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Abstract: The number of stored Ca^+ ions in an ion trap was measured optically by utilizing the metastable states. All the ions trapped are first pumped into the metastable D states. The ions in the metastable D states are transferred to the ground S state via the P state by exciting a $D \rightarrow P$ transition. Each ion then emits one photon through a subsequent $P \rightarrow S$ spontaneous emission. Thus, the number of photons is the same as the number of trapped ions initially in the metastable states. If a known fraction of ions are pumped into the metastable states, the total number of ions is determined by counting the photons with a known detection efficiency.

1 Introduction

In many ion trap experiments, it is of importance to know the number of trapped ions non-destructively. It is, however, not so easy to count the ions reliably and only a few, sometimes ambiguous, methods have been used so far. When a large number of ions are trapped in an ion trap, a rf resonance absorption method is normally used for the measurement of the number of trapped ions [1–3]. A signal of the collective motion of the trapped ions is detected electrically under proper conditions. The sensitivity and the accuracy of the method, however, are not so high because it requires the measurement of a small induced current between endcap electrodes which is often masked by noise from the driving rf field and the signal height and shape depends critically on the proper tuning of the amplifier circuits.

In laser spectroscopy of trapped ions, the intensity of the laser induced fluorescence can be used to estimate the number of stored ions. Actually, a small number of laser cooled ions is really countable by detecting discrete decay or stepwise cooling of the laser-cooled ions [4]. In the case of an ion cloud, one must know the mean excitation rate for the whole ensemble of stored ions to estimate the number. It is, however, not so simple to estimate the excitation rate, because one must normally assume several ambiguous parameters, such as the temperature of the ion cloud, the power and the diameter of the laser beam at the trap region and the fraction of ions irradiated. These parameters depend upon the experimental conditions.

In the present paper, we propose a new method to reliably count the number of ions in an ion trap.

2 Principle

Some ions, such as Ca^+, Ba^+ and Sr^+, have metastable D states from which P states can be accessed by E1 allowed optical transitions. For these ions, we can count the number of trapped ions by employing the one to one correspondence of emitted photons to trapped ions when we utilize metastable states. We use a UV pumping laser to excite the $S \rightarrow P$ transition and two IR lasers to excite the $D \rightarrow P$ transitions (See Fig. 1). The method works as follows. We firstly excite the $S \rightarrow P$ transition while the IR lasers are turned off for the optical pumping of all the trapped ions into the D states. Then, we excite the $D \rightarrow P$ transitions by the IR lasers. At this moment, a prompt increase of the fluorescence intensity occurs because each ion, quickly excited from the D states, emits one photon due to the spontaneous emission from the P to the S state. Since one UV photon corresponds to one ion, the total number of trapped ions, N, can be derived as

$$N = \frac{N_{\mathrm{det}}}{\epsilon}, \tag{1}$$

where N_{det} is the number of detected photons in the prompt signal of the fluorescence and ϵ is the detection efficiency.

If there is a relaxation of the D states to the ground state due to gas collisions, the method should be modified. In this case, complete optical pumping of the stored ions to the D states is not attained. We need, therefore, to know the population extent ρ_D of ions pumped in the D states to estimate the number of ions, then

$$N = \frac{N_{\mathrm{det}}}{\rho_D \epsilon}. \tag{2}$$

To explain the modified method, we take the three level system, in which the population extent ρ_D is given as

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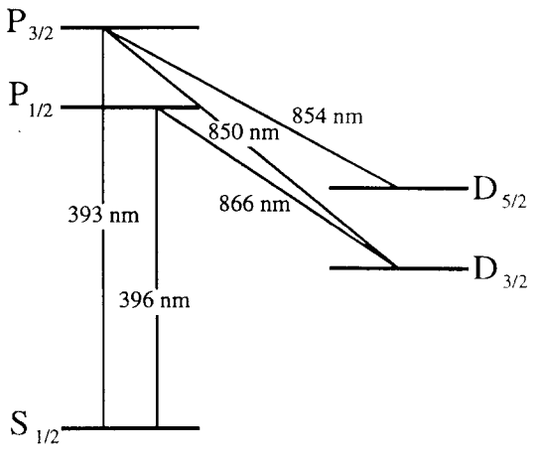

Fig. 1. Energy diagram of $^{40}Ca^{+}$ ion.

