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Laser cooling and quantum jumps of a single indium ion

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We performed laser spectroscopy of In^+ ions stored in a radio-frequency trap. The $5s^2\,{}^1S_0 \rightarrow 5s5p\,{}^3P_1$ transition at 230.6 nm was excited and its hyperfine splitting and isotope shift have been measured. The lifetime of the ${}^3P_1$ state was measured using intensity modulated laser excitation: $\tau({}^3P_1)=0.44(4)\ \mu\mathrm{s}$. A single indium ion was laser cooled using this intercombination line and dark periods in the fluorescence were observed when the metastable ${}^3P_0$ state was populated by a magnetic dipole decay from ${}^3P_1$. The radiative lifetime of the forbidden transition ${}^3P_0 \rightarrow {}^1S_0$ was determined from the duration of the dark periods: $\tau({}^3P_0)=0.14(2)$ s. This value is compared to calculations based on perturbation theory. Prospects for an optical frequency standard using the narrow $5s^2\,{}^1S_0 \rightarrow 5s5p\,{}^3P_0$ resonance at 236.5 nm of a single laser-cooled indium ion are discussed.

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I. INTRODUCTION

The use of the forbidden optical transition of a single laser-cooled ion stored in a Paul trap has been proposed as the basis for a new atomic frequency standard of high accuracy [1]. Impressive progress has already been achieved along these lines and very-high-resolution laser spectroscopy has been reported on Hg^+ [2] and Ba^+ ions [3]. Other ions that are investigated in this context are ytterbium, strontium, and calcium. A severe restriction in the choice of ions for these experiments is the availability of suitable transitions: a strong dipole transition is needed for laser cooling and a long-lived transition for the frequency standard, and both lines have to be in the wavelength range accessible for continuous wave laser sources. The prospective clock transitions of all the ions mentioned above are electric quadrupole transitions like ${}^2S_{1/2} \rightarrow {}^2D_{5/2}$ or higher-order multipole processes. The field dependence of those transitions may cause a problem for a frequency standard. Around zero magnetic field they show a linear Zeeman shift of the order MHz/G unless a Zeeman component $(m_F=0) \rightarrow (m_F=0)$ of an isotope with half-integer nuclear spin is used [2]. In this case however the hyperfine splitting of the ground state makes the realization of a cycling transition for laser cooling more complicated. A small but difficult controllable shift arises from the interaction with the electric quadrupole field of the trap [1].

To avoid these problems Dehmelt suggested the use of a transition between two states with vanishing angular momentum of the electrons like $6s^2\,{}^1S_0 \rightarrow 6s6p\,{}^3P_0$ in Tl^+ [1,4] or the corresponding lines of the other ions of the third main group of the periodic system: boron, aluminum, gallium, and indium. The three lighter of these cannot be laser cooled presently because the intercombination line ${}^1S_0 \rightarrow {}^3P_1$ is too weak and the wavelength of the strong dipole transition ${}^1S_0 \rightarrow {}^1P_1$ lies in the vuv below 160 nm and is thus inaccessible for continuous-wave (cw) lasers. In the case of thallium the wavelengths for ${}^1S_0 \rightarrow {}^3P_0$ and ${}^1S_0 \rightarrow {}^3P_1$ are 202.2 and 190.8 nm, respectively, and for indium they are 236.5 and 230.6 nm. Figure 1 shows a simplified energy-level diagram of In^+. The two indium wavelengths can be generated relatively easily using the β-barium borate (BBO) crystal for the frequency doubling of blue lasers. The clock transition at 236.5 nm is especially technically convenient, since it coincides with the fourth harmonic of the 946-nm transition ${}^4F_{3/2} \rightarrow {}^4I_{9/2}$ of the Nd:YAG laser (where YAG denotes yttrium aluminum garnet). So this intrinsically frequency stable solid-state laser can be used as the oscillator to drive the clock transition. Indium is thus presently the most promising of the group-III ions.

