International Journal of Mass Spectrometry 377 (2015) 403–409

Contents lists available at ScienceDirect

International Journal of Mass Spectrometry

journal homepage: www.elsevier.com/locate/ijms

Early observations of macroscopic quantum jumps in single atoms☆

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ARTICLE INFO

Article history:
Received 26 March 2014
Received in revised form 25 June 2014
Accepted 1 July 2014
Available online 11 July 2014

Keywords:
Paul trap
Quantum jumps
Quantum optics
History of science
Mercury
Barium

ABSTRACT

The observation of intermittent fluorescence of a single atomic ion, a phenomenon better known as 'macroscopic quantum jumps,' was an important early scientific application of the three-dimensional rf quadrupole (Paul) trap. The prediction of the phenomenon by Cook and Kimble grew out of a proposal by Dehmelt for a sensitive optical double-resonance technique, called 'electron shelving.' The existence of the quantum jumps was viewed with skepticism by some in the quantum optics community, perhaps due to the failure of some conventional calculations, for example the solutions to the optical Bloch equations, to predict them. Quantum jumps were observed nearly simultaneously by three different experimental groups, all with single, isolated ions in Paul traps. Some slightly earlier observations of excessive fluctuations in the laser-induced fluorescence of a single Hg^+ ion by a group at the National Institute of Standards and Technology, viewed in retrospect, were due to quantum jumps. Similarly, sudden changes in the resonance fluorescence of trapped Ba^+ ions observed by a group at the University of Hamburg were due to quantum jumps, although this was not understood at first. This shows how discoveries can be missed if unanticipated observations are ignored rather than investigated. A fourth experiment, performed not with a single, trapped ion, but with neutral atoms transiently observed in an atomic beam, and published at about the same time as the other experiments, has been almost totally neglected.

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1. Introduction

The Nobel Prize in Physics is awarded for an 'important discovery or invention.' In this context, one would say that J.J. Thomson 'invented' a mass spectrometer, with which he 'discovered' two isotopes of neon [1]. Much other scientific work would fall into a third category called 'measurements,' such as the determination of a ratio of atomic masses to an additional decimal place. For an observation to be called a 'discovery,' it should concern a phenomenon that was unexpected or about which there was some doubt regarding its existence.

The observation of 'macroscopic quantum jumps' in single atoms could be classified as a discovery, as there was controversy among theorists as to whether they would occur. Thus, it might be considered one of the first discoveries made with a three-dimensional rf (Paul) trap. Although a Paul trap can be used as a mass spectrometer, its role in this case was simply to confine a single atomic ion to a small region of space. The experimental and theoretical work related to this phenomenon involved the efforts of three future Physics Nobel Prize laureates: Dehmelt (1989), Cohen-Tannoudji (1997), and Wineland (2012).

2. Dehmelt's proposal for 'shelved-electron detection'

The seed of the idea that resulted in the experimental and theoretical work on 'macroscopic quantum jumps' was a proposal by Dehmelt [2] for a sensitive optical double-resonance detection method called 'shelved-electron detection.' This was based on an intuitive approach to the quantum dynamics, according to which an atom was considered to be always in a particular atomic level at any given time. This method of detection was proposed in the context of developing atomic frequency standards and clocks based on narrow optical resonances in single atoms. An extremely sensitive detection method would be required to efficiently detect transitions in a single atom.

Consider the simplified atomic energy-level diagram of Fig. 1. Level 1 is the ground state. Level 3 is an excited state with a short lifetime (high spontaneous decay rate). Level 2 is a long-lived metastable state. Suppose the atom is initially in the ground state. A laser resonant with the $1 \rightarrow 2$ transition is directed at the atom for some period. The experimenter wants to know if the laser has driven the atom to Level 2. Detecting absorption by the atom is not feasible, since at most one photon would be removed from the laser beam. Detecting fluorescence is not feasible either, since at most

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http://dx.doi.org/10.1016/j.ijms.2014.07.005
1387-3806/Published by Elsevier B.V.

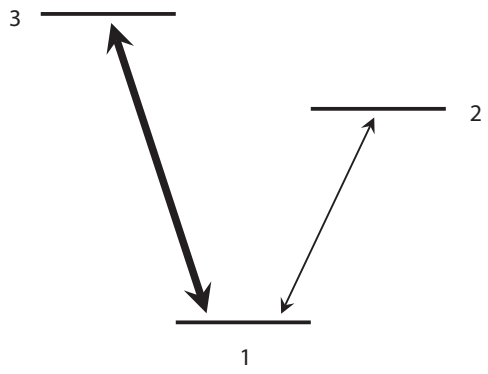

Fig. 1. Energy-level diagram of a three-level atom suitable for 'shelved-electron detection' and observation of 'macroscopic quantum jumps.' The $1 \leftrightarrow 3$ transition is 'strong,' while the $1 \leftrightarrow 2$ transition is 'weak.'

one photon would be emitted in the decay of Level 2. Instead, after attempting to drive the atom to Level 2, a laser resonant with the $1 \rightarrow 3$ transition is directed at the atom, and fluorescence photons are detected.