$$\rho_D = \frac{A_{PD} l_{SP}}{A_{PD}(l_{SP} + \gamma_{DS}) + \gamma_{DS}(A_{PS} + l_{SP})}, \tag{3}$$

where A_{if}, l_{if} and γ_{if} are the spontaneous emission rate, the excitation rate and the relaxation rate, respectively and the suffixes i and f denote the initial- and final-state, respectively. ρ_D can be estimated experimentally as follows. We measure fluorescence intensities at two different equilibrium conditions; I_a when all the lasers are turned on and I_b when the IR laser is turned off. The approximate value of ρ_D, which becomes accurate when pumping rate is high, is given as a ratio, η, of the measured intensities as shown in the following formula.

$$\begin{aligned}\eta &= (I_a - I_b)/I_a \\ &= \frac{A_{PD} l_{DP}(l_{SP} + \gamma_{DS})}{(l_{DP} + \gamma_{DS})\{A_{PD}(l_{SP} + \gamma_{DS}) + \gamma_{DS}(A_{PS} + l_{SP})\}} \\ &= (1 + \delta)\rho_D. \end{aligned} \tag{4}$$

If the excitation rate l_{DP} is much higher than the relaxation rate γ_{DS}, the correction factor δ is derived from the above equations as

$$\delta \cong \frac{\gamma_{DS}}{l_{SP}}. \tag{5}$$

These parameters can be measured in the following way. Since the detected photon count N_{det} is proportional to the number of ions in the D state, γ_{DS} corresponds to the decay constant of the variation of N_{det} as a function of the time interval between the pumping laser off and the IR laser on. Once γ_{DS} is measured, the excitation rate l_{SP} is also obtained. Since the stationary fluorescence intensity I_a in the usual condition is given as

$$I_a = c l_{SP} N, \tag{6}$$

l_{SP} is derived using (2-5) as

$$l_{SP} = I_a \eta / N_{det} - \gamma_{DS}. \tag{7}$$

Consequently, we can obtain the number of ions even in cases where not all the ions are pumped into the metastable states.

In the four level system where ions are pumped into the D states via the $S_{1/2} \rightarrow P_{3/2}$ transition, we can also use the above results in the following way: 1) When we pump ions into one of the D states, the population extent of the pumped D state becomes smaller due to the collisional fine structure mixing. However, (5-7) can be used to estimate the correction factor. 2) When ions are pumped into both D states, we can treat the D states as one state. The mean relaxation rate of both states substitutes for the γ_{DS} in the estimate of the correction factor. As long as the assumption of $l_{DP} \gg \gamma_{DS}$ is valid, we can deduce the population extent ρ_D by using (5-7). In the measurement of the ratio η, we should be careful of the coherent effect in the four level or three level system, the so called dark resonance which can reduce the fluorescence intensity [5, 6]. It can be avoided by making a slight misalignment and a proper detuning of the lasers.

One can deduce the mean excitation rate, l_{SP}, from the measurement of the decay rate of the fluorescence after turning the IR laser off. The decay curve of the fluorescence intensity is described as

$$I \propto C_1 \exp(-\lambda_+ t) + C_2 \exp(-\lambda_- t) + \frac{c}{b}, \tag{8}$$

and the two decay rates λ_+(fast) and λ_-(slow) are given as

$$\lambda_\pm = \frac{a}{2} \pm \frac{1}{2}\sqrt{a^2 - 4b}, \tag{9}$$

where $C_{1,2}$ are constants determined by the initial conditions, $a = \gamma_{DS} + A_{PS} + A_{PD} + l_{SP}$, $b = A_{PD} l_{SP} + \gamma_{DS}(A_{PS} + A_{PD} + l_{SP})$ and $c = \gamma_{DS} l_{SP}$. The fast component λ_+ is the order of the spontaneous decay rate of the P state, since the population of the P state decreases after turning off the $D \rightarrow P$ excitation. The slow component λ_- corresponds to the pumping rate from the S state to the D states. Inserting the measured decay constant λ_-, γ_{DS} and known transition probabilities [7] to (9), l_{SP} is obtained. From l_{SP}, we can also estimate the total number of ions with the measured stationary fluorescence intensity I_a by using (6). This alternative way to determine l_{SP} is, however, not so reliable since it requires the knowledge of many parameters and is obtained as the small difference of large quantities, which are of the same order of magnitude.