The decay ${}^3P_0 \rightarrow {}^1S_0$ between two levels with vanishing electronic angular momenta is forbidden in all orders of the multipole expansion of the radiation field. However for atoms with nonvanishing nuclear spin the hyperfine interaction induces a weak electric dipole transition between these states. This is due to small admixtures of the decaying states ${}^1P_1$ and ${}^3P_1$ to the otherwise stable ${}^3P_0$ level. This effect has been quantitatively investigated so far only in a few cases. In mercury the absorption coefficient for the forbidden transition was measured and the lifetime of ${}^3P_0$ was inferred from that [5]. The result was 1.7 s for ${}^{199}\mathrm{Hg}$ and 2.6 s for ${}^{201}\mathrm{Hg}$. Recently lifetimes

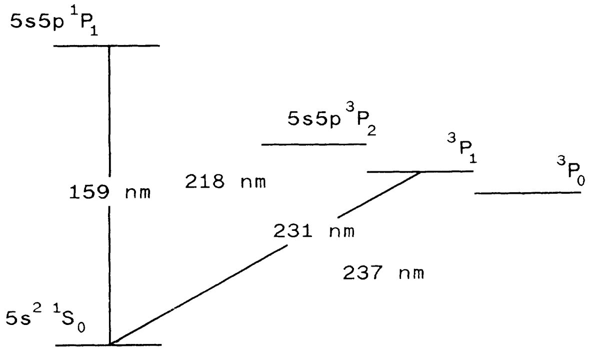

FIG. 1. Simplified energy-level scheme of the In^+ ion.

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in the picosecond range have been measured for hyperfine quenched $^3P_0$ states of highly charged heliumlike ions [6]. In the experiment reported here we use the perturbation-free environment offered by an ion trap to measure the lifetime of the $^3P_0$ state in In^+. We observe the quantum jumps of a single ion into and out of this level using Dehmelt's electron shelving scheme [1,4].

The paper is organized as follows. The next section describes the experimental setup for the storage and laser excitation of indium ions. Sections III and IV give the results of spectroscopy on the cooling transition $^1S_0 \rightarrow {}^3P_1$ and the measurement of the lifetime of the $^3P_1$ level. Section V describes laser cooling of a single indium ion and the observation of quantum jumps that allowed the determination of the lifetime of the $^3P_0$ state. This value is compared to theory in Sec. VI.

II. EXPERIMENT

We used two different types of radio-frequency traps that have already been described earlier: the experiments on large clouds of indium ions were performed with a hyperbolic Paul trap with a 5-mm-diam ring electrode [7], driven by an ac voltage of about 400-V amplitude at 4.3 MHz. For the experiments with single laser-cooled ions we used a Paul-Straubel trap with a 1-mm-diam ring electrode [8], driven by a voltage of 1000 V at 10.7 MHz. Together with a small indium oven, an electron source for ionization, and the detection optics for monitoring the fluorescence of the ions, the trap is mounted inside a stainless-steel ultrahigh-vacuum chamber. A turbomolecular pump and an ion getter pump are used to maintain a background pressure of a few times 10^{-8} Pa. The indium oven consists of a small tantalum tube (1 mm in diameter) that is closed on one end, filled with indium, and heated to about 550 °C by passing a current through a wire spot welded to the tube. The evaporated indium atoms are ionized by electron impact inside the trap.

The vacuum chamber is surrounded by three pairs of coils oriented along orthogonal axes, one of which coincides with the laser beam direction. These coils are used to compensate for the earth's magnetic field or to apply a controllable magnetic field. When the ions are optically pumped and the fluorescence intensity is observed as a function of the magnetic field, we get a narrow zero-field resonance due to magnetic depolarization of the ground state, analogous to the Hanle effect of an excited state [9]. This signal can be used to cancel magnetic stray fields inside the trap with a precision of a few mG.

The laser radiation at 231 nm for the excitation of the indium ions is generated by frequency doubling blue light from a stilbene-3 ring dye laser (Coherent 699-21), pumped by an UV argon ion laser. Frequency doubling is done with a BBO crystal which is placed inside an external cavity that is locked to the laser frequency [10] and enhances the intensity of the blue radiation. The BBO crystal is 7 mm long and has a Brewster cut for type-I angle phase matching. The system is capable of producing a maximum power of 3-mW single frequency light in the ultraviolet. Typically only 0.1 mW was used to excite the ions. The laser power is actively stabilized using a Pockels cell and a polarizer as a variable attenuator. A three-lens system ($f/1.4$) directly attached to the trap collects the fluorescence light from the trapped ions. It images the fluorescence onto a pinhole separating it from laser light scattered from the trap electrodes. The light passing the pinhole is detected with a solar blind photomultiplier (Hamamatsu R166P). The photomultiplier pulses are counted in a computer that is also used to control the laser frequency.