If the $1 \rightarrow 2$ transition was *not* driven, then the atom will be driven from Level 1 to Level 3 by the laser resonant with that transition and will then decay back to Level 1 with the emission of a photon. This process will repeat itself as long as the laser is applied. If the laser beam is intense enough to saturate the transition, the photon emission rate can be as high as one half the spontaneous decay rate, or around $10^8\ s^{-1}$ for a typical allowed transition.

If the $1 \rightarrow 2$ transition *was* driven, then the atom remains in Level 2, on average, for the natural lifetime of that state. The atom is then 'shelved' in the metastable state and can no longer be driven to Level 3 by the laser resonant with the $1 \rightarrow 3$ transition. The transition of the atom from Level 1 to Level 2 is then detected by the *absence* of many fluorescence photons.

In this example, the lasers are applied sequentially, not at the same time. This is necessary for the purpose of obtaining a narrow resonance profile on the $1 \rightarrow 2$ transition, since the laser resonant with the $1 \rightarrow 3$ transition would broaden the $1 \rightarrow 2$ transition. Cook and Kimble [3] examined the case in which both lasers are applied simultaneously. They concluded that the fluorescence would have the form of a random telegraph signal – 'on' when the atom was cycling between Levels 1 and 3, and 'off' when it was in Level 2. The transitions between the 'on' state and the 'off' state, called 'quantum jumps' would take place at random times. The transitions came to be called 'macroscopic quantum jumps' because the 'on' and 'off' states are distinguishable with a photodetector or, in favorable cases, by eye, through a microscope.

3. Theoretical doubts and controversies

The theoretical approach used by Cook and Kimble [3] was criticized by some quantum-optics theorists. Perhaps the main reason was that there was a general lack of experience in dealing with experiments involving single atoms, repeatedly observed, as opposed to ensembles of atoms, observed simultaneously. Properties of ensembles of atoms could often be understood in terms of solutions of the optical Bloch equations (the equations of motion for the elements of the atomic density matrix). The solutions of the optical Bloch equations were continuous in time, without quantum jumps.

In recent decades, it has become rare for there to be much doubt as to the outcome of a quantum-optics experiment. For this reason, the case of 'macroscopic quantum jumps' ought to be of some interest to historians of science. The period of maximal controversy was roughly from March 1985, when the paper of Cook and Kimble [3] was published, until the conclusion of the NORDITA (Nordic Institute for Theoretical Physics) Lecture Course on 'Quantum Fields and Laser Spectroscopy,' in Copenhagen in November 1985 [4,5]. During this period there was no clear experimental evidence to settle the question.

According to Claude Cohen-Tannoudji (personal communication to WMI, 2014):

> "There was indeed in the 1980s a strong doubt about the existence of quantum jumps. I remember a meeting in Copenhagen organized by Stig Stenholm around 1985. There was a long discussion about the existence of quantum jumps. Stig asked people to vote. About half of the people were claiming that these jumps could not exist! Jean Dalibard was at this meeting and we started immediately during the meeting to do the calculation of the delay function (or waiting time distribution) giving the distribution of the time intervals between 2 *successive* spontaneous emissions of a single 3-level atom. This was showing clearly that periods of darkness were appearing in the fluorescence signal. We even presented these calculations during the meeting and published them about one year after in Europhysics Letters [6]. Later on, we showed that this was even clearer in the picture of the radiative cascade of the dressed atom [7]. At that time, many people were thinking only in terms of optical Bloch equations and density matrices, giving average values of experiments performed on a large number of atoms. They were not used to calculations dealing with a single atom."

4. The NIST observations of quantum jumps

The experimental program of the Boulder Ion Storage group at the National Institute of Standards and Technology (NIST) developed along the lines originally outlined by Dehmelt, with the goal of demonstrating a frequency standard based on an optical transition in a single, trapped atomic ion. Demonstrating the existence of quantum jumps in systems not useful for frequency standards was not considered. The atom chosen for a demonstration of a single-ion frequency standard was Hg^+. The relevant levels are shown in Fig. 2. This system is of the same form as the three-level atom of Fig. 1, where the $^2S_{1/2}$, $^2D_{5/2}$, and $^2P_{1/2}$ states correspond to Levels 1, 2, and 3 respectively. The $^2P_{1/2}$ state of Hg^+ has a lifetime of 2.3 ns [8], while the $^2D_{5/2}$ has a lifetime of 86 ms [9]. The demonstration of a frequency standard based on the narrow 282 nm $^2S_{1/2} \rightarrow {}^2D_{5/2}$ transition required lasers resonant with the 194 nm

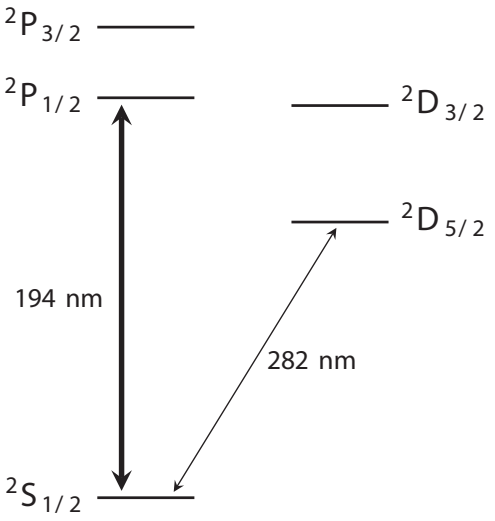

Fig. 2. Energy-level diagram of the Hg^+ ion, showing the transitions relevant to the two-laser demonstration of quantum jumps.

