

Observation of Quantum Jumps

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We have recorded the laser-excited resonance fluorescence of one to three Ba^+ ions and observed Bohr's "quantum jumps" when an ion decayed to the metastable $^2D_{5/2}$ state, suddenly quenching its fluorescence. Most of the jumps mark Raman-Stokes scattering from the $^2S_{1/2}$ and $^2D_{3/2}$ levels.

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In 1913, Bohr suggested that the interaction of light and matter happens in such a way that an atom undergoes instantaneous transitions of its internal state upon the emission or absorption of a light quantum.¹ These transitions have become known as "quantum jumps." They were mostly considered, at that time and even later, a kind of artifact of Bohr's model of the atom whose real existence was questionable and, anyway, not to be verified with the common large atomic ensembles. Recently the issue has been reopened, since available coherent light sources, as well as single ions prepared in ion traps^{2,3} and optically cooled,^{4,5} seemed to allow an experimental demonstration. The typical concept for this observation is based upon a weak absorption line, coupled, via the ground state or a metastable state, to a strong resonance line in a V-shaped three-level system of an individual atomic particle.⁶⁻¹⁰ On the strong line, resonance fluorescence is laser excited, which permits the particle to scatter continually up to 10^9 quanta per second. Occasional excitation of the *weak* transition, and the concomitant presence of the atomic system in the corresponding upper state, will initiate immediate quenching of the strong resonance fluorescence, if the absorption on the weak line is indeed accompanied by a quantum jump of the atom. Since the absorption of classical radiation involves the excitation and synchronization of the induced atomic dipole, the response may be slightly less abrupt.

A related concept even closer to Bohr's original idea makes use of weak *spontaneous emission*, coupled to the *upper* level of the strong resonance line. Quantum jumps of the atomic particle into an intermediate third level of long enough life will show up as random extinction of the strong resonance scattering. This concept has also been discussed recently.^{9,10}

We have observed quantum jumps of individual barium ions, marked as steps in their $^2P_{1/2}$ - $^2S_{1/2}$ resonance fluorescence, excited by intense laser light. The scattering abruptly vanishes upon transitions into the metastable $^2D_{5/2}$ level (see Fig. 1).

The essentials of the experimental apparatus have been described elsewhere.² Small clouds of one, two,

or three Ba^+ ions are confined in a miniaturized rf ion trap (cap distance 0.5 mm).¹¹ If optical cooling is perfect ($T < 10$ mK for single ion), their orbit sizes are $< 0.1 \mu\text{m}$, $4 \mu\text{m}$, and $9 \mu\text{m}$, respectively. The ions are simultaneously irradiated by the cw light of two dye lasers, Coumarine 102 at $\lambda = 493$ nm, and DCM at $\lambda' = 650$ nm, focused by an $f = 10$ cm lens to a spot of approximately $70\text{-}\mu\text{m}$ size. The red light serves to reexcite the ion(s) when they have decayed into the metastable $^2D_{3/2}$ state, i.e., to couple this level to the $^2P_{1/2}$ level. Both transitions are strongly saturated. Thus, most of the time the ions are nearly equally partitioned among the states $^2S_{1/2}$, $^2P_{1/2}$, and $^2D_{3/2}$, provided the particular tuning of the light frequencies avoids excitation of the two-photon (Raman, or "dark") resonance—and possibly pertinent higher-order resonances.

Resonance fluorescence is collected by a high-speed binary microscope, which permits simultaneous visual control, and is measured by a low-noise photomultiplier and a photon-counting system. The maximum signal was 4000 counts per ion per second. The green fluorescence was recorded in a series of scans of either

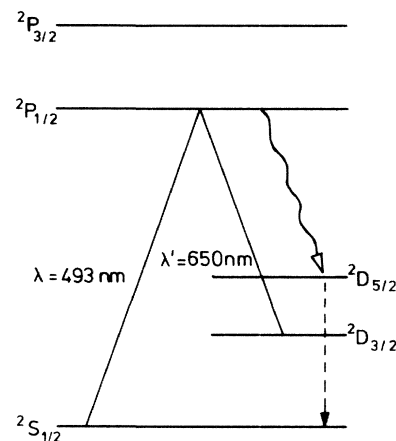


FIG. 1. Partial energy level scheme of Ba^+ , and its interaction with light. The dashed arrow indicates radiative and collisional deexcitation of the metastable level $^2D_{5/2}$.

the green or the red laser across the respective lines, while the other one was tuned down from resonance center by 300 MHz in order to provide for steady optical cooling,^{4,5} or with fixed tuning of the light. An example, with the red light scanned, is shown in Fig. 2. The maximum fluorescence is due to three ions; approximate envelopes for one and two ions are also indicated. The width of this excitation spectrum is determined by power broadening and to a lesser extent by residual Doppler broadening as a consequence of the radiative and collisional processes described below.