III. SPECTROSCOPY OF THE $^1S_0 \rightarrow {}^3P_1$ TRANSITION

To measure the wavelength and hyperfine splitting of the $5s^2\,{}^1S_0 \rightarrow 5s5p\,{}^3P_1$ transition at 230.6 nm we observed a cloud of about 10^5 ions. Indium in the natural isotopic abundance was used, consisting of 95.7% $^{115}\mathrm{In}$ and 4.3% $^{113}\mathrm{In}$. Both isotopes have a nuclear spin $I=\frac{9}{2}$. The ions were cooled by collisions with helium buffer gas of 10^{-3} Pa pressure. This results in a temperature of the ions of 1000 K and a corresponding Doppler width of 3 GHz for the optical transitions. By scanning the laser frequency we obtained the excitation spectrum shown in Fig. 2 with the three hyperfine components $F=\frac{9}{2} \rightarrow \frac{7}{2}, \frac{9}{2}, \frac{11}{2}$. A magnetic field of 5 G was applied in the direction of the laser beam to define the quantization axis. Without this field fluorescence on the $F=\frac{9}{2} \rightarrow \frac{7}{2}$ hyperfine component is not detectable, because the linearly polarized laser light pumps the ions into the Zeeman sublevels $m_F = \pm\frac{9}{2}$ of the ground state from where they are no longer excited. The magnetic field makes the ions "see" σ instead of π polarized light and ensures excitation from every ground-state sublevel. The observed relative intensities of the three hyperfine components depend on the population of the Zeeman sublevels and the respective transition probabilities.

The laser intensity was kept at a low value of about 1 W/cm^2 to avoid line shifts due to laser cooling or heating. Laser cooling of ion clouds leads to a redshift in the excitation spectrum [11] whereas in the case of buffergas

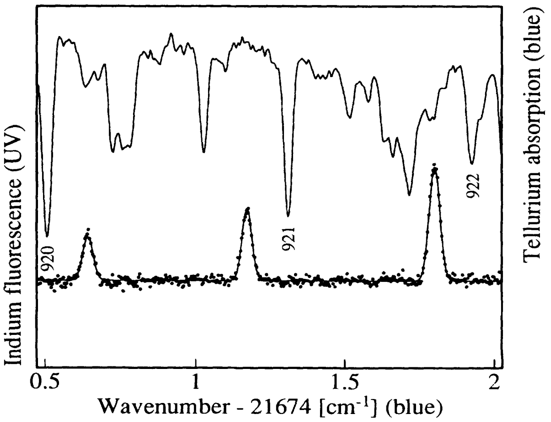

FIG. 2. Excitation spectrum of stored indium ions showing the hyperfine structure of the $5s^2\,{}^1S_0 \rightarrow 5s5p\,{}^3P_1$ transition at 230.6 nm and tellurium absorption spectrum at 461.2 nm, recorded as a wavelength reference. The numbers below the tellurium lines refer to Ref. [12].

cooled clouds higher laser intensities cause a blueshift of the line. The laser heating occurring for positive detuning broadens the high-frequency side of the line and causes an asymmetric line profile, so that the center appears shifted to higher frequencies. In Fig. 2 this is not detectable and the lines are fitted by simple Gaussians. The isotope shift between ^{115}In and ^{113}In is too small to be resolved here, and in addition the latter isotope is of small abundance.

As a wavelength reference we recorded the absorption spectrum of the tellurium molecule ^{130}Te$_2$. A large number of these lines in the blue spectral region are tabulated to an accuracy of 10^{-4} cm^{-1} [12]. We used enriched ^{130}Te in a quartz cell that is heated to about 500 °C to obtain the required vapor pressure. The tellurium absorption is measured using the blue fundamental laser radiation, while the indium excitation spectrum is recorded simultaneously with the doubled laser frequency in the UV. The three reference lines that are closest to half the frequency of the indium resonance and were used in the analysis are 920: 21 674.509 7 cm^{-1}; 921: 21 675.316 4 cm^{-1}; 922: 21 675.931 4 cm^{-1} (the numbers of the lines refer to Ref. [12]). They are marked in the tellurium spectrum in Fig. 2 above. The wave numbers and wavelengths for the three hyperfine components of $^1S_0 \rightarrow {}^3P_1$ are given in Table I. The uncertainty for these values is due to the nonlinearity in the laser scan and the linewidths of indium and tellurium. The hyperfine constants of the $^3P_1$ level as derived from this measurement are

$$A(^3P_1)=0.232(3)\ \text{cm}^{-1}\ ,$$

$$B(^3P_1)=-0.016(14)\ \text{cm}^{-1}\ .$$

An extensive survey of the spectrum of In$^+$ in the wavelength range 210–925 nm was published in 1938 by Paschen and Campbell [13] and is still the source for indium wavelengths in the tables. Our absolute wave numbers are about 0.3 cm^{-1} smaller compared to Ref. [13], whereas there is agreement to within one standard deviation in the hyperfine splittings. Recently Larkins and Hannaford reported measurements that confirm our values and increase the accuracy by another order of magnitude [14].

