Development of a trapped Yb^+ frequency standard: Efficient quenching of population trapping

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We report on a study to determine the efficiency of various buffer gases in quenching the population trapping states of laser excited ytterbium ions. The ions were confined in an rf/dc hybrid trap, and their $^2S_{1/2}$ { } $^2P_{3/2}$ transition was excited wit h 329 nm la ser light. Buffer gases used include H_2, N_2, CO_2, Ar, and 1 Ie at several different pressure regimes. We have identified N_2 to be highly efficient in quenching the population of the trapping states, and thus be the most suit abl e buffer gas to use with ytterbium for the development of a trapped ion microwave frequency standard.

The 171 isotope of ytterbium is a particularly attractive choice for trapped ion frequency stan idards applications. This ion has a nuclear spin I= 1/2 and thus a simple hyperfine level structure with an energy splitting of 12.64 GHz. The lower lying excited electronic states of singly ionized ytterbium are accessible from the ground state with light generated by frequency doubled semiconductor lasers [1 ,2]. This is a particularly useful feature of the ytterbium ion in connection with the development of practical standards.

The major ob stacle in the develop ment of a frequency st andard based on the ytterbium ion is the existence of population trapping states. These metastable States prevent the ion from making the desired clock transition by retaining it in an energy eigenstate for a time corresponding to the lifetime of the eigenst ate. The presence of low lying metastable levels in the electronic structure of the ytterbium ion has been the subject of much study and concern [3 8]. Such long lived states can "trap" the excited population of ions involved in the optical pumping of the clock transition. The observation of a "dark" state with estimated lifetime exceeding several (lays has been associated w it h populat ion trapping in the $4f^{13}6s^2\,^2F_{7/2}$ stat e [7] and has been examined in several investigations [1 ,4,5,9]. Initial studies of the F state have resulted in some manswered questions concerning the mechanism for its population; these studies have not identified an efficient sch eme to de- excite the ions back to the ground state. Unt il now the only effective approach to avoid population trappi ng in the "dark" state involved using additional lasers to depopulate the metastable D-states [3 5,9 1 1], which are believed to be the intermediary path to the excitation of the F state.

In this paper we examine the efficacy of several different buffer gases (including 11$_2$, N_2, CO_2, He, and Ar) in quenching the populat ion of the "dark" state in the ytterbium ion. Nitrogen is found to be extremely efficient at quenching the dark state with an estimated quenching rate of 3.78x1 0^4/sec/Pa for pressures less than 2.66×10^{-4} Pa. While the influence of nitrogen on the depletion of" the *population* of the "dark" state will be the primary focus of this paper, we will also present results obtain ed with other gases wh ic point to a possible path for population trapping due to molecular formation with laser excited ions.

Buffer gas studies were performed with naturally abundant ytt erbium, as well as with isotopically enriched ytterbium 171 ions confined in a hybrid linear rf/dc trap [1 2]. The isotopically enriched ytterbium 171 st udy was performed to examine the effect of collisions with the buffer gas on the col erence of t he microwave transition between the hyperfine levels. The trap is housed in a stainless steel vacuum chamber evacuated to a background pressure below 7×10^{-7} Pa. 11$_2$, N_2, CO_2, He, and Ar were introduced from a manifold connected with a high precision sapphire variable leak to the vacuum chamber

Ytterbium ions were generated by collision of counter-propagat ing beams of ytterbium atoms from a tantalum tube, and electrons from a hot filament. The beam di - rections were perpendicular to the trap axis. During ion loading a background buffer gas pressure of 6.7×10^{-4} I's, as read on the ion gauge, was maintained. Once a sufficiently large cloud of ions was loaded ($\sim 10^6$ both the elect ron b eam and the atom beam were tur ned off. After a short relaxation time the pressure was reduced to a 6.7×10^{-5} Pa and the steady state fluorescence was measured as a function of buffer gas pressure.