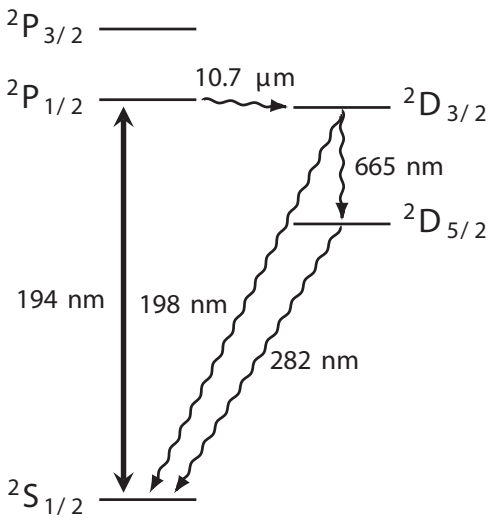

Fig. 3. Energy-level diagram of the Hg^+ ion and radiative decays relevant to the NIST experiment with only the 194 nm laser applied.

and 282 nm transitions. If the ion were irradiated with both lasers at the same time, one would expect to observe intermittent 194 nm fluorescence as the ion was driven to the $^2D_{5/2}$ state and either spontaneously decayed to, or was driven (by the 282 nm laser) to, the $^2S_{1/2}$ state.

Initially the NIST group ignored the possibility that there could be intermittent fluorescence, due to quantum jumps, with only the 194 nm laser applied. The relevant levels and transitions are shown in Fig. 3. The 194 nm $^2S_{1/2} \leftrightarrow {}^2P_{1/2}$ transition is not a perfectly cycling transition. About once in 10^7 times, instead of decaying directly to the $^2S_{1/2}$ state, an ion in the $^2P_{1/2}$ state decays to the $^2D_{3/2}$ state, which is metastable, with a lifetime of 9.2 ms [9]. The $^2D_{3/2}$ state decays to the ground state, either directly or by first decaying to the $^2D_{5/2}$ state, which is also metastable. While the ion is in one of the metastable states, the 194 nm fluorescence 'shuts off' for up to about 100 ms. The experimental signature of these quantum jumps is that they become more frequent as the 194 nm laser is tuned closer to resonance. The more frequently the ion is driven to the $^2P_{1/2}$ state, the more frequently it decays to the $^2D_{3/2}$ state.

In retrospect, these quantum jumps were probably visible in the first clear single-ion fluorescence curves observed by the NIST group in August, 1985. The ion fluorescence exhibited large fluctuations as the 194 laser was tuned close to resonance. At this time, the 282 nm laser was not available, so quantum jumps were not anticipated. Fig. 4 is a resonance curve in which the laser frequency was increased from below resonance. Figs. 4–7 are photographs of the original thermal-printer data. Due to the limited resolution of the printer, the data points are not all separated on the horizontal axis. The integration period was about 230 ms, so the dark, 'off' periods were mostly shorter than the counter integration period. Still, there were large fluctuations close to the peak of the resonance. The dots within the ellipse marked 'A' represent periods that were not much affected by 'off' periods. The dots within the ellipse marked 'B' were the most affected. This resonance curve was taken on 19 August 1985.

Fig. 5 is a plot of fluorescence versus time, taken with a shorter integration time of about 77 ms. The 194 nm laser was tuned close to resonance. The plot is from 3 March 1986. The detection efficiency was improved compared to the measurements of the previous August. The 'off' periods due to quantum jumps were still partly averaged over, but very large fluctuations are present.

Fig. 6 is a plot of fluorescence versus time taken immediately before the plot of Fig. 5, with the 194 nm laser tuned further away from resonance, on the low-frequency side. The vertical scale is the same. The average fluorescence level is less, since the laser is further from resonance. The number of low-fluorescence measurements is significantly less. This dependence of the number of low-fluorescence measurements on laser frequency is a clear signature of the single-laser type of quantum jumps involving the $^2P_{1/2} \rightarrow {}^2D_{3/2}$ spontaneous decays.

The electronic counter that was used in these experiments was not suitable for making successive measurements of short periods, because 5 ms was required to transfer data to a computer, during which time the counter was inactive. A counter and computer interface was designed and constructed by C.H. Manney (NIST), which had a storage buffer and enabled successive measurements to be made with only a few microseconds of deadtime. The first time this counter was used to observe the 194 nm fluorescence, clear evidence of quantum jumps, in the form of intermittent fluorescence, was observed. Fig. 7 is one of the first such observations, from 2 May 1986. The time bins are 10 ms. Collisions were eliminated as a cause of the intermittent fluorescence by varying the background pressure in the Paul trap. It was then realized that the quantum jumps were caused by the $^2P_{1/2} \rightarrow {}^2D_{3/2}$ spontaneous decays. A short time later, the 282 nm laser was introduced, and quantum jumps in the three-level system of Fig. 2, the original

