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Collisional Population Transfer in Yb Ions

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Abstract

The effect of population trapping in long-lived metastable states must be understood in order to achieve high precision in atomic frequency standards. We report here on transfer of Yb^+ ions into the long-lived $^2F_{7/2}$ state by means of collisions between He buffer gas and ions held in a linear Paul trap. Transfer rates were measured as a function of buffer gas pressure and repump laser power, and the collisional population transfer rates were extracted from the resulting data by solving the rate equations for the system. The measured transfer rate coefficients are $8.32(75) \times 10^{-11} cm^3/s$ and $8.65(33) \times 10^{-11} cm^3/s$ for the collisional processes $^2P_{1/2} \rightarrow ^2D_{5/2}$ and $^2D_{3/2} \rightarrow ^2F_{7/2}$, respectively.

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FIG. 1: A partial energy level diagram of $^{174}\text{Yb}^+$ showing the relevant clock transitions. Laser cooling and detection of ions is accomplished via the $^2S_{1/2}$ to $^2P_{1/2}$ at 370 nm.

I. INTRODUCTION

Trapped and laser-cooled ytterbium ions have been the subject of much experimental work in recent years. Singly-charged ytterbium ions are being used in quantum information studies[1] and in searches for variations in fundamental constants[2], but the bulk of work is toward more accurate atomic clocks. Of particular interest for frequency standards are the electric quadrupole transitions from the $^2D_{5/2}$ and $^2D_{3/2}$ [3] to the ground state but work is also being done on the electric octupole transition from the long-lived (6 yrs) $^2F_{7/2}$ to the ground state[4]. Fig. 1 shows the relevant energy levels.

The lifetimes of the two D states against electric dipole decay to the ground state have been measured [5–7] and the wavelength of the transition from the ground state to $^2F_{7/2}$ has also been measured[8]. Depopulation of the D states by collisions with ambient background gas is also of interest as this will affect the measured lifetimes and studies of this have also been done[5].

However, other collisional paths can play a role as well. We concern ourselves here with two pathways that, ultimately, leave the ions shelved in the $^2F_{7/2}$ state. This can proceed via collision-induced transfer from the $^2P_{1/2}$ state to the $^2D_{5/2}$ state followed by electric

dipole (E1) transition to the $^2F_{7/2}$ state or by E1 transition from the $^2P_{1/2}$ state to the $^2D_{3/2}$ state followed by collision-induced transfer to the $^2F_{7/2}$ state. Mixing of the two D states by collisions with buffer gas appears not to occur as this process was found not to occur in collisions between Sr ions and He[9], and the energy separation between the corresponding D states in Yb^+ is much greater than that in Sr^+ . The paths identified above have been addressed experimentally before[10], but those measurements may have been complicated by chemical reactions between the trapped Yb ions and background hydrogen gas[11].

II. EXPERIMENT

The studies reported here were conducted using isotopically-pure clouds of $^{174}Yb^+$ ions confined in a linear RF quadrupole trap. The trap comprises four, 1 mm-diameter, molybdenum rods in a rectangular arrangement with two diagonally-opposed rods having RF of several hundred volts amplitude applied and the remaining two rods being grounded. Confinement perpendicular to these rods is provided by this applied RF. Confinement parallel to the rods is provided by a DC voltage of a few tens of volts applied to tantalum tubes slipped over the ends of the grounded rods and insulated from them by thin alumina sleeves. The separation of these "endcaps" is 6 mm. A detailed description of this type of trap arrangement can be found in the literature[12].

The trap and Yb oven are enclosed in an ultrahigh vacuum system with base pressure of roughly 2×10^{-8} Pa maintained by a Varian Star-cell ion pump and SAES non-evaporable getter pump. Helium is passed through heated quartz into the vacuum system thereby producing ultra-pure buffer gas samples with operating pressures ranging from a 1×10^{-5} Pa to 1×10^{-2} Pa for the collision studies. During these measurements the ion pump was valved off from the system but the getter pump was left on thereby keeping such impurities as hydrogen and water to a minimum. Buffer gas pressures were measured by an ionization gauge, and the pressure readings were corrected for the decreased sensitivity of the gauge to He.

