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On the Incorrectness of the Claim about the Observation of Bohr Quantum Jumps in Experiments with Single Atoms

© E.G. Saprykin, V.A. Sorokin

Institute of Automation and Electrometry, Siberian BranchRussian Academy of Sciences,
Novosibirsk, Russia

e-mail: Saprykin@iae.nsk.su, vladsorokin1957@yandex.ru

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As a result of a retrospective analysis of works reporting the observation of quantum jumps in single ions from 1986 to 2015, the incorrectness of the claim about the detection of Bohr quantum jumps has been demonstrated, and the presumed cause of the misconception has been identified. The authors of the analyzed works mistook a nonlinear interference effect, which manifested in their experiment and accompanies the combinational scattering of radiation, predicted by Russians 10 years before their work, for Bohr jumps.

Keywords: barium ions, single atoms, quantum jumps, nonlinear interference effect.

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1. Introduction

In 1986, three groups of experimenters published articles containing the words — „observation" of quantum jumps [1–3]. From articles [1,3], the nature of the quantum jumps could be understood only by reading their text. Unlike them, in the abstract of work [2], the main result was claimed to be the observation of Bohr quantum jumps by the authors, illustrated in the text [2] by a graph of frequency scanning, absent in works [1–3].

Quantum jumps, introduced by Bohr in 1913, were one of the most significant consequences of his quantum mechanics [4], which proceeds from the postulate of the impossibility of an atom being in states that do not correspond to the energy of stationary states. Therefore, the transition between them cannot be gradual but must occur as a quantum jump, during which the energy of the intermediate state is undefined, as is the moment of the transition time. Over time and with the establishment of the new quantum mechanics, the attitude toward them changed, and at present, in quantum physics, they are considered as a sometimes useful heuristic device. Modern views of quantum physics on this issue require consideration beyond the scope of this article. Therefore, we will present the views on Bohr jumps that had formed in quantum physics by the time works [1–3] were carried out.

Already initially, the classics had no answer to the question: why, during a Bohr jump, is there no continuous spectrum emitted? And further, the ambiguous attitude toward them persisted. Thus, the authors [2] write about Bohr's quantum jumps as follows: „they were mainly considered at that time and even later as a peculiar artifact of Bohr's atomic model, whose real existence was doubtful and, in any case, not subject to verification on ordinary large atomic ensembles". The same authors noted in work [5]: „only in large atomic ensembles, with which ordinary experiments deal, was a continuous change in atomic variables in time assumed to occur. Although this consequence, as if slipping from the imagination, led to severe doubts, the stunning successes of Bohr's quantum mechanics soon calmed the skeptics".

It should be noted that Schrödinger belonged to their number; as a result of a discussion with Bohr, he was forced to accept his point of view, and Bohr jumps, also called Bohr-Schrödinger jumps, were included in wave mechanics. But experimentally, the question of the jump-like or gradual nature of optical transitions remained unresolved. Participants in the celebration of the fifth anniversary of the new quantum mechanics noted that it took place in an atmosphere of anticipation of an experimental demonstration of the gradual nature of optical transitions predicted by matrix mechanics. But the expectation of such experiments shifted to the next century, and the clarification of this issue continued on a theoretical plane.

Both of these situations were considered in 1976 in the first monograph on the dynamics of spectroscopic transitions ([6], section 4.4). It notes that in the Schródinger picture of the transition, only the eigenstates of the energy operator are real; intermediate states are forbidden and cannot be observed. Therefore, the calculated superposition states of energy have only a formal meaning, consisting in the fact that they are solutions to the Schrödinger equation identifying the initial and final states of the transition, and quantum jumps of this type were proposed by the author to be called Schrödinger jumps. In the alternative model of gradual transitions, the author [6] (see p. 125 of the Russian translation) proceeds from the assumption that the superposition state is the true state of the system. More precisely, during the transition from state 1 to state 2, the system passes through a continuum of superposition states that are just as „real“ as states 1 and 2. But only under the condition that their eigenvalues are not eigenvalues of the energy operator.[1] Therefore, interference in the process of such a gradual transition, for example, with attempts to measure energy, disturbs the system and leads to a jump-like transition of the system to one of the stationary energy states. Such jumps were proposed by him to be called Heisenberg jumps. In conclusion of section 4.4, the author designated his position regarding the nature of quantum jumps. In his opinion, there are no jump-like transitions as Schrödinger understood them, but there are transitions extended in time and Heisenberg quantum jumps interrupting them, forced by the „incorrect“ type of measurement.