The fl uorescence measurements were made by exc - ing the $4f^{14}(^1S)6s^2S_{1/2}$ <> $4f^{14}(^1S)6p^2P_{3/2}$ transition using 328.9 nm IIISCI' radiation directed along the trap axis The fluorescence rate from the decay of the excited $4f^{14}(1S)6p^2P_{3/2}$ level was monitored perpendicular to the trap axis, The fluorescence was monitored for 30 seconds, followed by blocking t he laser for an ot he r 30 seconds inter val; during this time the buffer gas pressure was manually adjusted from 6.7 x 10^{-5} to 1.3 x 10-4 Pa. After this period the laser was unblocked again and

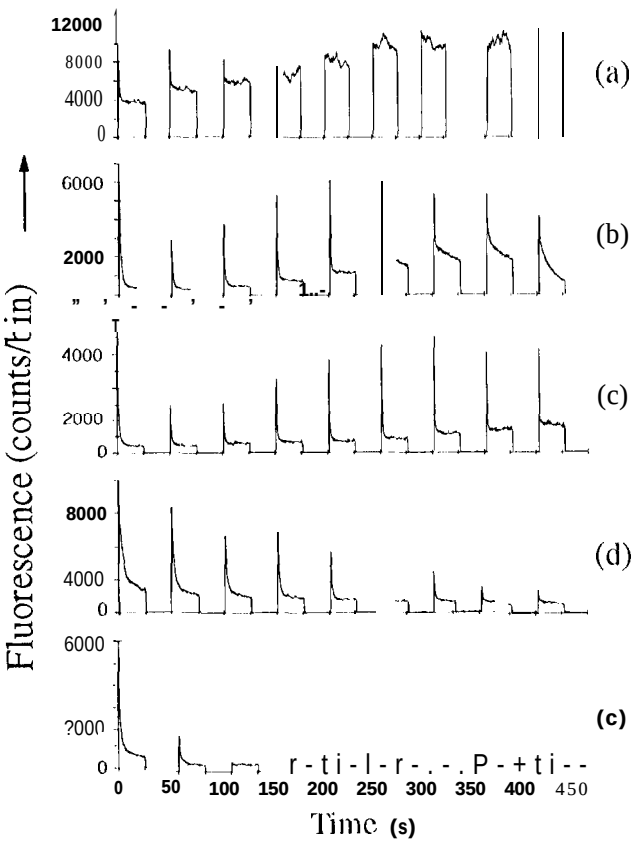

FIG. 1. Steady state fluorescence spectrum for (a) nitrogen, (b) helium, (c) argon (d) carbon dioxide arid (e) hydrogen. Fluorescence rate was recorded on a multichannel scaler with a 110 msec bin width.

fluorescence vs. time was monitored for another 30 second interval. This procedure was continued until the fluorescence rate was measured for each of the following nine buffer gas pressures ($6.7\times$ 10-5, I.3X 10-4, 2.7 $\times 10^{-4}$, 6.7x 10-4 1.3X 10^{-3}, 2.7 x 10^{-3}, 6.7×10^{-3}, 1.3×10^{-2}, 2.7 X 10^{-2}) Pa. 011((e the steady Still(. fluorescence measurement was completed the ions were re-loaded, to replace the ions lost at very high buffer gas pressures, and a measurement of the ion temperature was performed by scanning the laser 40 GHz at a rate of 7.63GHz/s to obtain the Doppler profile of the transition. The temperature measurements were performed at the same nine buffer gas pressures, each measurement comprising of 50 scans summed to improve the signal to noise ratio. The resonance spectrum obtained in this manner was then fitted to a sum of five Voight profiles, corresponding to the five dominant peaks present in naturally abundant ytterbium, using a commercial curve fitting program. Using the ratio of the peak fluorescence together with the temperature measurements a quenching rate for the "dark" state can be determined.