Loading of ions is achieved by multi-photon ionization of Yb atoms evolved from a heated tantalum tube filled with Yb foil. The resonant first step of this process at 399 nm provides the isotope selectivity with subsequent excitation to a high-lying Rydberg state enabling field ionization by the electric field produced by the trap RF. The atomic beam is collimated by

a series of 1 mm diameter apertures and is intersected at the center of the trap by the ionization beam. The collimation and 90° intersection angle serves to minimize the Doppler width of the resonant step, thereby maximizing the isotope selectivity of the loading. This method allows loading of isotopically pure samples of a few ions to a few times 10^5 ions. Although we could load any Yb isotope we wish with this method, we chose to work with ^{174}Yb ions for these measurements.

In addition to the photo-ionization beam, laser light at 370 nm is used to detect the ions while light at 935 nm and 638 nm can be used to remove ions from the metastable $^2D_{3/2}$ state and the long-lived $^2F_{7/2}$ state, respectively, thereby returning the ions to the detection cycle. An $f/1$ microscope objective collects 370 nm photons scattered by the ions and images them onto a photomultiplier tube (PMT), thus giving a time record of the ions' fluorescence rate.

The laser light at 935 nm and 638 nm are provided by commercially available laser systems from Sacher, while the 399 nm and 370 nm laser systems are based on Nichia diode lasers and were built at LANL. With the exception of the 399 nm photo-ionization beam, all beams enter the trap coaxially and at 45° to the trap symmetry axis. The photo-ionization beam enters the vacuum vessel through a different port to provide the isotope selectivity noted above.

III. RESULTS AND DISCUSSION

The rate of 370 nm photon scattering by the ions was recorded continuously as the ions were driven through the $^2S_{1/2} \leftrightarrow ^2P_{1/2}$ cycle by the 370 nm detection and the 935 nm and 638 nm repump laser beams. In the presence of buffer gas, blocking the 638 nm laser beam allows population to accumulate in the long-lived $^2F_{7/2}$ state thereby decreasing the number of ions in the detection cycle. This leads to a decrease in fluorescence at 370 nm, and the rate of the decay of this fluorescence is a measure of the rate of transfer of ions from the detection cycle to the long-lived state.

Fig. 2 shows typical response of the cloud fluorescence level to subsequent blocking and unblocking of the 638 nm repump laser beam, while Fig. 3 shows a plot of signal decay time, and hence the population transfer time, upon blocking the repump beam vs buffer gas pressure. Note that there is an overall slow loss ($> 50\times$ slower) of fluorescence signal even in the presence of the 638 nm repump light.

FIG. 2: Fluorescence rate from the trapped ions at 370 nm as the 638 nm repump beam is periodically blocked and unblocked. Helium buffer gas pressure for this data is approximately $3.3 \times 10^{-3} Pa$

This signal loss is probably due to reactions with hydrogen gas resulting in non-fluorescing ions, perhaps YbH^+ . Sugiyama and Yoda[11] surmised that the decay of signal observed by Bauch, *et al.*[10] was caused by just such a reaction and that the subsequent recovery of signal by slight detuning of the 370nm beam was actually evidence of photodissociation of the molecular ions rather than repumping of the ions from the $^2F_{7/2}$ state. This is clearly not the case in the measurements reported here due to the much lower energy of the repump photons. Furthermore, this wavelength corresponds to a known transition in Yb^+ ions, unlike the surmised, unknown repump transitions of Bauch, *et al.*. Also, we are able to shelve the ions in the $^2F_{7/2}$ state by driving the $^2D_{3/2}$ to $^1[3/2]_{3/2}$ transition at 860nm from which level the ions then decay to the F state. When this transition is excited, the ion cloud instantly goes dark and no fluorescence is observed until the ions are returned to the S-P cycle by the 638nm laser light.

Finally, fig. 3 clearly shows that the population transfer rate is linear with pressure indicating that the transfer occurs mainly, if not exclusively, through binary collisions. In light of all of this we attribute the decrease in 370nm fluorescence when the 638nm repump light is blocked entirely to transfer of population to the long-lived state through collisions

FIG. 3: Dependence of fluorescence signal decay rate upon blocking the repump beam on buffer gas pressure for 950 μ W (squares) and 200 μ W (circles) of 935 nm laser power.

between a neutral helium atom and an ytterbium ion. We can therefore fit the decay of the signal to an exponential to get a measure of the total transfer rate into the long-lived state.