3 Experimental apparatus

The experimental setup is as follows. A linear rf trap, which consists of three sections, each made from four separated cylindrical rods each with a diameter of 6 mm, was used to store Ca^{+} ions. The closest distance $2r_o$ between diagonal electrodes was 5.22 mm. The rf frequency was 8 MHz and the amplitude was 204 V. We also applied a DC voltage of 7-15 V to the two end sections. The Ca^{+} ions were produced by focusing a Nd:YAG pulsed laser beam on a metallic calcium target placed at one side of the trap. The peak power of the beam on the target was estimated to be more than 10^9 W/cm^2, which enabled us to easily prepare a laser ablated plasma consisting of Ca^{+} ions in the trap.

To excite the $S_{1/2} \rightarrow P_{3/2}$ transition of Ca^{+} ions, we used 393 nm UV laser light which was generated by intracavity second harmonic generation using an angle tuned $LiIO_3$ crystal in a Ti-Sapphire ring laser (Coherent 899-21) pumped by an Ar^{+} ion laser (Coherent Innova 300, 10 W). The linewidth of

the UV laser is less than 1 MHz and the maximum power is about 2 mW. To excite the $D \rightarrow P$ transitions, we used two laser diodes (LD, Spectra Diode Lab., SDL-5400). The power of LD's with a linewidth of about 30 MHz is typically 10 mW. By using two mechanical shutters, we can generate a laser beam pulse with a variable width and provide the necessary pulse sequence for the measurements. All the laser beams were merged by a half mirror and a dichroic mirror and were sent into the trap chamber through a glass window. The chamber was evacuated to less than 10^{-7} Pa by a turbo molecular pump backed by a rotary pump. For buffer gas cooling of trapped ions, we introduced helium gas to the vacuum chamber from the fore vacuum side of the turbo molecular pump in order to reduce impurities. The fluorescence from the ions, focused by two lenses which were mounted inside the chamber and a camera lens (Nikon 28 mm f/2.8), was detected by a position sensitive photon counting system (Hamamatsu PIAS-TI). An interference filter with 33% transmittance at 393 nm was inserted in front of the camera lens to detect only 393 nm UV photons. We also inserted a neutral density (ND) filter, in some cases, for decreasing the detection efficiency in order to avoid a pile-up effect in the detector.

4 Experimental results

The method of counting trapped ions has been tested in the case of Ca^+ ions. The Ca^+ ion has the excited $P_J (J = 1/2, 3/2)$ states and the metastable $D_J (J = 3/2, 5/2)$ states near the ground $S_{1/2}$ state as illustrated in Fig. 1. The D states have long lifetimes of about 1 second [8–10]. Figure 2 shows the time spectrum of the fluorescence. In the period A, the UV pumping laser (393 nm) and two IR (850, 854 nm) lasers were turned on and tuned to the resonance and the stationary intensity of UV fluorescence I_a was observed. In the period B, one of the IR lasers was detuned far from the resonance. Then, a fraction of ions was pumped into the metastable $D_{5/2}$ state. As a result, the fluorescence intensity decreased to I_b. At the end of period B, the IR laser was tuned on the resonance again, then, a prompt increase of the fluorescence intensity was observed as a spike peak due to the $D_{5/2} \rightarrow P_{3/2}$ excitation of ions in the $D_{5/2}$ state. The number of emitted UV photons at the moment of the prompt increase is the same as the number of ions in the $D_{5/2}$ state.

The number of detected photons accumulated for 100 cycles of measurements was 322±42 and the intensity ratio η was 66±2%. The error is mainly due to the background count. The relaxation rate of the $D_{5/2}$ was assumed to be of the order of 1 Hz under our experimental conditions [8]. The excitation rate Γ_{SP} of $(8.8 \pm 1.1) \times 10^2$ Hz was deduced from the detected counts in the spike peak and the relaxation rate γ_{DS} by using (7). The correction factor δ is estimated as less than 0.005, which allowed to take $\rho_D = \eta$. The result gives the total number of ions N to $(4.9 \pm 0.6)/\epsilon$, where ϵ is the detection efficiency. The detection efficiency, which was estimated from the geometry of the detection system, transmittance of the optical elements and the quantum efficiency of the detector, was 3×10^{-4} for this setup. Then the absolute number was obtained to be 1.6×10^4. The intensities of the laser radiations were about 3 μW for the UV and 10 mW for the IR laser.