The authors of article [7] in its review part note that „„...with the development of modern quantum theory, the Bohr concept of the quantum jump was relegated to a much more problematic status. As is known, the new theory does not describe discontinuous atomic transitions (jumps) but rather speaks only of the continuity of the evolution of the wave function“. In this case, the authors undoubtedly meant Schrödinger's wave mechanics. But in Heisenberg's matrix mechanics, the situation is similar. Here too, the continuity of optical transitions is assumed, but there is also the possibility of interrupting the transition by external perturbation.

What situation is actually realized had to be shown by experiments. The first experiment in which, 73 years after Bohr, the observation of Bohr quantum jumps was claimed was precisely work [2]. In it, as well as in works [1,3], the reduction of the ensemble of atoms involved in the experiments (to $1-3$ barium ions) was ensured by low ion concentration, trapping them in ion traps, and additional laser cooling.

2. Macroscopic Quantum Jumps in Single-Atom Spectroscopy

The extraordinary nature of the results of work [2] regarding the registration of Bohr jumps dictates the necessity to consider an alternative mechanism for generating quantum jumps within the concept of [8]. The initial prerequisite for carrying out works to implement such a mechanism was the V-scheme of double resonance as an amplification mechanism for detecting weak optical transitions in single-atom spectroscopy of excited ions, which was presented by Dehmelt in a 1975 report and more detailedly substantiated in the 1981 monograph. To implement the proposed mechanism, it was necessary to use laser radiation controlling the ion's stay in the ground state $6s^2S_{1/2}$. Laser radiation saturating the strong or weak transitions, acting separately, stimulated either intense photon scattering (fluorescence) on the strong transition $6p^2P_{1/2}$ or negligibly small fluorescence on the weak

[1] The author [6] does not provide examples of such operators, but one of them is the momentum operator.

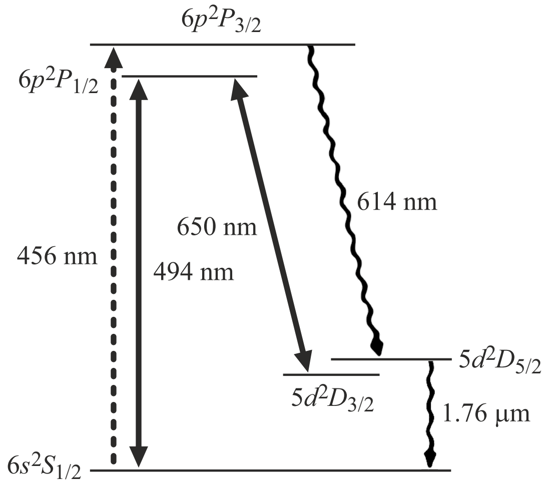

Figure 1. Scheme of levels involved in the experiments of works [2,5].

transition from the metastable state $5d^2D_{5/2}$ to the ground state. However, if, with the strong transition excited, at least one photon is absorbed from the field acting on the weak transition from the metastable state to the ground state, then intense fluorescence on the strong transition will be interrupted, and small fluorescence corresponding to the weak transition will appear. After some time, the reverse situation will occur, and intense fluorescence on the strong transition will resume. Such interruptions (jumps) of intense fluorescence reveal the presence of weak transitions. With continuous excitation of both transitions, a regime arises called „telegraph“, with a random distribution in time and variability of the widths of „dark“ dips associated with the duration of fluorescence of the metastable state. The observation of such jumps was reported in works [1–3].