Next, we solve the rate equations for this system in order to extract the transfer rates for each path of interest from the total transfer rate. We begin by noting that, in the presence of 638nm repump laser light, the transition rates between the $^2S_{1/2}$, $^2P_{1/2}$ and $^2D_{3/2}$ states during the detection cycle are much higher than the collisional decay into the $^2F_{7/2}$, and these three levels can be considered to have quasi-static populations determined by the optical pumping and decay rates. The rate equations for those populations are

$$\dot{\rho}_{s_{1/2}} = -B_{P,S} \cdot \rho_{s_{1/2}} + (A_{P,S} + B_{P,S}) \cdot \rho_{P_{1/2}} + B_{D,[3/2]} \cdot \rho_{D_{3/2}} \quad (1)$$

$$\dot{\rho}_{P_{1/2}} = B_{P,S} \cdot \rho_{s_{1/2}} - (A_{P,S} + B_{P,S} - A_{P,D}) \cdot \rho_{P_{1/2}} \quad (2)$$

$$\dot{\rho}_{D_{3/2}} = A_{P,D} \cdot \rho_{P_{1/2}} - B_{D,[3/2]} \quad (3)$$

Here, A_{PS} and A_{PD} are the spontaneous decay rates from the $^2P_{1/2}$ to the $^2S_{1/2}$ and the $^2D_{3/2}$ states, respectively, and B_{PS} is the optically stimulated rate for the P to S transition.

The $^2S_{1/2}$ and $^2P_{1/2}$ levels have the same degeneracy, so the driven $^2S_{1/2} \rightarrow ^2P_{1/2}$ is the same as the driven $^2P_{1/2} \rightarrow ^2S_{1/2}$ rate. We neglect the population in the $^3[3/2]_{1/2}$ state, since ions in that state decay quickly back to the $^2P_{1/2}$ state. In addition, the forbidden (E2) decay from $^2D_{3/2}$ to $^2S_{1/2}$ has been neglected since the rate for this transition ($\tau \simeq 53ms$) is very low compared to the 935nm repump rate.

Applying the steady state condition $\dot{\rho}_{S_{1/2}} = \dot{\rho}_{P_{1/2}} = \dot{\rho}_{D_{3/2}} = 0$, the equations can be solved to obtain the fractional population distribution

$$\rho_{S_{1/2}} = \frac{(A_{P,S} + B_{P,S} + A_{P,D})B_{D,[3/2]}}{(A_{P,S} + 2B_{P,S})B_{D,[3/2]} + A_{P,D}(B_{P,S} + B_{D,[3/2]})} \quad (4)$$

$$\rho_{P_{1/2}} = \frac{B_{P,S}B_{D,[3/2]}}{(A_{P,S} + 2B_{P,S})B_{D,[3/2]} + A_{P,D}(B_{P,S} + B_{D,[3/2]})} \quad (5)$$

$$\rho_{D_{3/2}} = \frac{B_{P,S}A_{P,D}}{(A_{P,S} + 2B_{P,S})B_{D,[3/2]} + A_{P,D}(B_{P,S} + B_{D,[3/2]})} \quad (6)$$

for which $\rho_{S_{1/2}} + \rho_{P_{1/2}} + \rho_{D_{1/2}} = 1$

While the ions continue to cycle through these states due to the action of the 638nm repump laser or due to the absence of collisions, the above steady state populations will remain unchanged and the ions will fluoresce at 370nm. The intensity of this fluorescence is proportional to $\rho_{P_{1/2}} \cdot A_{P,S}$. The effect of the removal of the repump light in the presence of collisions is a slow transfer of population from the S-P-D cycle into the $^2F_{7/2}$ state and a concomitant decay of the ion fluorescence. This can be treated as a slow leakage through the two pathways outlined in the introduction.

