

## Radiative Decay Rates in $\text{Hg}^+$ from Observations of Quantum Jumps in a Single Ion

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Radiative decay rates connecting the lowest four energy levels of  $^{198}\text{Hg}^+$  have been derived solely from an analysis of the fluctuations (quantum jumps) of the laser-induced fluorescence of the 194-nm first resonance transition of a single ion confined in a Paul trap. The natural linewidth of the 194-nm first resonance transition was also measured. The measured decay rates and branching ratios are in satisfactory agreement with theory.

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Recently it has become possible to observe for extended periods the fluorescence from a single, isolated ion, kept in place by the electric fields of an ion trap. This capability has made it possible to study quantum jumps (sudden transitions between atomic energy levels)<sup>1-3</sup> and nonclassical photon statistics (photon antibunching).<sup>3,4</sup> In this Letter we report measurements of five radiative decay rates connecting the lowest four energy levels of  $^{198}\text{Hg}^+$ . These measurements depend on the observation of transition between the energy levels of a single ion, as exhibited by the changes in its fluorescence. This would not be possible with a sample of many ions observed simultaneously. Related methods have previously been used to measure lifetimes of metastable levels in  $\text{Ba}^+$  (Refs. 1 and 2) and  $\text{Hg}^+$  (Ref. 3).

An unusual feature of the techniques used in the present work is that decay branching ratios are measured without our observing the decay photons directly. It is not necessary to calibrate the detector efficiency, and it is necessary only to observe one wavelength. Instrumental noise, such as that due to changes in the laser intensity or that due to statistical fluctuations in the numbers of photons detected, is unimportant compared to the inherent quantum mechanical randomness in the behavior of a single atom. This property is shared by the quantized optical double resonance technique recently used to observe narrow resonances in a single  $\text{Hg}^+$  ion.<sup>5</sup>

The lowest four energy levels of  $\text{Hg}^+$  are shown in Fig. 1. After the ion is excited from the ground  $5d^{10}6s^2S_{1/2}$  level to the  $5d^{10}6p^2P_{1/2}$  level, it usually decays back to the ground state by electric dipole ( $E1$ ) radiation, but has a small probability of undergoing an  $E1$  decay to the metastable  $5d^96s^2D_{3/2}$  level. This probability is small because it requires configuration mixing in order to occur (the nominal configurations differ in two orbitals). In addition, the transition rate is proportional to the cube of the energy difference between the  $^2P_{1/2}$  and the  $^2D_{3/2}$  energy levels, which is small compared to that between the  $^2P_{1/2}$  and the  $^2S_{1/2}$  energy levels. From the  $^2D_{3/2}$  level, the ion decays either directly to the ground state, by electric quadrupole ( $E2$ ) radiation, or

to the metastable  $5d^96s^2D_{5/2}$  level, by magnetic dipole ( $M1$ ) and  $E2$  radiation. According to calculations,<sup>6</sup> the  $E2$  decay rate to the  $^2D_{5/2}$  level is negligible compared to the  $M1$  rate. The  $^2D_{5/2}$  level decays to the ground state by  $E2$  radiation. According to simple estimates,<sup>7</sup> the magnetic quadrupole ( $M2$ ) and electric octupole ( $E3$ ) allowed decay rate from the  $^2P_{1/2}$  level to the  $^2D_{5/2}$  level is much too small to observe in the present experiment.

The apparatus has been described previously.<sup>3,8</sup> The  $\text{Hg}^+$  ions were confined in a radio-frequency (Paul) trap under high-vacuum conditions. The ions were loaded into the trap by ionization of Hg atoms inside the trap volume with an electron beam. A beam of narrow-band cw 194-nm radiation of about 5  $\mu\text{W}$  was focused to a waist size  $w_0$  of approximately 5  $\mu\text{m}$  at the position of the ions. The frequency of the 194-nm radiation was tuned below the resonance, in order to provide laser cooling of the ions. The 194-nm fluorescence photons were collected by a system of lenses and focused on the Cs-Te photocathode of a solar-blind photomultiplier tube. The numbers of photons counted in consecutive time intervals

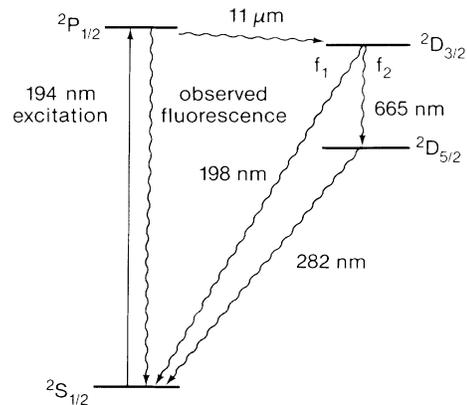


FIG. 1. Diagram of the lowest four energy levels of  $\text{Hg}^+$ . The decay rates from all three excited levels were determined by observation of only the 194-nm fluorescence.