The observation of laser-cooled indium ions with largely reduced Doppler broadening enabled us to measure the isotope shift between ^{113}In and ^{115}In on this transition. The respective resonance frequencies of the $F=\frac{9}{2}\rightarrow\frac{11}{2}$ hyperfine component of single ions of both isotopes were measured relative to a Doppler-free line in the tellurium spectrum, obtained with saturation spectroscopy. Since the nuclear moments of the isotopes are slightly different, hyperfine interaction contributes a shift of 60 MHz between the two isotopes for this component ($A_{115}/A_{113}=1.002\,13$ and $B_{115}/B_{113}=1.0138$ are the ratios of magnetic dipole [15] and electric quadrupole moments [16], respectively). The pure isotope shift of the $^1S_0 \rightarrow {}^3P_1$ line is

$$\nu_{115}-\nu_{113}=-0.64(7)\ \text{GHz}\ .$$

The negative sign indicates that the volume effect is dominating the isotope shift in this case.

TABLE I. Wave numbers and wavelengths for the three hyperfine components $(5s^2\,{}^1S_0,\frac{9}{2})\rightarrow(5s5p\,{}^3P_1,F)$ of In$^+$.

F	Wave number (cm^{-1})	Wavelength (nm)
$\frac{7}{2}$	43 349.30(2)	230.6133(1)
$\frac{9}{2}$	43 350.36(2)	230.6076(1)
$\frac{11}{2}$	43 351.62(2)	230.6009(1)

IV. MEASUREMENT OF THE LIFETIME OF THE $^3P_1$ LEVEL

The lifetime $\tau(^3P_1)$ of the $^3P_1$ level, the upper level of the cooling transition, is an important parameter for the project of an indium frequency standard. It determines the maximum rate of fluorescence photons emitted from a single ion, the power of laser cooling, and the minimum achievable temperature.

To measure $\tau(^3P_1)$ we excited a cloud of trapped indium ions with square wave intensity modulated laser radiation and observed the decay of the $^3P_1$ population using a delayed coincidence technique. An acousto-optic modulator (AOM) was placed between the dye laser and the enhancement cavity used for the frequency doubling. The radio-frequency drive of the modulator was turned on and off every 2.5 μs, so that the undiffracted beam, which was used for the frequency doubling, had either the full intensity or was attenuated by the amount of light diffracted into the sidebands. Since the modulation frequency was well above the unity gain frequency of the servo loop for the enhancement cavity, the cavity length remained locked to the laser frequency. The resulting UV beam was modulated with an intensity ratio 1000:1 and a rise time of 100 ns, limited by the transit time of the acoustic wave in the modulator. The pulse generator that controlled the driver of the AOM also gave the start pulse for a time-to-amplitude converter (TAC). The TAC was stopped by the first photon detected from the trap. The pulse-height distribution from the TAC was recorded in a multichannel analyzer and transferred to a computer for further analysis. This signal directly represents the temporal behavior of the fluorescence because the photon count rate (≈ 10 kHz) was much smaller than the modulation frequency of 200 kHz. To take account of the finite rise time and the small amount of laser radiation still present when the modulator drive was on, the time dependence of the stray light from the empty trap was measured using the same procedure. If $s(jT)$ represents the data taken with stray light (T, time interval of the multichannel analyzer; j, channel number) and $f(jT)$ those taken with fluorescence, a fit to the function

$$f(jT)=e^{-jT/\tau}\left[k_1+k_0\sum_{k=1}^{j}s(kT)e^{kT/\tau}\right]+k_2 s(jT) \qquad (1)$$

was carried out. Equation (1) represents the general solution for the fluorescence intensity under time-dependent excitation plus a background term describing laser stray light. k_0, k_1, and k_2 are fitting parameters. A set of data with the fitted function is shown in a logarithmic plot in Fig. 3. The decay constant τ gives the lifetime of the $^3P_1$ state,

$$\tau(^3P_1)=0.44(4)\ \mu\text{s}\ ,$$

which is relatively long because In^+ does not deviate strongly from ideal *LS* coupling, where this intercombination line is forbidden.