The leakage rate per ion can be written

$$\Gamma = (0.83) \cdot \gamma_{P,D} \cdot \rho_{P_{1/2}} + \gamma_{D,F} \cdot \rho_{D_{3/2}} \quad (7)$$

where $\gamma_{P,D}$ and $\gamma_{D,F}$ are the collisional transfer rates for $^2P_{1/2} \rightarrow ^2D_{5/2}$ and $^2D_{3/2} \rightarrow ^2F_{7/2}$, respectively. The coefficient 0.83 comes from the branching ratio for $^2D_{5/2} \rightarrow ^2F_{7/2} : ^2D_{5/2} \rightarrow ^2S_{1/2}$. About 17% of ions transferred to the $^2D_{5/2}$ state decay to the ground rather than the $^2F_{7/2}$. As this rate depends on $\rho_{P_{1/2}}$ and $\rho_{D_{3/2}}$, it depends implicitly on the 370nm and 935nm intensities at the ion's position.

Due to the cloud and laser beam geometries, only the central portion of the ion cloud is illuminated by the detection and repump beams, and we must therefore account for both the variable cloud density and beam intensities in our data analysis. We assume a fractional

density distribution given by

$$n(\vec{r}) = \frac{1}{\pi R_c^2} \frac{1}{\sqrt{\pi} Z_c} \exp \left[-\frac{|\vec{r}|^2}{R_c^2} \right] \exp \left[-\frac{|z|^2}{Z_c^2} \right] \quad (8)$$

with the property

$$\int n(\vec{r}) dV = 1.$$

Modeling of our trap geometry yields the values $R_c = 30\mu m$ and $Z_c = 950\mu m$.

The laser beam intensities can be represented by

$$I(\vec{r}) = I_0 \exp \left[-\frac{|\hat{k} \times \vec{r}|^2}{R_0^2} \right] \quad (9)$$

where $\hat{k}$ is the direction vector of the beams and R_0 is the beam radius at the focus. For the 935nm beam, the focal radius was $215\mu m$; for the 370nm beam it was $112\mu m$.

The 935nm and 370nm beams drive electric dipole transitions, so the transition rate is proportional to the intensity. This rate can be written

$$B(\vec{r}) = B_0 \exp \left[-\frac{y^2 + \frac{1}{2}(x-z)^2}{R_0^2} \right] \quad (10)$$

where the driving rate at the center of the trap is B_0 . This formula corresponds to a beam propagating through the trap in the x-z plane, rotated from the trap axis (z-axis) by 45° .

To find the driving rate at the center of the cloud, a very small ion cloud was loaded - much smaller than the foci of the two driving beams. This allowed us to temporarily neglect the intensity distribution of the beams. The 370nm fluorescent signal is proportional to the population in the $^2P_{1/2}$ state given by eq 6. As the 935nm power increases, the population in the $^2D_{3/2}$ state decreases, and the population in $^2P_{1/2}$ tends to

$$\rho_{P_{1/2}} = \frac{B_{P,S}}{(A_{P,S} + 2B_{P,S})} = \frac{\beta_{370}P_{370}}{(A_{P,S} + 2\beta_{370}P_{370})} \quad (11)$$

In this series of measurements, 500-600 μW of 935nm power into the trap was sufficient to clear the $^2D_{3/2}$ state. The 935nm power was held at 930 μW and the fluorescence measured as a function of 370nm power. This saturation curve was fit to eq 11 to obtain β_{370} , the proportionality constant between the input 370nm power and the $^2S_{1/2} \rightarrow ^2P_{1/2}$ driving rate. For this system, β_{370} was measured to be $2.44 \times 10^6 s^{-1}/\mu W$, giving a driven rate of $6.1 \times 10^8 s^{-1}$ at 250 μW of power into the trap.

A similar procedure was then performed for the 935nm driving rate. The 370nm beam was held at a constant $250\mu\text{W}$ and the 935nm power varied. The measured saturation curve was fit to eq 6 to obtain $\beta_{935} = 2.34 \times 10^3 \text{ s}^{-1}/\mu\text{W}$. At $930 \mu\text{W}$, the 935nm driven rate was $2.18 \times 10^6 \text{ s}^{-1}$.