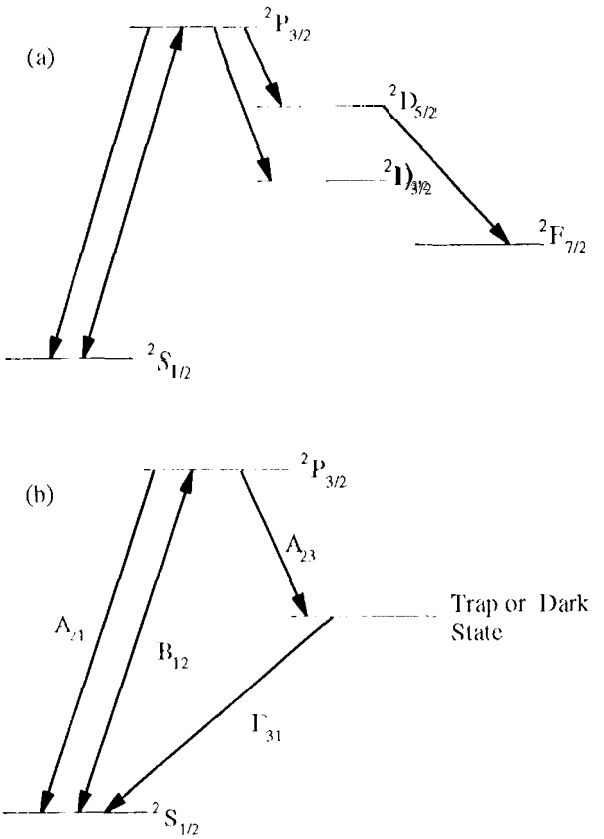

FIG. 2. (a) Partial energy level diagram showing Yb^+ levels involved in present study. (b) Further simplification where the D and F states have been combined into one state which we call the "dark" or "trap" state.

Steady State fluorescence and resonance spectra were obtained for each of the five buffer gases examined in this study. Fig. 1 show the fluorescence measurements for all five buffer gases, It is clear from the figure that N_2 is the most efficient of the buffer gases studied in its ability to quench the "dark" Still (. In order to obtain a more quantitative measurement of the influence of N_2 on the ytterbium ion, fluorescence vs. time measurements were repeated with higher resolution and at lower pressures. Fifteen values of N_2 pressures between 6.7×10^{-5} and $1.3\text{X}10^{-3}$ P?I were used to study its quenching efficacy.

Fig. 2(a) shows a partial energy level diagram for Yb^+ and2(b) Silo\!'S a further simplification where we have combined the $^2D_{3/2}$, $^2D_{5/2}$, and $^2F_{7/2}$ metastable states into a single "dark" state. This])roc('(lure is justified since the "dark" state is presumed to be resulting from population trapping via a mechanism that involves the 1) states, or the F state which is populated through the D states, or the combination of all three states.

Using the oscillator strengths and branching ratios quoted in Fawcett and Wilson [1 3] we set up a till'((level rate equation.

$$\dot{\mathcal{N}}_1 = -\rho(f_{12})g(f_{12})B^f_{12}\mathcal{N}_1 + \rho(f_{12})g(f_{12})B^f_{12}\mathcal{N}_2 + A_{21}\mathcal{N}_2 + \Gamma_{31}\mathcal{N}_3. \quad (1)$$

$$\dot{\mathcal{N}}_2 = \rho(f_{12})g(f_{12})B^f_{12}\mathcal{N}_1 - \rho(f_{12})g(f_{12})B^f_{12}\mathcal{N}_2 - A_{21}\mathcal{N}_2 - A_{23}\mathcal{N}_2 \quad (2)$$

$$\dot{\mathcal{N}}_3 = A_{23}\mathcal{N}_2 - \Gamma_{31}\mathcal{N}_3. \quad (3)$$

$$\mathcal{N}_o = \mathcal{N}_1 + \mathcal{N}_2 + \mathcal{N}_3 \quad (4)$$

where A an B are the Ei n stein coefficients, Γ_{31} is the quenching rate of the "dark" state, and $g(f_{12})$ is the lineshape factor given as the convolution of the Doppler broadening, lifetime broadening and the laser linewidth. Fawcett and Wilson quote the radiative decay rates gA_{ik}, where $g = 2J + 1$ is the statistical weight of the upper level. $A_{21} = 1.189 \times 10^8$ and we set $A_{23} = A(4f^{14}6p^2P_{3/2} \rightarrow 4f^{14}5d^2D_{3/2}) + A(4f^{14}6p^2P_{3/2} \rightarrow 4f^{14}5d^2D_{5/2}) = 3.383 \times 10^5 + 1.458 \times 10^6 = 1.796 \times 10^6$. B_{ik} is related to the A coefficient through the relation

