The Observation of Quantum Jumps in Hg^+

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Introduction

Quantum jumps (sudden changes of quantum state) have been observed in isolated samples of one or a few atomic ions [1-4]. The method of detecting quantum jumps uses a kind of optical-optical double resonance originally proposed by Dehmelt [5]. Consider an atom, such as Hg^+, which has both a strongly allowed transition (at wavelength λ_1) and a weakly allowed transition (at wavelength λ_2) from the ground state (see Fig. 1). Assume that radiation is present at both λ_1 and λ_2. The atom fluoresces strongly at λ_1 until it absorbs a λ_2 photon and makes a transition to the metastable state. The λ_1 fluorescence is shut off until the atom decays to the ground state.

The first experiments were concerned mainly with verifying that quantum jumps did occur [1-3]. For a single ion, this means that the fluorescence from the strong transition switches back and forth between zero and a steady value rather than assuming some constant intermediate value. Values of metastable lifetimes were obtained from the time intervals between quantum jumps, but these values were no more accurate than those obtained previously. Here we report the use of quantum jumps in measuring radiative decay rates and branching ratios which had not been known accurately. We also report their use as a means of detection in high-resolution spectroscopy.

Hg^+ System

The lowest energy levels of Hg^+ are shown in Fig. 1. The $5d^9 6s^2\ ^2D_{3/2}$ and $5d^9 6s^2\ ^2D_{5/2}$ states are predicted to be metastable [6]. Once excited to the $5d^{10} 6p\ ^2P_{1/2}$ state, the atom usually decays back to the $5d^{10} 6s\ ^2S_{1/2}$

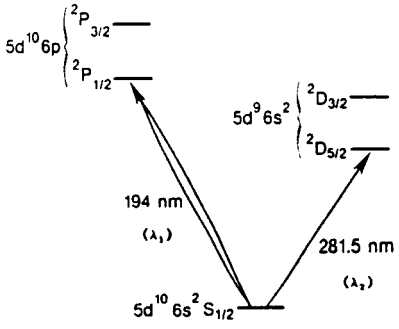

Fig. 1. Energy levels of Hg^+

ground state, but it has a probability, estimated to be about 10^{-7} [7], of decaying to the $^2D_{3/2}$ state. The probability is small because the transition frequency is low and because it requires configuration mixing to occur (the nominal configurations differ by two orbitals). From the $^2D_{3/2}$ state, the atom decays either directly to the ground state with probability f_1 or to the $^2D_{5/2}$ state with probability $f_2 = 1-f_1$. According to theory, $f_1 \approx f_2$ [6].

Apparatus

The apparatus has been described previously [2,8]. The Hg^+ ions are confined in a radiofrequency (Paul) trap under ultra-high-vacuum conditions. A single Hg^+ ion has been kept in the trap for over a week. The cw 194 nm radiation required to excite the $^2S_{1/2}$ to $^2P_{1/2}$ transition was obtained by sum-frequency mixing the output of a frequency doubled 514.5 nm Ar^+ laser with a 792 nm dye laser in a potassium pentaborate crystal [9]. The 281.5 nm radiation required to excite the $^2S_{1/2}$ to $^2D_{5/2}$ transition was obtained by frequency doubling the output of a dye laser. The 194 nm fluorescence was collected by a lens system and detected by a photomultiplier tube.

Analysis of Single-Ion Quantum Jumps - Lifetimes and Branching Ratios

Typical fluorescence data are shown in Fig. 2 for one, two, and three ions. The quantum jumps are clearly visible and occur in a time of less than 1 ms, which is the spacing between data points. For these data, only the 194 nm radiation was present. The intensity was high enough that quantum jumps due to the weak $^2P_{1/2}$ to $^2D_{3/2}$ decay occur several times per second.

Let P_1, P_2, and P_3 be the probabilities that an Hg^+ ion is in the $^2D_{3/2}$, the $^2D_{5/2}$, and the $^2S_{1/2}$ states, respectively. Let γ_1 and γ_2 be the total radiative decay rates of the $^2D_{3/2}$ and the $^2D_{5/2}$ states, respectively. The ensemble averaged behavior of one atom after it decays to the $^2D_{3/2}$ state can be derived from the following set of rate equations:

$$dP_1(t)/dt = -\gamma_1 P_1(t), \tag{1}$$

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

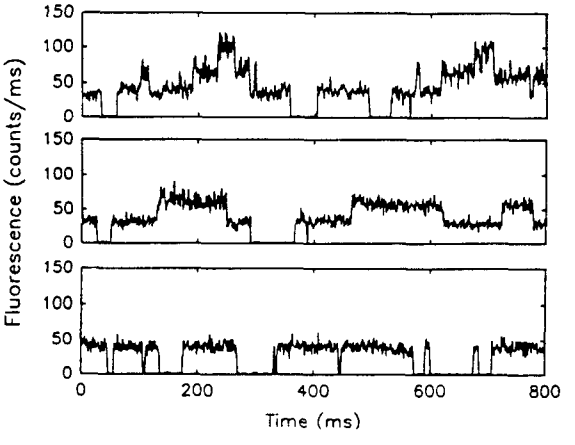

Fig. 2. Fluorescence intensity as a function of time for three ions (top), two ions (middle), and one ion (bottom), showing the quantum jumps