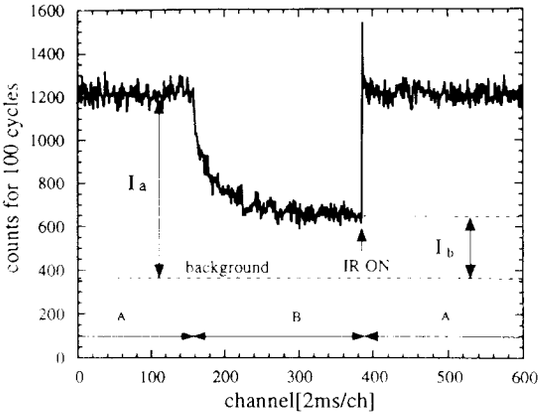

Fig. 2. Fluorescence intensity as a function of time showing the present method in the case of a weak UV laser (3 μW). The fluorescence spike at the moment when the IR laser is tuned on indicates the number of ions in the trap (For details, see text.).

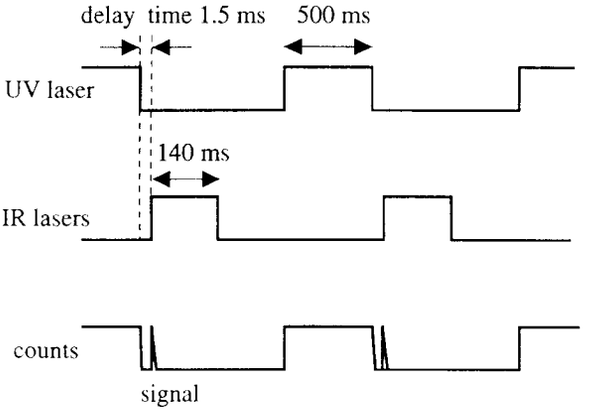

Fig. 3. Time sequence of the experiments shown in Fig. 4. The periods given in the figure are typical values.

We also performed an improved measurement in which a very high signal-to-noise ratio was attained. The time sequence of the method is shown in Fig. 3. The key point of the improvement is to stop the UV laser when we observe the prompt increase of the fluorescence. Origins of the noise in the previous experiment were stray photons from the UV laser,

