2$ state; and 2. indirect decay such as $n^3S_1\to2^{2j+1}P_j\to2^3S_1$ . Data from the NIST database suggest that only about 11.7% of the total decay rate goes to $3^5P_2$ channel thus providing us with an estimate for Einstein coefficient and reduced dipole moment of the Stokes transition. The remaining radiative channel is the decay of the dump level to the ground state. This decay as the other two indirect decays are assumed to have 1/3 probability to reach a magnetic sublevel of the metastable state. The final two decay channels are driven by the black body radiation and describe the exchange between the target Rydberg state and the dump level.

## Results for $v_z=0$

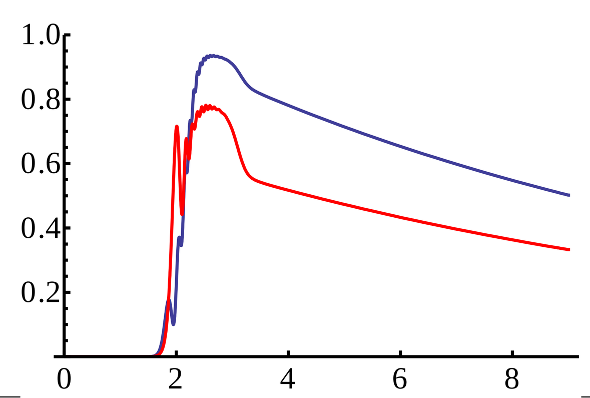
Efficiency of production of the Rydberg Helium is judged by the amount of He* atoms remaining in the ground state. Here is efficiency for two models. They seem to be identical (blue line for three level model matches red line for 11 level model well) and there is no noticeable effect of the temperature (left T=300K and right is T=500K).



Why can't we obtain similar production efficiency in experiments? Let us add Doppler effect due to non-zero component of atomic $v_z$.