$$B_{ik} = \frac{\lambda^3}{8\pi h}A_{ik} \quad (5)$$

Since the laser linewidth ($\sim$ 1 MHz) is much smaller than either the Doppler broadening ($\sim$ 1 GHz) or the lifetime broadening ($\sim$ 19 MHz) of the S to 1' transition we assumed the la ser to be perfectly mon o chromat ic, reducing $g(f_{12})$ to a convolution of Only the lifetime and Doppler broadening. The lifetime lineshape is given by the lorentzian

$$G_{LT}(f, f_o, \Delta f_{LT}) = \frac{1}{2\pi}\frac{\Delta f_{LT}}{(f_o - f)^2 + (\frac{\Delta f_{LT}}{2})^2} \quad (6)$$

while the Doppler broadening is give by the gaussian

$$G_d(f', f_o, \Delta f_d) = \sqrt{4\frac{\ln 2}{\pi}}\frac{1}{\Delta f_d}e^{-4\ln(2)\left(\frac{f'-f_o}{\Delta f_d}\right)^2}. \quad (7)$$

The resulting Voight profile is given by

$$\mathcal{V}(f, f', \Delta f_{lt}, \Delta f_d) = \frac{1}{2\pi}\sqrt{\frac{4\ln 2}{\pi}}\frac{\Delta f_{lt}}{\Delta f_d} \times \int_{-\infty}^{+\infty}\frac{e^{-4\ln 2(\frac{f'-f_o}{\Delta f_d})^2}}{(f_o - f)^2 + (\frac{\Delta f_{lt}}{2})^2}df_o. \quad (8)$$

Here Δf_d is the Doppler full width at half maximum which we are able to obtain through the resonance fluorescence data fitted with t he sum of five Voight profiles as previously discussed (see fig. 3). Δf_{lt} is the lifetime width and is given by $A_{12}/2\pi$. f' is the center of the Doppler broadened transition where we have shifted the origin to set $f' = 0$ and also set the laser frequency f to overlap the center of this transition. Throughout the experiment the la ser frequency is monitored with the aid of the ytterbium hollow cathode lamp. With $f' = 0$ and $f \to 0$ the lineshape factor $g(f) = g = \mathcal{V}(f, f', \Delta f_{lt}, \Delta f_d)$

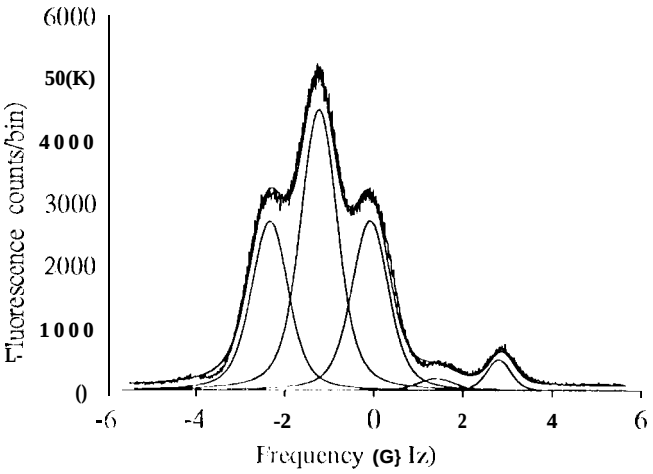

FIG. 3. R esona nce spe c tr um of excitted ytterbium at 1.3×10^{-2} Pa of N_2, fit to a sum of five Voight profiles in order to obtain the doppler width at half maximum and thus the temperature.

in equations 1 an d 2. Steady state solutions of equation 1 - 4 may be obtained by letting $\dot{\mathcal{N}}_1 = \dot{\mathcal{N}}_2 = \dot{\mathcal{N}}_3 = 0$ and by r earranging the equations to get

$$\mathcal{N}_2 = \frac{pgB^f_{12}\Gamma_{31}\mathcal{N}_o}{pgB^f_{12}(2\Gamma_{31} + A_{23}) + \Gamma_{31}(A_{21} + A_{23})} \quad (9)$$