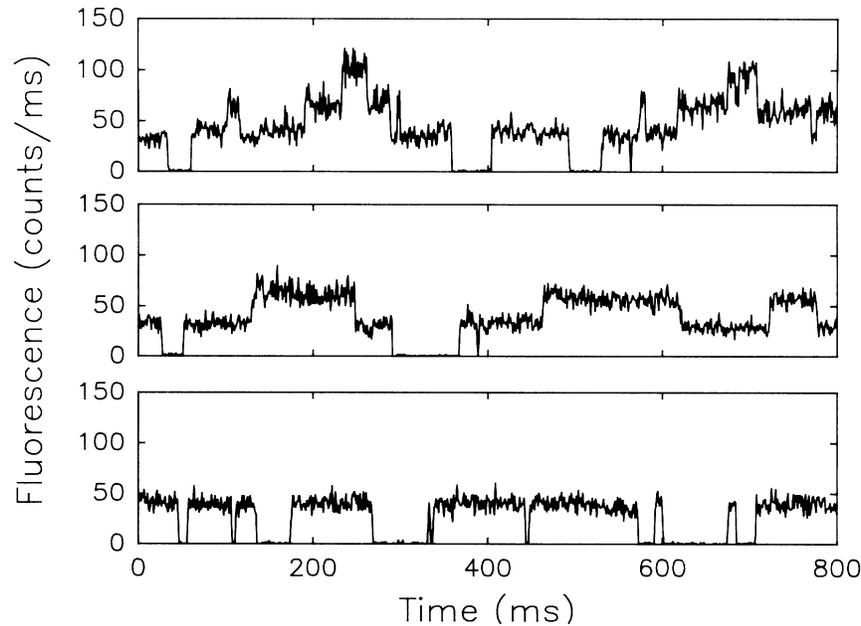


FIG. 2. Intensity of 194-nm laser-induced fluorescence as a function of time for samples of three ions (highest trace), two ions (middle trace), and one ion (lowest trace) in the trap. The integration time per point was 1 ms, and the points were connected by straight lines.

were recorded by a computer.

Figure 2 shows traces of the fluorescence intensities as functions of time when three ions, two ions, and one ion were in the trap and irradiated with radiation near the  $^2S_{1/2}$ -to- $^2P_{1/2}$  transition frequency. When a single ion was in the trap, as in the lowest trace in Fig. 2, the 194-nm fluorescence intensity had two observable states. When the ion was cycling between the  $^2S_{1/2}$  level and the  $^2P_{1/2}$  level (the "on" state), the fluorescence intensity had a constant value; when it was in either the  $^2D_{3/2}$  or the  $^2D_{5/2}$  metastable level (the "off" state), it was zero. When more than one ion was in the trap, as in the highest and the middle traces of Fig. 2, the fluorescence intensity at a given time was proportional to the number of ions that were in the on state at that time, so that it took one of several discrete values.

The total radiative decay rate of the  $^2P_{1/2}$  level ( $\gamma_P$ ) was determined by measurement of the width of the 194-nm transition. The decay rate  $\gamma_P$ , in units of inverse seconds, is equal to the natural linewidth, in units of radians per second. Only the low-frequency half of the resonance was observed, since the ion heated rapidly when the frequency of the 194-nm radiation was above the resonance center.<sup>9,10</sup> The frequency of the 194-nm radiation was swept slowly enough that the transitions between the on and off states were clearly resolved, and a smooth curve was interpolated through the on-state data. The effect of power broadening was removed by an extrapolation to zero intensity of the 194-nm radiation. A similar method has been used previously by Nagourney,

Janik, and Dehmelt to measure the natural linewidth of the 280-nm resonance line of  $Mg^+$ .<sup>10</sup> The temperature was measured in a separate experiment to be a few millikelvins,<sup>5</sup> for the range of frequency detunings required for this line-shape analysis. This temperature was low enough that the Doppler broadening was not significant. The result was  $\gamma_P = (3.8 \pm 0.6) \times 10^8 \text{ s}^{-1}$ .