Our value for the lifetime is close to the results of a number of *ab initio* calculations which lie in the range 0.33–0.59 μs [17]. A previous measurement of $\tau(^3P_1)$ in In^+ with the beam-foil method gave the result 0.125(25) μs [18], which seems to be too small for this intercombination line. Most of the lifetimes of other levels reported in Ref. [18] are below 10 ns. A reason for the large deviation may be that the method used is not suitable for the measurement of long lifetimes.

The lifetime of the $^3P_1$ level of 0.44 μs implies a natural linewidth $\gamma=1/\tau=2\pi\times360$ kHz for the cooling transition. This is about two orders of magnitude narrower than the lines used previously in laser cooling of Ba^+, Mg^+, or Hg^+. Since the secular frequencies ω of the indium ion in the trap were typically $2\pi\times1$ MHz in this experiment, we are already in the strong binding regime $\gamma<\omega$ and are cooling on resolved sidebands. This opens the possibility of reaching the quantum ground state of the ion's oscillation in the trap. The minimum kinetic energy achievable with laser cooling in this case is given by [19,20]

$$E_{\min}=\hbar\omega\left[\frac{1}{2}+\alpha\frac{\gamma^2}{\omega^2}\right]\ , \tag{2}$$

where α is a geometry-dependent factor of the order of magnitude 1. The term $\alpha\gamma^2/\omega^2=\langle n\rangle$ is the mean occupation number for the oscillator levels and is already as small as 0.1 in our case. The lower temperature limit would thus be $T_{\min}=\hbar\omega/k_B\ln(1+\langle n\rangle^{-1})=20$ μK. Cooling on resolved sidebands in the strong binding regime has already been reported using the clock transition in the Hg^+ ion [21], but there the ion was precooled on a strong transition. This is not necessary in the case of indium, but the weakness of the cooling power for the initially high temperatures and the small fluorescence signal associated with the narrow cooling transition pose experimental difficulties.

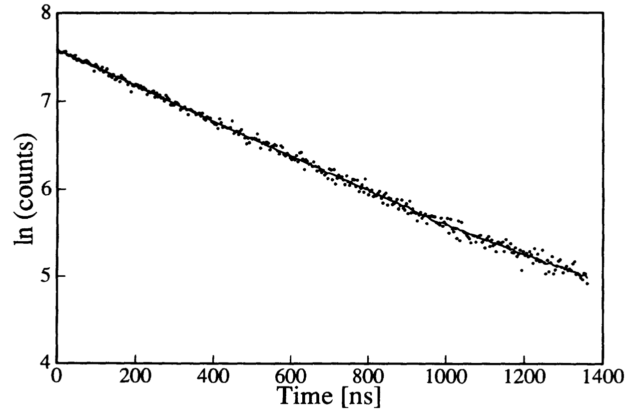

FIG. 3. Decay curve of the fluorescence from the $^3P_1$ level.

V. OBSERVATION OF QUANTUM JUMPS OF A SINGLE INDIUM ION AND LIFETIME MEASUREMENT OF THE $^3P_0$ LEVEL

For the experiments with single ions we use a Paul-Straubel trap [8,22] with a 1-mm-diam ring electrode. This trap combines a steep potential for good localization of the ion that can only be achieved in a small trap with a good detection efficiency for the fluorescent light and a small contribution of laser stray light due to the open electrode structure [8]. Without damping through a buffer gas the trap still can store several ions, but at a typical well depth of 4 eV these can be evaporated from the trap by heating them with the laser detuned to the blue side of the resonance. After this procedure a single ion usually remains which is then trapped in a very stable manner. It can be kept for hours even when it is not permanently laser cooled. We also observed Coulomb crystals [11,23] of two laser-cooled indium ions separated by 3 μm.