The $\mathcal{N}_2$ level population is the only one of importance since the observed fluorescence level is directly proportional to this population. Several time constants are necessary to fit the fluorescence rise and decay curves. The initial peak fluorescence is dominated by a very fast time constant in which an equilibrium is reached between the ground st ate population depleted by the laser and the decay of the 1' state via the various availa ble decay channels. This peak fluorescence is proportional to the maximum value of $\mathcal{N}_2$ ($\mathcal{N}_2^{max}$) given by setting $A_{23}/\Gamma_{31} = 0$ in eqn. 6. This is justified since the ratio between A_{21} and A_{23} is large (66:1), which in turn implies that the peak population in level 2 is approximately equal to the steady state population in that level if Γ_{31} is very large. This is followed by a much slower t ime constant in which fluorescence is lost due to ions getting shelved into a metastable State. The steady state level beyond several seconds is determined by the act ual value of Γ_{31} The measured ratio R of the steady state fluorescence vs. the peak height is therefore equal to $\mathcal{N}_2/\mathcal{N}_2^{max}$, from which we can determine Γ_{31} as follows:

$$\Gamma_{31} = \frac{pgB^f_{12}A_{23}\mathcal{R}\mathcal{N}_2^{max}}{pgB^f_{12} - (2pgB^f_{12} + A_{21} + A_{23})\mathcal{R}\mathcal{N}_2^{max}}. \quad (10)$$

A plot of Γ_{31} vs. pressure is shown in Pig. 4. The data fit well to a polynomial of order 2 reflecting thermalizing e ffects [1 4,1 5] and multiple collisional effects that may be present We have fit t he first seven data points to a

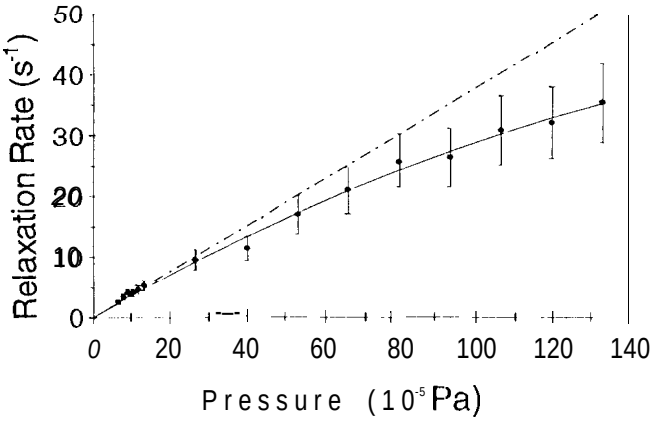

FIG. 4. Quenching rate of ytterbium's trap state vs. N_2 buffer gas]) 1(ss(1,(. The curved line represents a least squares fit of the data to the polynomial Γ_{31} : al' + bl^2; a = 3. 63 ± 0.12 × 10^4 /s/Pa and b = 7.45 ± 1.15 × 10^6/s/Pa^2, while the straight line represents a linear least squares fit to pressures < 26.6 x 10^{-5} Pa.

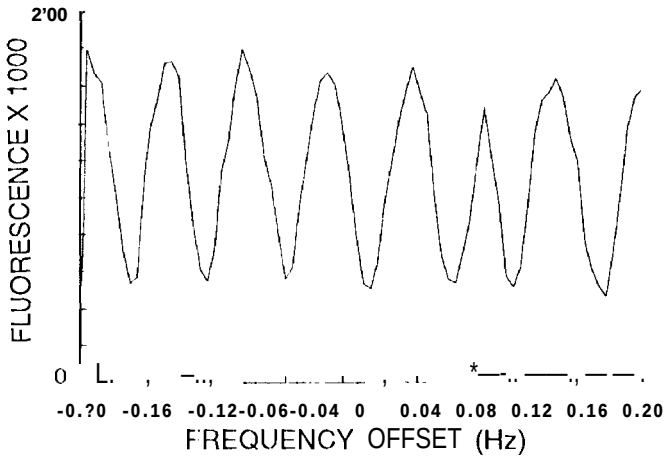

FIG. 5. 25 mHz ramsey fringes corresponding to a 20 second separation between $\pi/2$ pulse.

straight line to obtain a quenching rate Γ_{31} = (3.78 ± ().99X 104/S/I Pa at low pressures ($P \leq 2.66 \times 10^{-4}$ Pa). The uncertainty corresponds primarily to fluctuations in the laser power.