The conspicuous steps mark the appearance and disappearance of an individual ion from visibility by its resonance fluorescence. An event of *disappearance* occurs when a well-cooled ion, insensitive to collisional heating at the background pressure 10^{-8} mbar, decays into the metastable $^2D_{5/2}$ level. This is mostly due to electronic Raman-Stokes scattering far off the intermediate $^2P_{3/2}$ level (numerical estimate for scattering from $^2S_{1/2}$ and $^2D_{3/2}$ levels: $3 \times 10^{-3} \text{ s}^{-1}$), and to analogous collision-induced processes. Also, small even-parity contributions may be mixed into the $^2P_{1/2}$ level by the strong light fields and allow some $E2$ decay to the $^2D_{5/2}$ level. Residing there, the ion does not partake in optical cooling which affects only the $S_{1/2}$, $P_{1/2}$, and $D_{3/2}$ levels. In contrast, the ion is somewhat heated by parametric coupling of its secular motion ($\nu_z = 1.3\text{--}1.6$ MHz) to the driven (or micro) motion at $\Omega/2\pi \cong 39$ MHz. With a larger orbit, it now becomes

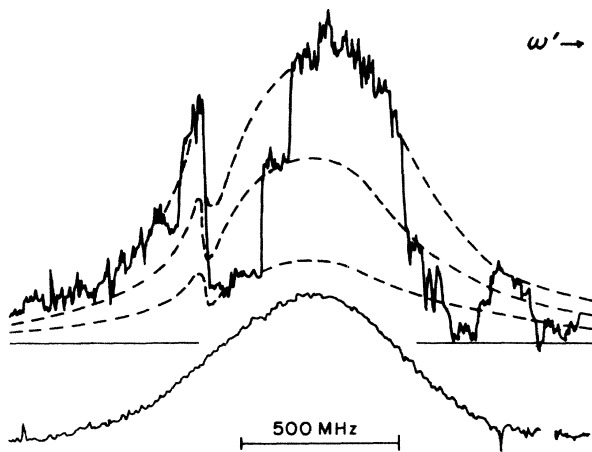


FIG. 2. Recording of the Ba^+ resonance fluorescence at 493 nm (upper trace) vs scanned frequency of red laser light (full scan $\cong 10$ min). Background pressure 10^{-8} mbar. Three ions are present in the trap. Downward steps mark transitions to $^2D_{5/2}$ accompanied by quenching of the interaction with the laser light. The feature 300 MHz below line center marks the two-photon (or Raman) resonance $^2S_{1/2}\text{--}^2D_{3/2}$. The formation of Raman coherence considerably reduces light scattering and optical cooling. The lower trace is the optical galvanic signal from a hollow-cathode discharge.

more susceptible to collisional heating. The orbit can grow even larger than the size of the light beam, and fluorescence remains absent for a substantial lapse of time. Thus, only “down” steps of the fluorescence mark quantum jumps, whereas its increments may happen much later than the radiative or collisionally induced transition from the $^2D_{5/2}$ level into the $^2S_{1/2}$ ground state, or into the $^2D_{3/2}$ state.

Note the Raman resonance showing up some 300 MHz below line center. It is marked by heating two of the ions—and the concomitant disappearance of their fluorescence—due to strongly reduced cooling when a coherent superposition of the $^2S_{1/2}$ and $^2D_{3/2}$ states is formed. Also note the more frequent occurrence of jumps on the *high*-frequency wing of the line, since there the red light is optically heating the ions, rather than cooling them as on the low-frequency wing.

Figure 3 shows the recording, versus time, of the light scattered off a single ion in a trap of 1-mm cap distance, and illuminated off the laser-beam focus. The background pressure was 8×10^{-10} mbar.