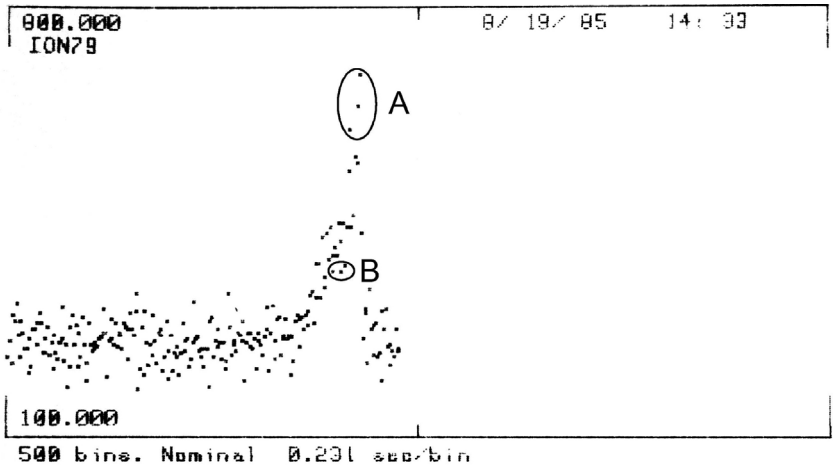

Fig. 4. Fluorescence detected from the 194 nm transition of an Hg^+ ion. The 194 nm laser frequency increases from left to right. Dots within the ellipses marked A and B represent periods when the ion spent the least or most time, respectively, in a metastable state. The vertical scale is in frequency units (counts per second). The number of photons counted is given by the number of counts per second multiplied by the time bin (0.231 s). This number can be used to estimate the statistical noise of photodetection. The vertical scale limits are 2.31 and 184.8 counts per bin. The background can be estimated from the low-laser-frequency data.

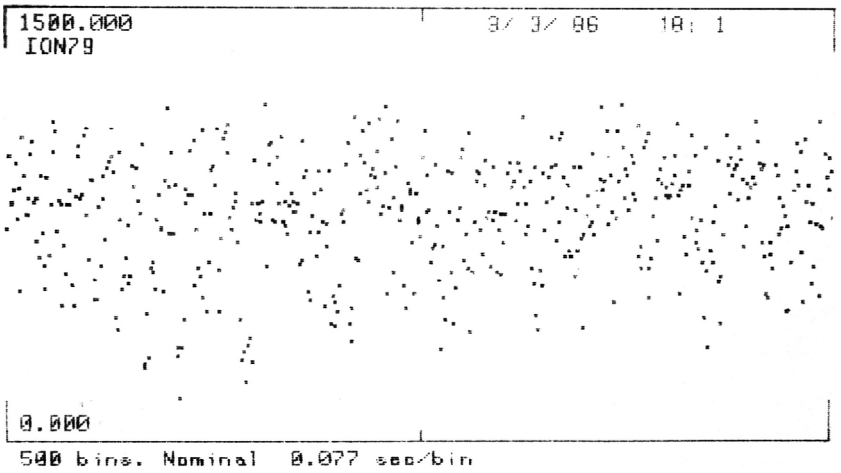

Fig. 5. Sequence of 194 nm Hg^+ fluorescence measurements of duration 77 ms. Vertical scale limits are 0 and 11.55 counts per bin. Frequency of laser is fixed and close to resonance. Measurement time increases from left to right. The lowest-fluorescence measurements represent periods when the ion spent much of the time in a metastable state. No background has been subtracted from the data.

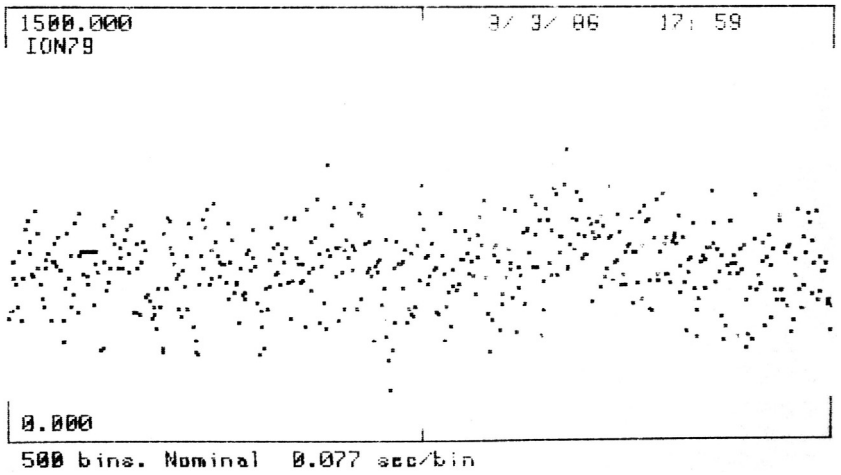

Fig. 6. Sequence of 194 nm Hg^+ fluorescence photocounts of duration 77 ms. Frequency of laser is fixed and detuned further below resonance, compared to Fig. 5. The horizontal and vertical scales are the same as in Fig. 5.