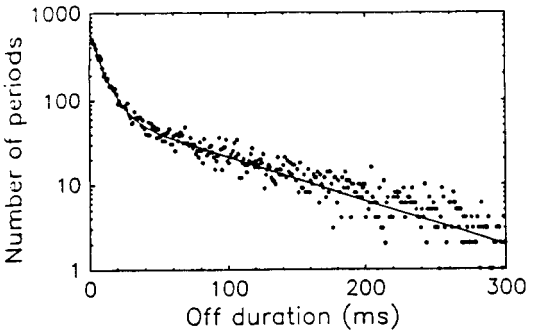

Fig. 3. Distribution of fluorescence-off periods for a single ion (dots) and least-squares fit (solid curve)

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

The quantity which is measured is the probability distribution $W_{off}(\tau)$ of a fluorescence-off period having a duration τ. This is proportional to $dP_3(t)/dt$ evaluated at $t=\tau$ with the initial conditions $P_1(0) = 1$ and $P_2(0) = P_3(0) = 0$. Solving Eqs. (1-3) yields

$$W_{off}(\tau) = \text{const.}[f_2\gamma_2\exp(-\gamma_2\tau) + (f_1\gamma_1-\gamma_2)\exp(-\gamma_1\tau)]. \qquad (4)$$

The experimental fluorescence-off distribution was least-squares fitted to Eq. (4) to obtain values for γ_1, γ_2, and f_1. Figure 3 shows the data and the least-squares fit. The values obtained from the fit are $\gamma_1=112 \pm 7\ s^{-1}$, $\gamma_2=12.0 \pm 0.6\ s^{-1}$, and $f_1=0.498 \pm 0.031$. These values are in good agreement with calculations [6]. The value of γ_2 is in good agreement with previous measurements [2,8,10], but the value of γ_1 is about a factor of 2 higher than the only previously reported value [10]. No previous measurements of f_1 exist.

Application of Quantum Jumps to Spectroscopy - Quantized Detection

We have used the bistable nature of the single-ion fluorescence intensity (high when the ion is cycling between the $^2S_{1/2}$ and $^2P_{1/2}$ states and zero when it is in a metastable state) to detect the weak $^2S_{1/2}$ to $^2D_{5/2}$ transition [11]. The measurement cycle was as follows: If the fluorescence was high enough to indicate that the ion was cycling between the $^2S_{1/2}$ and $^2P_{1/2}$ states, the 194 nm radiation was turned off and the 281.5 nm radiation was turned on for 20 ms, which is much less than the $^2D_{5/2}$ state lifetime.

The 194 nm radiation was turned on again, and the fluorescence photons were counted for 10 ms to see whether the ion had made a transition to the $^2D_{5/2}$ state. We have used the quantum multiplication aspects of double-resonance detection previously [12]. The new feature was that the result of the measurement was a 0 or a 1, depending on whether the number of photons detected was below or above a threshold level. This eliminated certain kinds of instrumental noise, such as intensity fluctuations of the detection laser, leaving only the inherent quantum fluctuations of the atom. Resonance linewidths as small as 30 kHz were observed at 281.5 nm, and the motional sidebands were clearly resolved. Motional sidebands are caused by the frequency modulation of the laser frequency in the frame of the ion by the Doppler effect. They are offset from the central resonance by multiples of the frequencies of harmonic motion of the ion. The intensities of the motional sidebands indicated that the ion had been cooled to near the theoretical minimum of 1.7 mK.

Quantum Jumps of Two or Three Ions

We have observed quantum jumps with two and three ions (see Fig. 2). Others have reported that multiple jumps (simultaneous quantum jumps of two or more ions) occur frequently in a sample of Ba^+ ions [4]. They attribute the multiple jumps to a collective interaction of the ions with the light field. We find that the Hg^+ ions act independently. Apparent multiple jumps occur only at a rate which is consistent with the finite time resolution of the apparatus.

We gratefully acknowledge the support of the Air Force Office of Scientific Research and the Office of Naval Research.

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