For full appreciation of the dynamics of individual trapped ions, we have to elaborate on the outlined model. Collisions with the thermal atoms ($W_{\text{kin}} \cong 50$ meV) do not suffice to heat substantially a single well-cooled ion, or even less-cooled clouds of two or three ions, in the pseudopotential well of the trap whose depth is on the order of 3 eV. Optical cooling, at line saturation, acts at a rate on the order of 14 eV/s,⁴ such that collisional heating is quickly compensated when the interaction with light continues. A $^2D_{5/2}$ ion lacks this cooling. Its radiative lifetime is 47 ± 16 s.¹² The collisional quenching rate by hydrogen is $9 \times 10^5/\text{mbar s}$.¹³

The determination of even the collisional quenching time of the $^2D_{5/2}$ level would require, for the experi-

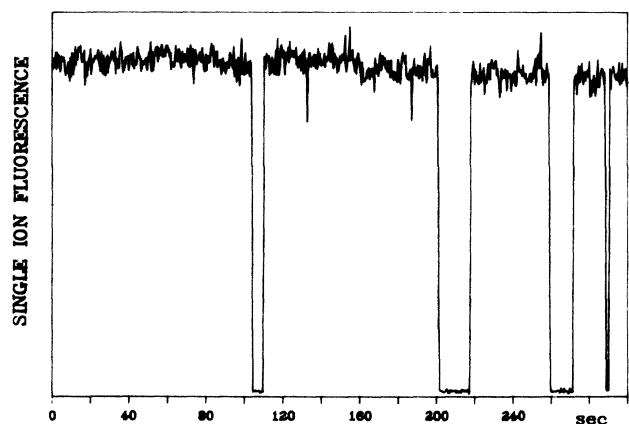


FIG. 3. Resonance fluorescence, at 493 nm, of *single* Ba^+ vs time. Green laser 300 MHz down from line center, red laser at center frequency. The signal corresponds to 4000 counts/s.

ment of Fig. 2, quantitative modeling of the collisional dynamics. This is not so, however for the preceding decay of the fluorescing states.

For evaluation of this part of the dynamics, we consider an effective two-level system,⁷ where $P_+ = P(S_{1/2}) + P(P_{1/2}) + P(D_{3/2})$ is the probability for finding the ion in a state where it interacts with the light, and P_- is the probability for the ion being in the $^2D_{5/2}$ level. The probability that no transition to this level has occurred between t and $t + T$ is, for N ions, $P_{0+}^N(t, T)$, where $P_{0+}(t, T) = \exp(-T/\tau_+)$ and τ_+^{-1} is the “+” state decay rate. Inversely, the probability for a transition to have happened in time T is $F^{(N)}(T) = 1 - P_{0+}^N$. Its derivative is the probability density of “on” times, and

$$P^{\text{on}}(T) = (dF^{(N)}/dT)\tau_+ = NP_{0+}^N$$

indicates how often an on time of length T appears. Figure 4 shows a plot of

$$\ln[P^{\text{on}}(T)/N] = -NT/\tau_+$$

for 36 on periods taken randomly from five recordings such as Fig. 2. To avoid systematic effects connected with the selection of short on times, the first interval has been neglected with the linear regression. The $1/e$ time for decay of the on state is $\tau_+ = 45 \pm 27$ s (3 standard deviations). One out of three jumps marks a radiative transition, whereas two are collisionally induced. On the other hand, evaluating 156 on times of recordings as in Fig. 3 yields $\tau_+ = 140 \pm 24$ s. Here, most of the jumps are radiative, as derived from an estimate of the Raman-Stokes rates from the intensity values of the lasers (60 times and 10^3 times saturation of the green and red resonance transitions, respectively).

In conclusion, we have demonstrated the existence of Bohr's “quantum jumps” by observing the quenching of the resonance fluorescence of individual trapped barium ions, when they undergo a very weak spontaneous transition to a level not affected by the laser light which excites the ion. We have also shown that this technique can be applied to the determination of extremely small transition rates. Some insight has been shed upon the dynamics of individual ions in a rf ion trap when simultaneously exposed to radiative and collisional interaction. An interesting question remains as to whether or not weak *absorptive* excitation gives rise to similar steps in the fluorescence output of individual particles.

We appreciate receiving manuscripts from C. Cohen-Tannoudji, J. Javanainen, and R. G. Brewer prior

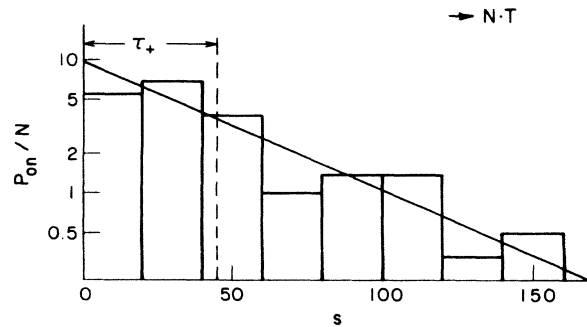


FIG. 4. Statistics of 36 on intervals of Ba^+ resonance fluorescence, as in Fig. 2. τ_+ is the mean time duration for the ion to interact with the laser light.

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Note added.—Since this Letter was submitted similar work has been reported by Nagourney, Sandberg, and Dehmelt.¹⁴

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