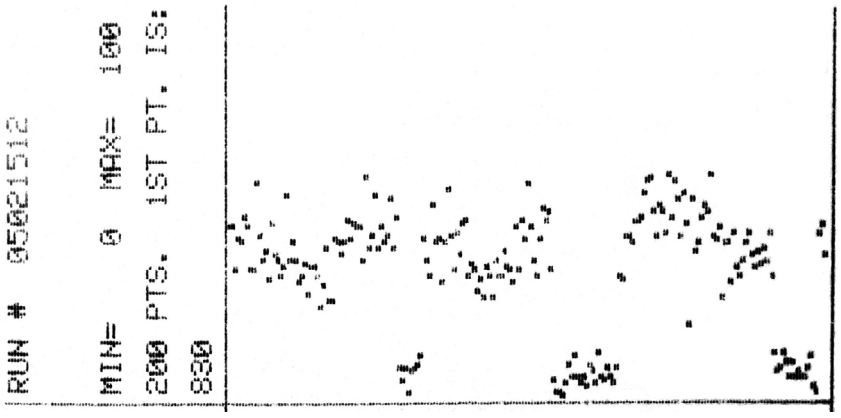

Fig. 7. Time sequence of 194 nm fluorescence measured with a low-deadtime counter. This is one of the first clear observations of quantum jumps in Hg^+, 2 May 1986. Time bins are 10 ms. Hence, the vertical scale limits correspond to 0 and 10^4 counts per second or 0-100 counts per bin. No background has been subtracted from the data.

goal of the experiment, were observed. The article reporting observations of quantum jumps for the two-laser, three-level system [10] was submitted on 23 June 1986 and published on 6 October 1986.

The existence of the weakly allowed $^2P_{1/2} \rightarrow {}^2D_{3/2}$ decay in Hg^+, leading to quantum jumps with only a single laser, had been anticipated in an obscure 1981 conference proceedings by the same group [11], but forgotten by them by 1985. In the context of those conference proceedings, the decay had been regarded as an extraneous effect, resulting in a tolerable loss of signal. In 1985, the NIST group was so focused on demonstrating the quantum jumps in the two-laser system that the evidence for the single-laser

quantum jumps was ignored at first. The details of the calculation of the $^2P_{1/2} \rightarrow {}^2D_{3/2}$ decay rate have never been published, and are reproduced in Appendix A.

The statistics of the quantum jumps observed with just the 194 nm laser were later analyzed to obtain the decay rates for the $^2D_{3/2}$ and $^2D_{5/2}$ states [9]. An anonymous reviewer of Ref. [9] was not overly impressed:

> "While the authors do not say so, this paper essentially is a worthwhile quantitative study of a small wart on the pretty face of the shelved-electron amplifier scheme. . . .While this may not be a great discovery, the work is, in my opinion, up to the average level of papers in PRL. I recommend publication."

5. Other single, trapped ion experiments

Two other experimental groups were working at the same time as the NIST group to demonstrate the existence of the macroscopic quantum jumps in single-ion fluorescence (Dehmelt's group at the University of Washington and Toschek's group at the University of Hamburg). The two experiments were similar to each other in that they both used single Ba^+ ions confined in Paul traps. The experiments were designed primarily to demonstrate the existence of the quantum jumps and only secondarily as part of an atomic frequency standard program, which would have required at least one more laser.

5.1. University of Washington group

Laser cooling and fluorescence detection of a Ba^+ ion requires at least two lasers, one at 494 nm, to excite the ion from the ground $6s^2S_{1/2}$ state to the $6p^2P_{1/2}$ state, and another one at 650 nm to excite the ion out of the $5d^2D_{3/2}$ state, to which the $6p^2P_{1/2}$ state can decay, where it would otherwise be trapped. The atomic levels and transitions are shown in Fig. 8. Both the $5d^2D_{3/2}$ and $5d^2D_{5/2}$ states are metastable. If lasers resonant with the $6s^2S_{1/2} \rightarrow 6p^2P_{1/2}$ and the $5d^2D_{3/2} \rightarrow 6p^2P_{1/2}$ transitions are applied, the ion cycles between the three states and emits photons at 494 nm and 650 nm.

If the ion is somehow transferred to the $5d^2D_{5/2}$ state, the laser-induced fluorescence at both 494 nm and 650 nm ceases. One way to do this is with a 1.76 μm laser resonant with the $6s^2S_{1/2} \rightarrow 5d^2D_{5/2}$ transition. This would be analogous to the three-level system shown in Fig. 1. The method actually used by the University of Washington group was to focus light from a barium hollow-cathode lamp, passed through an interference filter centered around the 456 nm $6s^2S_{1/2} \rightarrow 6p^2P_{3/2}$ transition. If light from the lamp excited the ion to the $6p^2P_{3/2}$ state it would

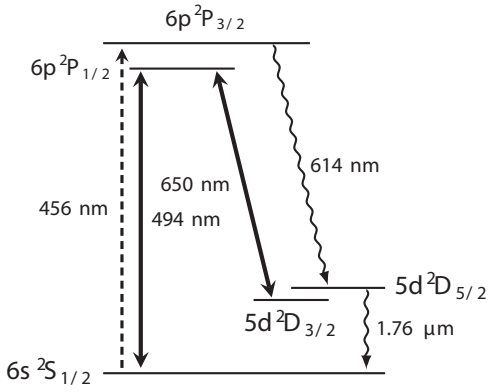

Fig. 8. Energy-level diagram of Ba^+, showing the levels and transitions involved in the experiments of the University of Washington and University of Hamburg groups.