In order to laser cool the ion on the narrow $^1S_0\rightarrow{}^3P_1$ transition its hyperfine component $F=\frac{9}{2}\rightarrow\frac{11}{2}$ was excited with circular polarized light in zero magnetic field. This effectively prepares a two-level system $(F=m_F=\frac{9}{2})\rightarrow(F=m_F=\frac{11}{2})$ by means of optical pumping. We observed photon count rates up to 1.5 kHz from a single ion in saturation, indicating that our overall detection efficiency is about 0.1%. Usually we worked below saturation to get a better ratio of fluorescence signal to laser stray light, where we achieved a value of 12. By scanning the laser we recorded excitation spectra of the cooling transition with a linewidth of about 20 MHz. This is limited partly by laser frequency jitter and also to a larger extend by frequency modulation caused by the micromotion of the ion at the trap frequency of 10.7 MHz. A linewidth of 20 MHz corresponds to a modulation index of 2 and a residual micromotion amplitude of 70 nm that might be caused by a slight displacement (≈1 μm) of the ion from the trap center. Very precise control of stray dc fields is required to minimize this effect. The presence of micromotion sidebands leads to a strong variation in the temperature as the laser is scanned across the line profile so that we can presently only give an upper limit of 100 mK for the temperature. If the laser frequency is stabilized to a linewidth below the natural linewidth of the cooling transition corresponding to 360 kHz and the micromotion is further reduced much lower temperatures will be possible.

If we look at the temporal behavior of the resonance fluorescence of a single indium ion on the transition $^1S_0\rightarrow{}^3P_1$ we observe dark periods whenever the metastable $^3P_0$ level is populated. This can happen either by direct laser excitation from the ground state, as it will be done for the frequency standard, or through a magnetic dipole decay from the $^3P_1$ level. The possibility of observing quantum jumps in such a three-level system with only one driven transition has been discussed theoretically [24] and has been realized in the Hg^+ ion [25].

In a simple rate equation model let A_0 be the decay rate from ${}^3P_0 \to {}^1S_0$, A_1 that from ${}^3P_1 \to {}^1S_0$, and A_2 that from ${}^3P_1 \to {}^3P_0$. Since $A_1 \gg A_0, A_2$ the fluorescence intensity which is proportional to $A_1\rho_{11}$ (ρ_{11}, population of ${}^3P_1$) switches from "on" to "off" at a rate $A_2\rho_{11}$ and back "on" at the rate A_0. The magnetic dipole decay rate A_2 for the transition ${}^3P_1 \to {}^3P_0$ at $\lambda = 9.32$ μm wavelength can be estimated as [26]

$$A_2 = \frac{2\pi e^2 h}{9\epsilon_0 m_e^2 c^2 \lambda^3} = 0.0223\ \mathrm{s}^{-1} . \tag{3}$$

The branching ratio A_1/A_2 is about 10^8 and with the beginning saturation of the cooling transition ($\rho_{11} = 0.25$) we expect one ${}^3P_1 \to {}^3P_0$ decay every 3 min.

To observe these dark periods in the experiment we counted the photomultiplier pulses in time intervals of 18 ms and stored these numbers in the computer. The laser power was measured synchronously and checked to be constant. Samples of these data exhibiting quantum jumps are shown in Fig. 4. We see an abrupt decrease of the count rate to the stray-light level and the reappearence of the fluorescence after some 100 ms. These events occur at a rate that is a little lower but still consistent with the estimate given above. The duration of the dark periods can be used to evaluate the lifetime of the ${}^3P_0$ level. We observed about 150 decays and calculated from these data the function $d(t)$ as the number of dark periods with a duration shorter than t. This function was fitted to $d(t) = a(1 - e^{-(t-t_0)/\tau})$ to get the lifetime τ. Events with a duration shorter than $t_0 = 70$ ms were not considered, to insure the statistical significance of every event. Figure 5 shows a logarithmic plot of $a - d(t) = ae^{-(t-t_0)/\tau}$ together with the fit, indicating a simple exponential behavior. The result for the lifetime of ${}^3P_0$ is

$$\tau({}^3P_0) = 0.14(2)\ \mathrm{s} .$$

The uncertainty results from the small number of observed decays. It was determined from the distribution of decay constants found in 100 data sets of the same size as the measured one, generated in a Monte Carlo simulation of an exponential decay. Among the quantum jumps evaluated in Fig. 5 about 5% were recorded with an ${}^{113}\mathrm{In}^+$ ion, the others with ${}^{115}\mathrm{In}^+$. According to theory (see Sec. VI) the lifetimes in the two isotopes should differ by only 0.2%. We checked that separate evaluation of the ${}^{115}\mathrm{In}^+$ data alone did not change the decay constant or the mean-square deviation of the data from the fit.