Highly effective quenching of metastable states in many of the alkali-earths with N_2 as a buffer gas has been known for many years and has been attributed to large cross sections for energy transport between the alkali-earths' electronic energy into vibrational and rotational states of the nitrogen molecule [14-16]. We believe the same mechanism is responsible for quenching of the alliali-like singly ionized ytterbium ion by the N_2 buffer gas.

As the data displayed in Fig. 1 shows He, Ar and CO_2 exhibit a considerably smaller efficiency for depleting the population of the dark state as compared to N_2. The absence of quenching efficiency with argon and helium is not surprising since the noble gases do not have the vibrational and rotational structure of diatomic or polyatomic molecules and therefore may only collisionally transfer the electronic energy into kinetic energy, a process with a much smaller cross section. A previous study of ytterbiumion's "(Fil)" state lifetime as a function of buffer gas pressure using He, Ne, and Ar indicated relaxation times on the order of minutes even at pressures approaching 1 Pa [7].

It was however surprising to see that neither CO_2 or H_2 were efficient at quenching the '{dark" state population. In fact, with hydrogen as the buffer gas the fluorescence quickly decayed to the level of the background light and could not be re-established even after extremely long dark periods (~ several hours). This would imply that either the trap time with hydrogen is short or hydrogen is particularly ineffectual in reducing the duration of the dark period. We verified that the trap time with hydrogen is quite long by waiting several hours, after initially loading the ions, before exposing the ions to the uv radiation. Since hydrogen is quite effective in quenching metastable states in other excited ions and atoms,[6,14,17] arid has been shown to be nearly all order of magnitude more efficient than nitrogen in quenching the $^2D_{3/2}$ state in Yb^+ [6], we conjecture that molecular formation between hydrogen and excited ytterbium ions may be occurring. CO_2 proved to be a very ineffective buffer gas yielding very snort trap times, thus being ruled out as a candidate buffer for use in the ytterbium ion frequency standard.

A dual decay time characteristic was exhibited when helium was used as the buffer gas at pressures of 6.7 x 10^{-3} Pa and greater. A nonlinear behavior of the decay rate of the $^2D_{3/2}$ in Yb^+ at high He pressures has been previously observed [6,18] and discussed in detail in ref [18]. We also found that at very high He pressures the trap time was greatly reduced and a competing effect between quenching the "dark" state and loss of ions from the trap appeared to occ '('111.

Since our main objective is to exploit the 12.64 GHz transition between the hyperfine levels of the ground state of $^{171}Yb^+$ we performed microwave optical double resonance spectroscopy using N_2 as a buffer gas and found no loss of coherence in the ground state hyperfine levels due to the N_2 buffer gas. In fact we were able to transfer the population of the ions between the lower and upper hyperfine states over a 25 second period using the Ramsey separated pulse technique which yielded a linewidth of 20 mHz (see fig. 5), corresponding to a line Q Of 6.3 x 10^{11}. Furthermore, nitrogen was so efficient at quenching the "dark stat(" that we were able to obtain single shot photon counts of 1 30,000 above a 50,000 count background, corresponding to a signal-to-noise ra-

tio exceeding 350:1 in the shot noise limit. A potential stability of $1.8 \times 10^{-14}/\sqrt{\tau}$ is thus realizable with the present trapping configuration [19].

In summary we have determined a highly effective means for quenching the "trap" state in the ytterbium ion through introduction of N_2 into the vacuum system. A quenching rate of $(1\{.634\ 0.12 \times 10^4)/s/Pa - (7.454\ 1.15 \times 10^6)/s/Pa^2$ was determined by measuring the ratio of the peak fluorescence to the steady state fluorescence level of naturally abundant ytterbium irradiated with 329nm radiation. We found no loss of ion coherence due to the presence of nitrogen and were able to cool the ion temperature down to 600" kelvins. This approach provides a simple solution to the problem of population trapping, and enables the realization of the full potential of ytterbium as the active ion in a trapped ion frequency standard.

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