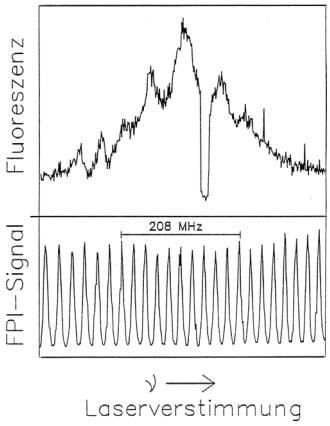

Fig. 9. Early data (1984–1985) from the University of Hamburg group [13, Fig. 28]. Resonance fluorescence of a single Ba^+ ion as a function of the frequency of the 650 nm laser (top). Transmission signal of Fabry–Pérot interferometer used for frequency calibration (bottom). The laser frequency and time increase from left to right. The sudden decrease in fluorescence followed by a sudden increase is probably due to quantum jumps to and from the $5d^2D_{5/2}$ dark state. The multiple peaks in the signal are probably due to rf micromotion of the ion. (Number of data points = 370, time per point = 1 s.)

have a fair probability of decaying to the metastable $5d^2D_{5/2}$. This would cause the fluorescence to stop. When the ion returned to the ground $6s^2S_{1/2}$ state, either by spontaneous decay or due to a collision, the fluorescence would resume. The fluorescence was observed to switch 'on' or 'off' over periods of tens of seconds. From the distribution of the 'off' periods, they estimated the natural lifetime of the $5d^2D_{5/2}$ state to be 32 ± 5 s. The University of Washington group was the first to publish experimental evidence for the existence of macroscopic quantum jumps. Their article [12] was submitted on 5 May 1986 and published on 30 June 1986.

5.2. University of Hamburg group

The University of Hamburg group carried out an experiment very similar to that of the University of Washington group, also on single, trapped Ba^+ ions. The system was even simpler in that no lamp was required to populate the metastable $5d^2D_{5/2}$ state. Only 494 nm and 650 nm lasers were present. Apparently the ion was driven from the $6s^2S_{1/2}$ state to the $5d^2D_{5/2}$ state by a far-off-resonant Raman process involving the 494 nm light. Their article [14] was submitted on 12 May 1986 and published on 6 October 1986.

Similarly to the NIST group, the University of Hamburg group made early observations that, in retrospect, were due to quantum jumps (P.E. Toschek, R. Blatt, personal communications to WMI, 2014). As early as 1984, sudden changes were observed in the level of resonance fluorescence of Ba^+ ions (Fig. 9). Some experimental plots showing these sudden changes were presented at conferences and published [15,16] before they were understood. In fact, a plot that eventually was published as evidence for quantum jumps in Ref. [14] was recorded in December 1984 (P.E. Toschek, R. Blatt, personal communications to WMI, 2014). After learning of the University of Washington group's use of a resonance lamp to populate the $5d^2D_{5/2}$ state, the University of Hamburg group also used this method to observe quantum jumps [17].

6. The neglected experiment of Finn and Greenlees

A fourth experiment, by Finn and Greenlees [18], was performed and published at about the same time as the other three experiments (submitted 23 June 1986, published 1 November 1986).

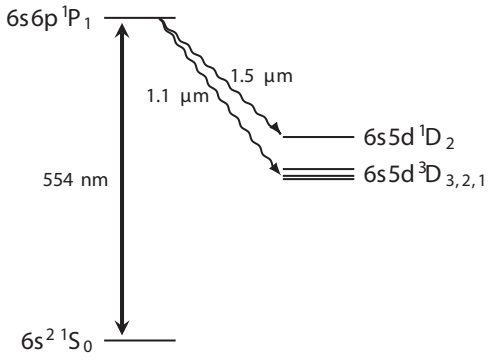

Fig. 10. Energy-level diagram of the neutral barium atom, showing the levels and transitions involved in the experiment of Finn and Greenlees.

Unlike the other experiments, it was performed with a series of neutral atoms in a dilute atomic beam, not single, trapped ions.

Fig. 10 shows the relevant levels of the Ba atom. The ground $6s^2 {}^1S_0$ state is laser-excited to the $6s6p {}^1P_1$ state, and the 554 nm photons emitted in the decay back to the ground state are detected. An atom continues to emit 554 nm photons until it decays to one of the metastable *D*-states or it leaves the region of laser excitation. The ratio of decays to the ground state to any of the *D*-states is 334 ± 30 [19]. Since decay from any of the *D*-states is electric-dipole-forbidden, an atom, once it is in a *D*-state remains there until after it leaves the experimental region.