Given the distributions of pumping rates and fractional cloud density within the trap, the total leakage rate can be calculated.

$$\Gamma_T = \int \Gamma(\vec{r}) dV \quad (12)$$

$$= (0.83) \gamma_{P,D} \int \rho_{P_{1/2}}(\vec{r}) n(\vec{r}) dV + \gamma_{D,F} \int \rho_{D_{3/2}}(\vec{r}) n(\vec{r}) dV \quad (13)$$

The integrals can be calculated for any combination of 935nm and 370nm beam powers, leaving the two free parameters $\gamma_{P,D}$ and $\gamma_{D,F}$.

In other words, one can calculate the relative weights for the two collisional pathways. In the experiment, the He buffer gas pressure is fixed and the $638\text{nm } ^2F_{7/2}$ repump blocked. The decay rate of the 370nm fluorescence signal is measured as a function of 935nm beam power. Figure 4 shows such a curve for a He pressure of 3.4×10^{-6} torr. A least-squares fit to this data gives $\gamma_{P,D} = 10.0(9) \text{ s}^{-1}$ and $\gamma_{D,F} = 10.4(5) \text{ s}^{-1}$.

Since the trapped cloud and buffer gas are in thermal equilibrium the measured rate of transfer of the ions into the long-lived state is an average of the energy-dependent cross section over the Maxwellian distribution of the ion-atom collision velocities. Hence, from these measured rates we derive rate constants $k_{P,D} = 8.32(75) \times 10^{-11} \text{ cm}^3/\text{s}$ and $k_{D,F} = 8.65(33) \times 10^{-11} \text{ cm}^3/\text{s}$. By comparison, the ion-induced dipole rate constant for collisions between helium atoms and ytterbium ions[13] is $k_L = 5.32 \times 10^{-10}$ which suggests that, on average, one out of every six collisions between a neutral atom and an ion in the relevant state results in transfer of that ion into the metastable state.

It is surprising that these rates are identical to within the measurement uncertainty when the P-D energy separation (360 meV) is roughly twice the D-F separation (190 meV). This issue remains unresolved at this point.

Finally, it is interesting to note that there is no evidence of quenching of the $^2F_{7/2}$ state by collisions with the neutral He gas: the ion cloud fluorescence drops to the background level when the 638 nm repump laser is blocked and remains there until the repump beam is unblocked. This is, perhaps, not surprising in view of the large energy separation between this level and the ground state.

FIG. 4: Fluorescence Signal Decay Rate vs 935nm Repump Power. The diamond points are measured values, while the squares are the fit points.

However, there is some evidence that collisions with other gases can lead to quenching of the metastable state. Specifically, after the gas handling manifold of the experiment had been exposed to atmosphere for a lengthy interval without subsequent baking, the fluorescence from the trapped ions dropped to roughly 50% upon blocking the 638nm repump laser. This is consistent with the establishment of equilibrium between collisional transfer rates into and out of the metastable state.

Residual gas analyzer scans showed significant amounts of water vapor and smaller amounts of nitrogen and oxygen. After baking the system, the concentrations of these other gases had dropped by at least an order of magnitude, and the fluorescence signal returned to its former mode of behavior. It is impossible to say which of the other gases was responsible for the collisional quenching, however the greater concentration of water vapor

combined with the intrinsic polarity of the water molecule makes it a likely candidate.

IV. CONCLUSION

From this data we can draw some basic conclusions as to acceptable levels of background gas concentrations in Yb ion experiments. The major residual gas in ultrahigh vacuum systems is hydrogen which, due to its larger polarizability, has an ion-induced dipole collision rate approximately three times the rate for helium. Assuming that the transfer probability per collision is the same as Yb-H₂ collisions as for Yb-He collisions, this implies a maximum hydrogen pressure on the order of 10^{-9} Torr for experiments requiring manipulations of single Yb ions lasting for several tens of seconds. Our own experience with crystals of a few ions showed that many of the ions would go dark in a few minutes in the absence of 638nm repump light with total background pressures of a few 10^{-10} Torr. Fortunately, pressures an order of magnitude lower are achievable with standard ultrahigh vacuum techniques.

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