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 → 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.

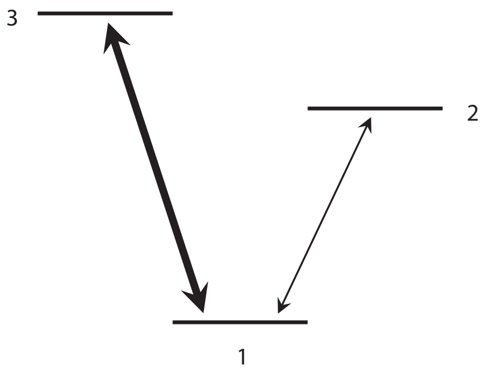

Fig. 1. Energy-level diagram of a three-level atom suitable for 'shelved-electron detection' and observation of 'macroscopic quantum jumps.' The 1 ↔ 3 transition is 'strong,' while the 1 ↔ 2 transition is 'weak.'

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 → 3 transition is directed at the atom, and fluorescence photons are detected.

If the 1 → 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 $10^8\ \mathrm{s}^{-1}$ for a typical allowed transition.

If the 1 → 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 → 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 → 2 transition, since the laser resonant with the 1 → 3 transition would broaden the 1 → 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

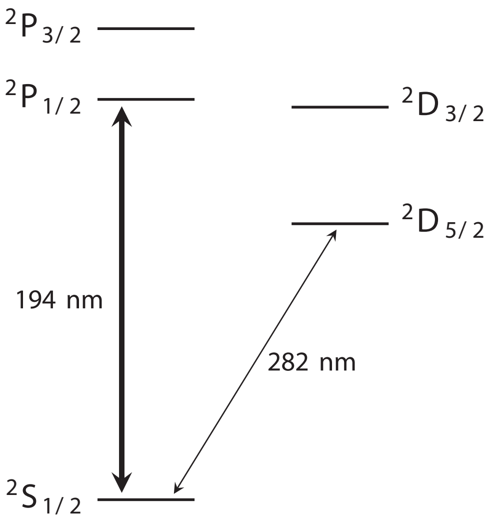

Fig. 2. Energy-level diagram of the Hg^+ ion, showing the transitions relevant to the two-laser demonstration of quantum jumps.