The radiative decay rate from the  $^2P_{1/2}$  level to the  $^2D_{3/2}$  level [ $\gamma(P \rightarrow D)$ ] was determined by measurement of the rate of transitions from the on state to the off state for a single ion as a function of the 194-nm fluorescence intensity. The transition rate  $R_+$  from the on state to the off state was obtained by division of the number of transitions in a run by the time that the ion was in the on state in that run. This rate is equal to the product of  $\gamma(P \rightarrow D)$  and  $P(^2P_{1/2})$ , the mean population of the  $^2P_{1/2}$  level when the ion is in the on state. The fluorescence intensity in the on state is also proportional to  $P(^2P_{1/2})$ .  $P(^2P_{1/2})$  could be varied by variation of either the intensity or the frequency detuning from resonance of the 194-nm radiation. It was found experimentally that  $R_+$  was proportional to the observed photon counting rate in the on state. A calibration of the value of  $P(^2P_{1/2})$  was made by measurement of the fluorescence intensity near resonance for different intensities of the incident 194-nm radiation. The value of  $P(^2P_{1/2})$  at resonance is equal to  $(2+1/S)^{-1}$ , where  $S$ , the saturation parameter, is a quantity proportional to the incident 194-nm intensity. Figure 3 is a plot of  $Y$ , the observed counting rate at resonance, as a function of  $X$ , the 194-

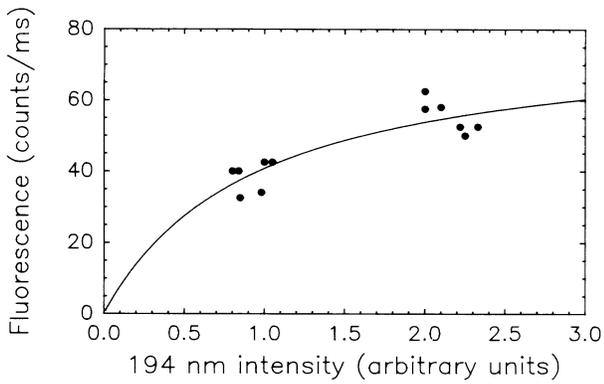


FIG. 3. Fluorescence intensity at resonance of a single  $\text{Hg}^+$  ion as a function of the intensity of the 194-nm radiation (dots) and a least-squares fit (curve).

nm intensity in arbitrary units. The solid curve is a least-squares fit of the data by the function  $Y = A(2 + B/X)^{-1}$ . The value of  $P(^2P_{1/2})$  for a given 194-nm intensity and detuning is the observed counting rate divided by  $A$ . The value of  $\gamma(P \rightarrow D)$  was determined by division of the value of  $R_+$  for a particular run by the value of  $P(^2P_{1/2})$  for the same run. The result was  $\gamma(P \rightarrow D) = 52 \pm 16 \text{ s}^{-1}$ . The uncertainty in the determination of  $A$  from the least-squares fit and the uncertainty in the determination of  $R_+$  contributed about equally to the uncertainty in  $\gamma(P \rightarrow D)$ .

The average behavior of the ion from the time it decays to the  $^2D_{3/2}$  level until it returns to the ground state is governed by the rate equations for the probabilities  $P_1$ ,  $P_2$ , and  $P_3$  of being in the  $^2D_{3/2}$  level, the  $^2D_{5/2}$  level, and the  $^2S_{1/2}$  level, respectively:

$$dP_1(t)/dt = -\gamma_1 P_1(t), \quad (1)$$

$$dP_2(t)/dt = f_2 \gamma_1 P_1(t) - \gamma_2 P_2(t), \quad (2)$$

$$dP_3(t)/dt = f_1 \gamma_1 P_1(t) + \gamma_2 P_2(t). \quad (3)$$

Here  $\gamma_1$  and  $\gamma_2$  are the total radiative decay rates of the  $^2D_{3/2}$  level and the  $^2D_{5/2}$  level respectively. The probabilities for the  $^2D_{3/2}$  level to decay to the  $^2S_{1/2}$  level or to the  $^2D_{5/2}$  level are  $f_1$  and  $f_2$  ( $f_1 + f_2 = 1$ ). The initial conditions are  $P_1(0) = 1$  and  $P_2(0) = P_3(0) = 0$ . The quantity observed experimentally is the probability distribution  $W_{\text{off}}(\tau)$  of the duration of off periods as a function of the duration  $\tau$ . This quantity is proportional to  $dP_3(t)/dt$  evaluated at  $t = \tau$ , which is the average rate at which the ion returns to the ground state after a period  $\tau$ . Solving Eqs. (1)–(3) yields the expression

$$W_{\text{off}}(\tau) \propto f_2 \gamma_2 \exp(-\gamma_2 \tau) + (f_1 \gamma_1 - \gamma_2) \exp(-\gamma_1 \tau). \quad (4)$$

The distribution of about 20000 off periods is shown in Fig. 4, together with a least-squares fit by a sum of two