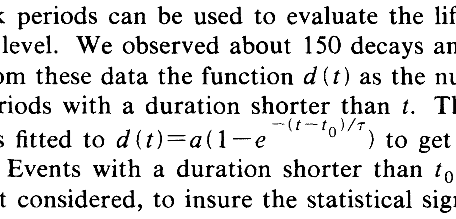

FIG. 4. Fluorescence from a single indium ion, showing dark periods due to quantum jumps to the metastable ${}^3P_0$ state.

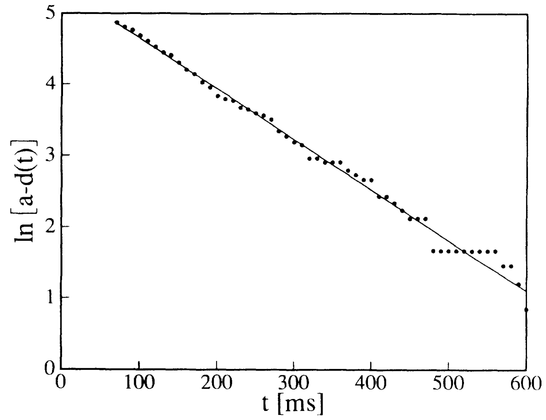

FIG. 5. Distribution of the durations of observed dark periods [see text for the definition of $a - d(t)$]. The fitted decay constant is the inverse lifetime of the ${}^3P_0$ level.

VI. COMPARISON OF THE ${}^3P_0$ LIFETIME WITH THEORY

To compare the measured lifetime of the ${}^3P_0$ state to a theoretical estimate we used the semiempirical perturbation theory approach by Garstang [27], also outlined in [5]: the magnetic dipole component of the hyperfine interaction leads to small admixtures of the states ${}^3P_1$ and ${}^1P_1$ to ${}^3P_0$ and this gives rise to a small electric dipole moment between ${}^1S_0$ and ${}^3P_0$. The mixing coefficients are evaluated in first-order perturbation theory using the lifetimes and hyperfine splittings of the states ${}^1P_1$ and ${}^3P_1$ as input parameters. In the following we give only a brief outline of the theory and the parameters relevant to the indium ion.

The real states $|{}^1P_1'\rangle$ and $|{}^3P_1'\rangle$ are intermediately coupled and can be expressed in terms of pure LS states $|{}^{2S+1}L_J\rangle$ or pure jj states $|j_1, j_2, J\rangle$:

$$\begin{aligned} |{}^3P_1'\rangle &= \alpha|{}^3P_1\rangle + \beta|{}^1P_1\rangle = c_1|\tfrac{1}{2},\tfrac{3}{2},1\rangle + c_2|\tfrac{1}{2},\tfrac{1}{2},1\rangle , \\ |{}^1P_1'\rangle &= -\beta|{}^3P_1\rangle + \alpha|{}^1P_1\rangle = c_2|\tfrac{1}{2},\tfrac{3}{2},1\rangle - c_1|\tfrac{1}{2},\tfrac{1}{2},1\rangle . \end{aligned} \tag{4}$$

The mixing coefficients [28] are calculated using the ratio of the lifetimes $\tau({}^1P_1) = 0.90$ ns [29] and $\tau({}^3P_1) = 0.44$ μs: $c_1 = 0.51$, $c_2 = 0.86$, $\beta = -0.079$, $\alpha = (1 - \beta^2)^{1/2}$. The perturbed ${}^3P_0$ state is described by

$$|{}^3P_0'\rangle = |{}^3P_0\rangle + \alpha_0|{}^3P_1'\rangle + \beta_0|{}^1P_1'\rangle . \tag{5}$$

The mixing is diagonal in F, so only the components $F = I$ of ${}^3P_1$ and ${}^1P_1$ contribute. The mixing coefficients are [28]

$$\alpha_0=\frac{\sqrt{I(I+1)}}{E(^3P_0)-E(^3P_1')}\left[\frac{c_2}{2}(a_s-a_{1/2})+\sqrt{2}c_1a'''\right],$$
$$\beta_0=\frac{\sqrt{I(I+1)}}{E(^3P_0)-E(^1P_1')}\left[\frac{c_1}{2}(a_{1/2}-a_s)+\sqrt{2}c_2a'''\right] \tag{6}$$