In the experiment, the detection of a 554 nm photon indicated that a Ba atom was in the experimental region excited by the laser. The detection of another photon after a delay of a few microseconds indicated that the atom was still fluorescing and had not decayed to one of the *D*-states (provided one accepts the quantum-jump picture). Photons were also detected within short time intervals between these two detection periods. The photon detection efficiency was too low to show quantum jumps for any individual atom. The photon counts in equivalent time intervals had to be summed for many atoms passing through the apparatus in order to obtain a statistically significant signal. If the photon counts in equivalent time intervals were summed for all atoms, whether or not a photon was detected in the most-delayed period (the ensemble-averaged case), a progressive decrease in fluorescence rate during the intermediate period was observed. The solution of the optical Bloch equations would predict an exponentially decreasing fluorescence rate. The hypothesis that this solution should be applied to the case of a single atom would be consistent with the ensemble-averaged observations. However, the subset of atoms for which a second photon was detected at the most-delayed period showed an essentially constant fluorescence rate during the intermediate period. This is not consistent with the solution of the optical Bloch equations. These observations can be interpreted as confirmation that a single atom fluoresces at a constant rate until it makes a quantum jump to one of the *D*-states and then ceases to fluoresce. The results were enough to disprove the hypothesis that the fluorescence rate of a single atom should be calculated according to the optical Bloch equations. The results were consistent with the existence of quantum jumps, although they do not clearly demonstrate the existence of the dark, 'off' periods. The predictions of the optical Bloch equations are seen to apply to ensemble averages but not to single atoms.

The experiment was little noted at the time of publication and has since been almost entirely neglected. According to an online database [20], the number of citations to the experiments of the University of Washington group [12], the University of Hamburg group [14], the NIST group [10], and Finn and Greenlees [18] are 500, 374, 373, and 21, respectively. The most recent citation to Ref. [18] is from 2001. According to another online database [21], the corresponding numbers of citations are 799, 530, 598, and 23. The lack of response to Ref. [18] is probably due to the fact that the results were less clear-cut than those of the single-ion experiments [12,10,14]. Had it been submitted a year earlier, it might have had a greater impact.

7. Resolution of the theoretical doubts

As alluded to in Section 3, theoretical discussions to resolve the question of whether or not the quantum jumps could be observed were going on at the same time as the experiments. Javanainen presented arguments at the November 1985 Copenhagen meeting, based on the quantum theory of photodetection, that predicted 'on' and 'off' periods in the fluorescence of a three-level atom. These calculations were published in March 1986 [22]. The calculations of Cohen-Tannoudji and Dalibard [6], first obtained at the same conference, were published in May 1986. They calculated the delay function $w(\tau)$, which is the probability, if one has detected a photon at time t, to detect the *next* photon at time $t+\tau$, not just *any* photon. The delay function clearly showed the existence of extended dark periods. In the end, it turned out that the predictions of rigorous quantum calculations were in agreement with the intuitive picture of Dehmelt, at least as far as anything that could be observed experimentally. According to Javanainen [22]:

> "The concept of 'quantum jumps' represents a questionable extrapolation of classical reasoning into quantum mechanics. Nevertheless, in the present example it leads to a prediction which agrees with the full-fledged theory of photodetection."

By the time that the first clear experimental evidence of quantum jumps was published in June 1986 [12], probably most of the quantum optics community was already convinced of their existence. Reviews of the theory of quantum jumps were published some years after the experiments described here were carried out [23,24]. The topic is included in some textbooks on quantum optics, for example, Ref. [25, pp. 329–333].

8. Concluding remarks: theoretical blinders, experimental blinders

Some observations can be made on the existence of 'blinders,' both in theory and in experiment. Some theorists were accustomed to seeing problems in terms of the tools which had previously proven useful for describing experiments with large numbers of atoms, such as the optical Bloch equations. Their intuition seemed to fail for the case of single atoms, and they were unprepared to accept the phenomenon of macroscopic quantum jumps. The experiences of both the NIST group and the University of Hamburg group show that experimenters also can have blinders. They failed to pursue the cause of some unanticipated experimental observations (excess fluctuations of fluorescence in the former case and abrupt changes in the fluorescence in the latter). In fact they were observing the effects of quantum jumps. If, in the NIST experiment, the integration period of the photon counter had been reduced, the quantum jumps, in the form of extended dark periods, would have become obvious. The calculation (Appendix A) needed to predict the quantum jumps was available, but was neglected.

Acknowledgments

We thank C. Cohen-Tannoudji for unpublished information about the theoretical discussions related to the possibility of observing quantum jumps. We thank P. Toschek and R. Blatt for unpublished information on their experiments on trapped Ba^+ ions. We thank S. Jefferts, J. Bollinger, and two anonymous referees for helpful comments on the manuscript.