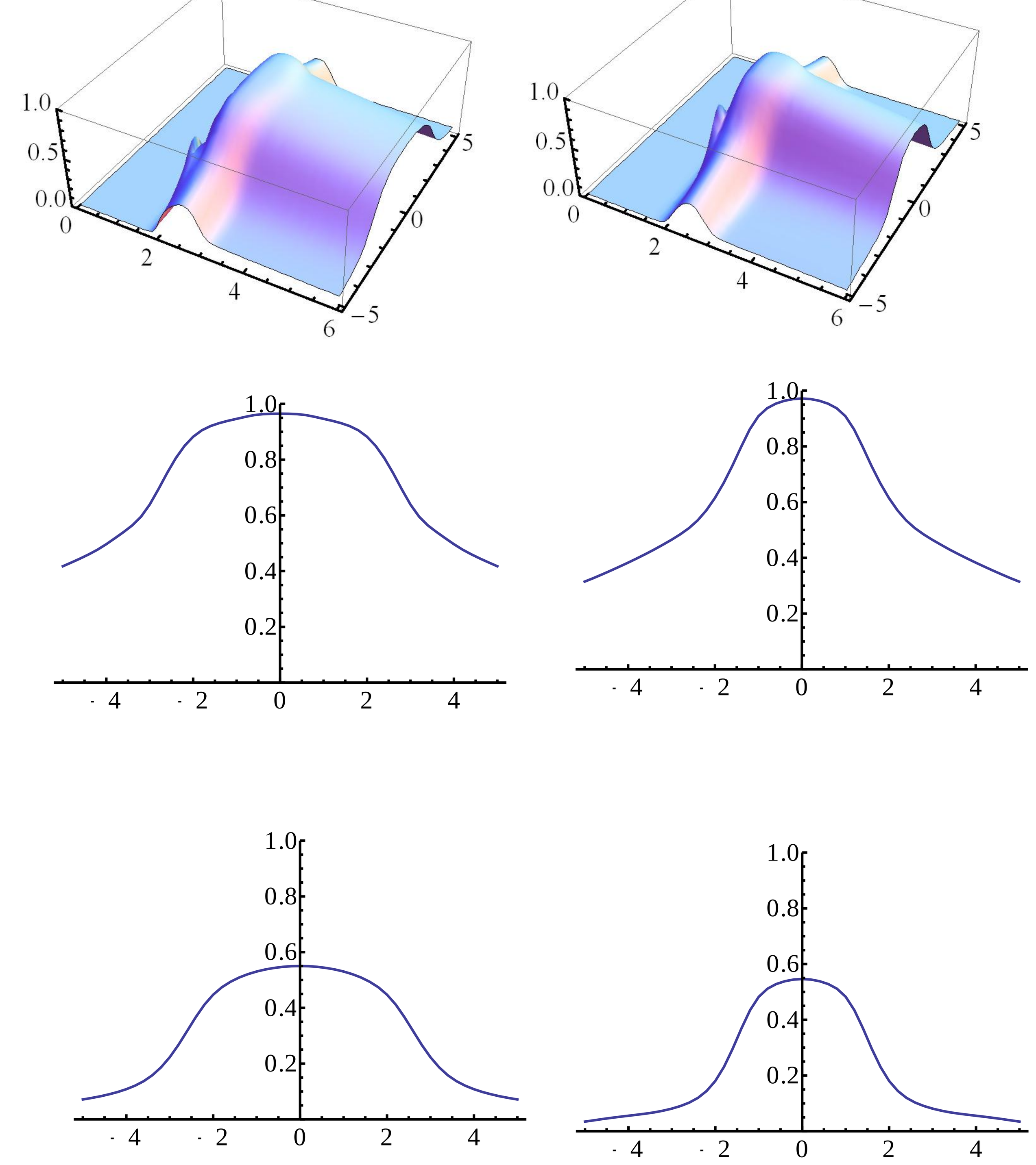
## Results for $v_z=1.5m/s$

Due to alignment issues or divergence of the beam, orthogonality is guaratied to about 1 mrad. It means that there will be always a Doppler shift on one photon as well as on two photon transition and due to Ladder nature of the scheme the Doppler shift on a two photon transition is a sum of the Doppler shifts for blue and red fields. Now there is a significant reduction in the production efficiency for the multilevel model:

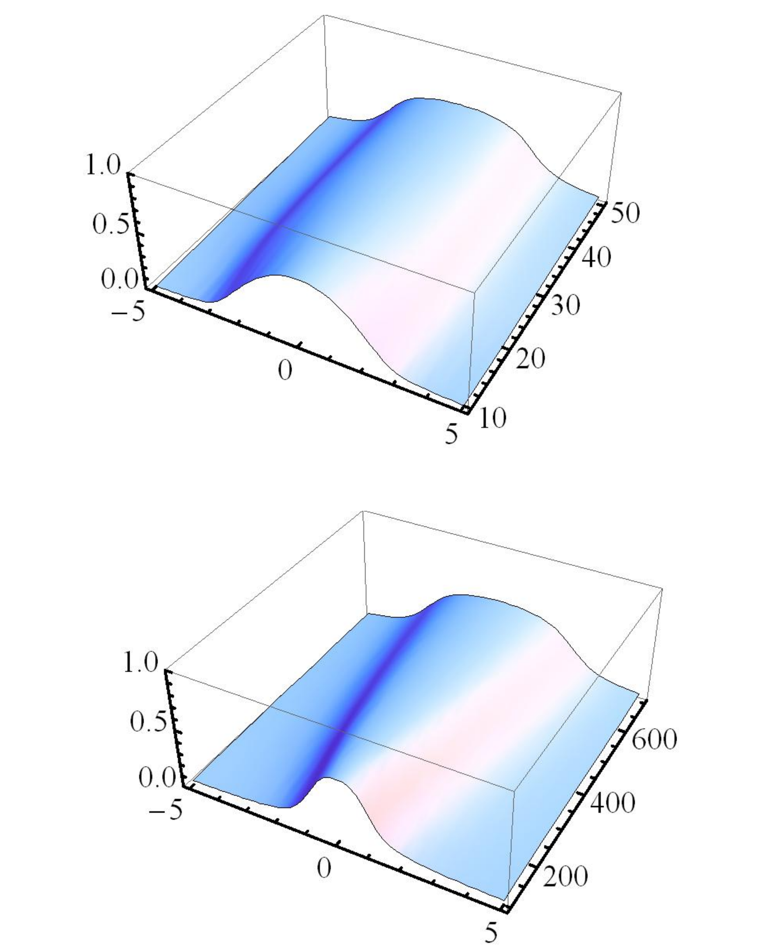


## Discussion

We look at the efficiency for different transverse velocities. One can see that at $v_z=0$ they match but the multi level model is less tolerable to the Doppler shifts resulting from transverse velocity.



The tolerance of a three level model is because effective Rabi frequencies used were higher. Here is a dependence of efficiency on the laser powers:



## Conclusion

We have presented two models that describe STIRAP production of Rydberg states in metastable Helium. Our models included effects of different decay channels, black body radiation as well as level multiplicity. Yet what we discovered is that the reason for low production efficiency was the presence of transverse velocity component that lead to the Doppler shifts. Efficiency increase is possible with utilization of higher power red laser.

## References

*[1] B. Brezger et al., J. Vac. Sci. Technol. B, **15**(6) p. 2905(1997).*
*[2] S. Nowak et al., Appl. Phys. B, **63** p. 203 (1996).*
*[3] I.L. Glukhov, J. Phys. B, **43** p125002 (2010).*

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