in terms of the single-electron hyperfine coefficients a, the nuclear spin I, and the level energies E. The coefficients a were calculated from the hyperfine splittings in the In^+5s5p configuration [13,14] using the Breit-Wills theory [30] for hyperfine structure in intermediate coupling: $a_s=0.64$ cm^{-1}, $a_{3/2}=0.014$ cm^{-1}, $a_{1/2}=0.091$ cm^{-1}, $a'''=-0.005$ cm^{-1}. With the energies [13] $E(^1P_1')=63\,034$ cm^{-1}, $E(^3P_1')=43\,350$ cm^{-1}, and $E(^3P_0)=42\,276$ cm^{-1} the mixing coefficients are $\alpha_0=-1.1\times10^{-3}$ from $^3P_1$ and $\beta_0=3.5\times10^{-5}$ from $^1P_1$. The lifetime $\tau(^3P_0)$ can now be calculated as

$$\tau(^3P_0)=\left[\frac{E(^3P_1')}{E(^3P_0)}\right]^3\frac{\beta^2}{(\alpha_0\beta+\beta_0\alpha)^2}\tau(^3P_1')=0.20\text{ s}\,. \tag{7}$$

The uncertainty of the experimental parameters that enter the calculation leads to an uncertainty of about ±0.03 s for $\tau(^3P_0)$. The result is valid for both stable isotopes ^{113}In and ^{115}In since they have the same nuclear spin and their magnetic moments differ only by 0.2% [15].

The theoretical lifetime is slightly larger but still in reasonable agreement with the experimental value. Possibly the calculation underestimates the admixture of the $^1P_1$ level because its hyperfine splitting is known with poor accuracy compared to that of the $^3P_1$ level [13,14]. Effects that shorten the experimentally measured lifetime seem unlikely. At the typical background pressure of 6×10^{-8} Pa collisional quenching of the $^3P_0$ state should be negligible.

A similar perturbational calculation can also be used to estimate the g factor of the $^3P_0$ state. It differs from the pure nuclear g factor of the $^1S_0$ ground state because of the admixtures also responsible for the decay of $^3P_0$. This effect is important for the optical frequency standard since it leads to a small anomalous Zeeman effect of the $^1S_0\rightarrow{}^3P_0$ transition. It has been investigated with high precision in the case of mercury [31]. To first order in α_0 and β_0 the g factor of the perturbed $^3P_0$ state is given by

$$g(^3P_0)=g(^1S_0)+\left\{\frac{8}{3I(I+1)}\right\}^{1/2}(\alpha_0a-\beta_0\beta)\,. \tag{8}$$

The ground-state g factor for indium is known from a NMR measurement [32]: $g(^1S_0)=-0.664\times10^{-3}$ and the calculation yields $g(^3P_0)=-1.01\times10^{-3}$. The corresponding field dependence of the π components $(^1S_0,m_F=\pm\frac{1}{2})\rightarrow(^3P_0,m_F=\pm\frac{1}{2})$ is 240 Hz/G. Consequently for a projected relative uncertainty of 10^{-18} in the transition frequency [1] a control or shielding of the magnetic field in the microgauss range is required. A calculation using rate equations indicates that the ion can be optically pumped into the $m_F=\pm\frac{1}{2}$ sublevels of the ground state with 84% probability by exciting the hyperfine component $(^1S_0,F=\frac{9}{2})\rightarrow(^3P_1,F=\frac{9}{2})$ of the cooling transition with linear polarized light below saturation.

VII. CONCLUSION

We presented the indium ion as a possible candidate for an optical frequency standard. Although the cooling transition in indium is a relatively slow intercombination line with only 360 kHz natural linewidth we were able to cool a single ion using this transition and to obtain a rate of fluorescence photons that is high enough to detect the dark periods due to electron shelving in the $^3P_0$ level. Besides these technical difficulties the small linewidth of the cooling transition enabled us to reach the strong binding regime using moderate trapping field strength. This opens the possibility of cooling the ion into low quantum levels or even the ground state of the single ion oscillator.

A frequency standard will benefit from the insensitivity of the indium ion's $^1S_0\rightarrow{}^3P_0$ transition frequency against external perturbations as well as from the availability of a frequency stable oscillator like a diode laser pumped Nd:YAG laser. A suitable source has been developed in our laboratory for this purpose [33]. The accuracy to which the wavelength of this forbidden transition is known has recently been improved significantly by the measurement of Larkins and Hannaford [14], so that direct laser excitation shall be possible in the near future.

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