Appendix A. Calculation of the $^2P_{1/2} \rightarrow {}^2D_{3/2}$ decay rate in Hg^+

The 10.7 μm $5d^{10}6p^2P_{1/2} \rightarrow 5d^96s^22D_{3/2}$ decay in Hg^+ (Fig. 3) is forbidden to the extent that the configurations are pure. This is because two electrons change their orbitals in this transition ($6p \rightarrow 6s$ and $5d \rightarrow 6s$), and the electric dipole matrix element (actually, any electromagnetic multipole matrix element) vanishes in such a case. However, the exact wavefunctions are mixtures of configurations, and such configuration mixing could allow the decay to occur. Although the $^2P_{1/2} \rightarrow {}^2D_{3/2}$ decay had not been directly observed prior to 1985, the related decay between other fine-structure components ($^2P_{3/2} \rightarrow {}^2D_{5/2}$) of the same configurations *had* been observed, by observations of the emitted 398 nm photons. The branching ratio of two decays from the $5d^{10}6p^2P_{3/2}$ state, $(5d^{10}6p^2P_{3/2} \rightarrow 5d^{10}6s^2S_{1/2})$ / $(5d^{10}6p^2P_{3/2} \rightarrow 5d^96s^22D_{5/2})$, was measured by Crandall et al. [26] to be 350 ± 30%.

In Ref. [11, Appendix B], the assumption was made that the ratios of the decay rates ($^2P_J \rightarrow {}^2D_{J'}$) for all J and J' could be calculated according to the rules for the decays of *LS*-coupled states. This would be a good approximation, for example, if the decay was allowed through mixing of the $5d^{10}6d$ configuration into the $5d^96s^22D_{3/2,5/2}$ states. Then only one electron has to change its orbital ($6p \rightarrow 6s$). This is not the only possible form of configuration interaction that would allow such a decay, but it is computationally the simplest to deal with. The rate $\gamma(LSJ \rightarrow L'S'J')$ for a state $|(LS)J\rangle$ to decay to a state $|(L'S')J'\rangle$ of lower energy is given by

$$\gamma(LSJ \rightarrow L'S'J') = C\frac{(\Delta E)^3}{2J+1}|((LS)J||D^{(1)}||(L'S')J')|^2 \tag{A.1}$$

$$= C(\Delta E)^3(2J'+1)\begin{Bmatrix} L & J & S \\ J' & L' & 1 \end{Bmatrix}^2 |(L||D^{(1)}||L')|^2, \tag{A.2}$$

where C is a constant, ΔE is the energy difference between the two states, and $((LS)J||D^{(1)}||(L'S')J')$ is a reduced matrix element of the electric dipole operator $\mathbf{D}^{(1)}$ [27, Ch. 14].

With the approximation of Eqs. (A.1 and A.2), the branching ratio of the two decays from the $^2P_{1/2}$ state can be related to the branching ratio of decays from the $^2P_{3/2}$ state reported in Ref. [26]:

$$\frac{\gamma(^2P_{1/2} \rightarrow {}^2D_{3/2})}{\gamma(^2P_{1/2} \rightarrow {}^2S_{1/2})} = \frac{2}{3}\frac{\begin{Bmatrix} 1 & \frac{1}{2} & \frac{1}{2} \\ \frac{3}{2} & 2 & 1 \end{Bmatrix}^2}{\begin{Bmatrix} 1 & \frac{1}{2} & \frac{1}{2} \\ \frac{1}{2} & 0 & 1 \end{Bmatrix}^2}\frac{\begin{Bmatrix} 1 & \frac{3}{2} & \frac{1}{2} \\ \frac{1}{2} & 0 & 1 \end{Bmatrix}^2}{\begin{Bmatrix} 1 & \frac{3}{2} & \frac{1}{2} \\ \frac{5}{2} & 2 & 1 \end{Bmatrix}^2} \times \left[\frac{E(^2P_{1/2}) - E(^2D_{3/2})}{E(^2P_{1/2}) - E(^2S_{1/2})}\right]^3 \times \left[\frac{E(^2P_{3/2}) - E(^2S_{1/2})}{E(^2P_{3/2}) - E(^2D_{5/2})}\right]^3 \times \frac{\gamma(^2P_{3/2} \rightarrow {}^2D_{5/2})}{\gamma(^2P_{3/2} \rightarrow {}^2S_{1/2})} \tag{A.3}$$

$$= \frac{10}{9}\left(\frac{933}{51485}\right)^3\left(\frac{60608}{25094}\right)^3\frac{1}{350} \tag{A.4}$$

$$= 2.66 \times 10^{-7} \tag{A.5}$$

This value, rounded to 3×10^{-7}, was published in Ref. [11]. The radiative lifetime of the $6p\ ^2P_{1/2}$ state has been measured to be 2.91 ± 0.11 ns [8]. The $6p^2P_{1/2}$ lifetime, together with the calculated value given by Eq. (A.5), yields $\gamma(^2P_{1/2} \rightarrow {}^2D_{3/2}) = 91.5 \pm 27\,s^{-1}$. This agrees with the experimental value of $52 \pm 16\,s^{-1}$ [9], within the combined uncertainties. An unpublished calculation, based on R.D. Cowan's atomic structure codes [27] and referred to in Ref. [9], predicted $\gamma(^2P_{1/2} \rightarrow {}^2D_{3/2}) = 55\,s^{-1}$ (D. Al-Salameh, W. Silfvast, personal communication to WMI, 1987). Another calculation, also based on Cowan's codes, predicted $\gamma(^2P_{1/2} \rightarrow {}^2D_{3/2}) = 57.7\,s^{-1}$ [28]. Both Cowan-code calculations are in good agreement with the experimental value.

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