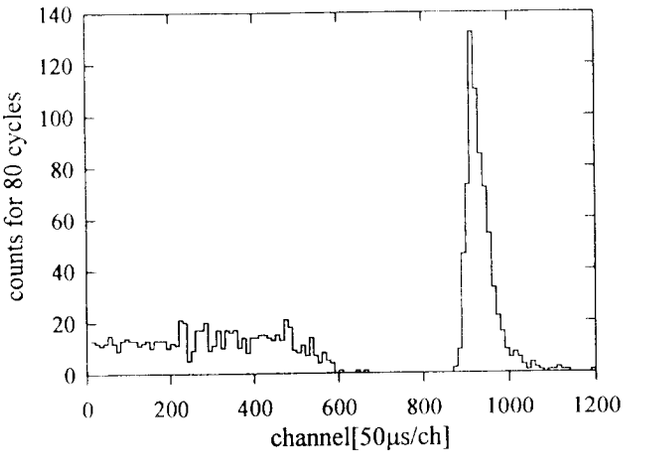

Fig. 4. Typical result for the improved measurement accumulated for 80 cycles

fluorescence from ions in the ground state and dark count of the detector. The first two, which are in fact the dominant ones, can be suppressed completely by blocking the UV laser at the moment of turning the IR lasers on. The experimental result is shown in Fig. 4. Since the background count rate due to the noise became zero, we could increase the UV laser power to about 180 μW in order to pump all the trapped ions into both D states ($\rho_D = 1$). We had to take care of a pile-up effect at the moment of the spike peak, since the detection efficiency and the capacity of ions for the new setup was higher than previously and the dead-time of the detector was about 10 μs. The power of the IR lasers was decreased to about 0.9 mW and, when necessary, an ND filter was inserted to the optical detection system. The channel-advance time of the data acquisition system (a multi-channel scaler) was also shortened to resolve the structure of the spike peak. Consequently, the observed time spectrum of the spike peak was not prompt but exponentially delayed. The round rising edge indicated a slow response of the mechanical shutter. We also take into account the relaxation of pumped ions in the metastable D states to the ground state due to gas collisions [8, 9, 11] during delay-time of 1.5 ms time interval between turning the UV laser off and IR lasers on, which was necessary to avoid overlapping of the noise to the spike peaks. The relaxation rate of about 2 Hz, according to our decay rate measurement under the same conditions, means the loss of ions in the D states during the period of 1.5 ms was less than 0.3%. The sum of the detected UV photons was 700±26 counts after 80 cycles of measurements. The total number of ions was $N = (8.75 \pm 0.35)/\epsilon$. The estimated detection efficiency with the ND filter in the new setup was 3×10^{-4} which gives the absolute number of ions as $N = 3 \times 10^4$. The uncertainty of the detection efficiency may be of the order of 30 %. In comparing ion numbers under different conditions, however, this factor cancels and ratios of ion numbers are much more accurate.

In some particular cases, we should be careful with the fine structure mixing effect due to buffer gas collisions. It may become serious when we pump into only one metastable state ($D_{3/2}$) using UV excitation of the $S_{1/2} \rightarrow P_{1/2}$ transition instead of the $S_{1/2} \rightarrow P_{3/2}$. In order to show this effect, we performed an experiment as shown in Fig. 5. In this measurement, the experimental condition was the same as previously but only one IR laser (854 nm, $D_{5/2} \rightarrow P_{3/2}$) was used. The time spectrum shows a slow decay component as well as a prompt one. This slow part was due to a feed from another metastable state, $D_{3/2}$, to the $D_{5/2}$ state from which ions were excited by the IR radiation. The time constant of 6.6±0.3 ms corresponds to the mixing rate under the present condition. The same effect was observed when we used the other IR laser (850 nm) only to excite the $D_{3/2} \rightarrow P_{3/2}$. If one uses the $S_{1/2} \rightarrow P_{1/2}$ transition to pump into the $D_{3/2}$ state and detect the fluorescence due to the excitation of the $D_{3/2} \rightarrow P_{1/2}$, one might miss a fractional amount of the ions, which are populated in the $D_{5/2}$ state by the mixing effect, in the estimate of the total number of ions. One should use two IR lasers by which both $D_{5/2}$ and $D_{3/2}$ states can be excited even though one uses the $S_{1/2} \rightarrow P_{1/2}$ excitation for the pumping.

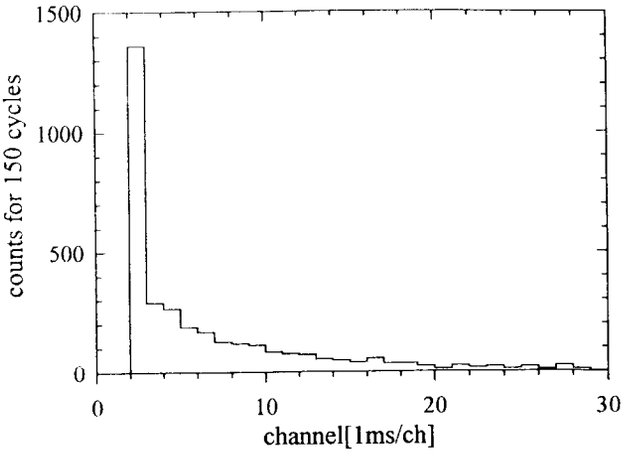

Fig. 5. Mixing effect between the fine structure doublet. The slow decay time constant of 6.6 ± 0.3 ms corresponds to the mixing rate from the $D_{3/2}$ to the $D_{5/2}$ state

5 Summary

We have developed and demonstrated a new, simple and reliable method to count the total number of trapped ions having an appropriate energy level scheme. Since the ambiguity comes only from the estimate of the detection efficiency, this method is very reliable particularly for the estimate of the relative number of ions in the same setup as long as the efficiency is constant. This assumption is true except for the case that the view of the detector does not cover the whole area where ions are distributed. It is also possible to measure the detection efficiency accurately by counting fluorescence from a saturated single ion which can be cooled by a single laser radiation, such as Be^+ or Mg^+.

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