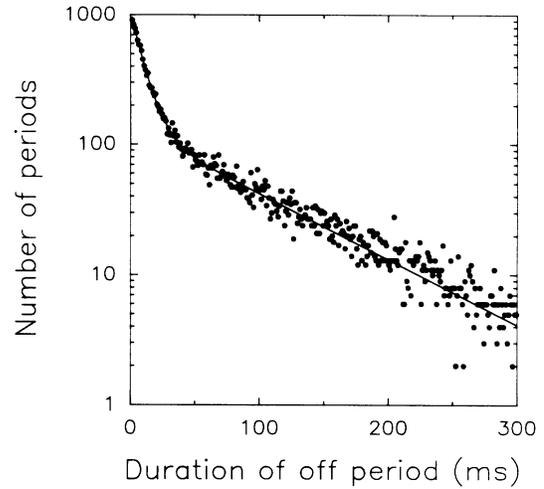


FIG. 4. Distribution of fluorescence-off periods for a single ion (dots) and a least-squares fit (curve). These data represent an analysis of approximately 40000 quantum jumps (up or down).

decaying exponentials. The values of the parameters obtained from the fit are  $\gamma_1 = 109 \pm 5 \text{ s}^{-1}$ ,  $\gamma_2 = 11.6 \pm 0.4 \text{ s}^{-1}$ , and  $f_1 = 0.491 \pm 0.015$ . The partial decay rates of the  $^2D_{3/2}$  level are  $f_1 \gamma_1 = 53.5 \pm 2.0 \text{ s}^{-1}$  and  $f_2 \gamma_1 = 55.5 \pm 4.0 \text{ s}^{-1}$ .

In order to place a limit on the effects of collisions on the measured lifetimes, data were collected at higher pressure. No shift of the decay rates of the metastable levels was observed, even when the pressure of the background gas, consisting mostly of Hg, was increased by over a factor of 10 above the normal level. The relative pressure was estimated from the time required to create an ion in the trap by electron-impact ionization and also from the decrease in the confinement time of the ion in the trap. The latter measurement was based on the observation that ions were lost from the trap when both 194-nm resonance radiation and neutral Hg atoms were present. We think that this process was caused by reactions of excited-state ions with neutral Hg atoms. The kinetic energy given to an ion by such a reaction could have been sufficient to cause it to be lost from the trap. All other perturbing effects, such as transitions out of the metastable levels induced by the 194-nm radiation or the trap electric fields, are estimated to have been negligible.

The present measurement of  $\gamma_P$  agrees with a previous beam-foil measurement of  $(4.3 \pm 0.7) \times 10^8 \text{ s}^{-1}$  by Erikson and Poulsen<sup>11</sup> but disagrees somewhat with a previous optical-absorption measurement of  $2.65 \times 10^8 \text{ s}^{-1}$  by Bruneteau *et al.*<sup>12</sup> An unpublished calculation by Al-Salameh and Silfvast<sup>13</sup> (AS) gives a value of  $5 \times 10^8 \text{ s}^{-1}$ , in fair agreement with the present measurement. Other theoretical values are given in Ref. 11. No previous measurements of  $\gamma(P \rightarrow D)$  have been reported. The present value is in good agreement with the value of

$55 \text{ s}^{-1}$  calculated by AS.<sup>13</sup> No previous measurements of the partial decay rates  $f_1\gamma_1$  and  $f_2\gamma_1$  of the  ${}^2D_{3/2}$  level have been reported. Johnson<sup>14</sup> has reported a value for the total decay rate  $\gamma_1$  of  $50 \pm 5 \text{ s}^{-1}$ , which disagrees with the present value. Garstang<sup>6</sup> has calculated the decay rate  $f_1\gamma_1$  to be  $54 \text{ s}^{-1}$ , in very good agreement with the present value. The other decay rate  $f_2\gamma_1$  has been calculated by Garstang<sup>6</sup> to be  $42 \text{ s}^{-1}$  and by AS<sup>13</sup> to be  $75.6 \text{ s}^{-1}$ . The present value is in fair agreement with both calculations. Two previous measurements of the  ${}^2D_{5/2}$  level decay rate  $\gamma_2$  have been reported by Bergquist *et al.*<sup>3,8</sup> The values obtained by the two methods were  $11 \pm 2 \text{ s}^{-1}$  (Ref. 8) and  $11 \pm 4 \text{ s}^{-1}$  (Ref. 3), in good agreement with the present value. A previous measurement by Johnson<sup>14</sup> of  $\gamma_2 = 10.2 \pm 0.5 \text{ s}^{-1}$  disagrees slightly with the present value. The values of  $\gamma_2$  calculated by Garstang<sup>6</sup> ( $9.5 \text{ s}^{-1}$ ) and by AS<sup>13</sup> ( $12.6 \text{ s}^{-1}$ ) are in fair agreement with the present value.

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