The scheme of levels Ba^+ used in works [1,2,5] and the fields exciting them is shown in Fig. 1. The only source of excitation of levels Ba^+ in [w]as „green“ laser radiation with a wavelength of 494,nm, directly populating the $6p^2P_{1/2}$ level, followed by spontaneous decay to metastable levels $5d^2D_{3/2}$ and $5d^2D_{5/2}$. In the priority work [1], they were additionally populated from the $6p^2P_{3/2}$level, excited by a barium lamp with a wavelength of 456 nm. After the publication of work [1], this method was also used in [5]. In addition, radiation from a „red“ laser with a wavelength of 650 nm acted on the transition $6p^2P_{1/2}-5d_2D^{3/2}$, which in the normal mode, detuned 300,MHz downward from the transition center, contributed to additional optical cooling of the ions and registration of the „quantum telegraph“ mode.

In the [2] „red“ laser was frequency scanned with the aim of obtaining spectral dependencies of the green fluorescence. At the same time, the „green“ laser was detuned by -300 MHz from the line center to provide stable optical cooling. This experiment showed that stable optical cooling persists as long as the detuning of the scanning laser frequency remains negative. Upon shifting into the anti-Stokes wing of the combination scattering (CS) line, the detuning sign changes, cooling turns into heating, and the spectral shape changes. This is specific to the laser cooling system used. An example of such scanning, interpreted in [2] as evidence of Bohr jumps, is shown in Fig. 2 of [2].[2] The specific form of the graphs depends on laser intensities. They are not presented in the paper. However, the wide optogalvanic signal track shown in Fig. 2 reveals significant field effects. The full width at half maximum of this contour is 700 MHz, determined by the intensity of the 494 nm laser. The intensity of the 650 nm laser also contributes to the intensity of the green fluorescence, showing dips — effects of population transfer from the $6p^2P_{1/2}$ level to the less populated $5d^2D^{3/2}$ level.

Downward jumps corresponding to transitions to the $5d^2D_{5/2}$ level were interpreted by the authors of [2] as the revival of Bohr jumps, made possible by the shift from experiments with atom ensembles to experiments with single atoms. The incomplete description of the scheme parameters and the acting fields, as well as the absence of other spectral dependencies (except Fig. 2 in [2]) that display the influence of parameters on spectral shapes, preclude identification of the true cause of these jumps. However, in the years preceding these experiments and the publication of [2], physicists involved in advancing the [8] theme had access to such information (via personal contacts, conferences, and other sources) and could form their own perspective on the matter.

Since 1975, many physicists began to implement Dehmelt's idea of a new method to detect weak transitions. Some are mentioned in Dehmelt's review [8], covering works from 1956−1981 others in the review [11], spanning the 1985−1986. A broader list is given in [7] (1995), with further references in the retrospective 2015 review [9], authored by experimental participants. A team of qualified physicists united to advance the topic.[3] Each spent several years realizing aspects of the [8] concept; hence, their opinion on the reliability or error of the claims by [2] regarding the observation of Bohr jumps is important.

However, attempts to ascertain how subsequent authors treated the claim by [2] regarding Bohr jumps in the spectral dependence of Fig. 2 showed the result from [2] was largely ignored by colleagues, with no citations referencing it. Citations only exist towards results in [2] that align with the [8] concept.

For example, it is omitted in [12], which only cites the experiments [1,3], or cited among a list of works observing quantum jumps in V- or Λ-cascade configurations [13], or as works consistent with the theory of suppression of quantum jumps by frequent measurements (Zeno effect) [14], or as demonstrations of the „telegraph" mode [15,16].

[2] Only much later did the authors of [9], preparing their publication, learn from the authors of [2] that this result was taken from a series of experiments from 1984−1985 which had previously been misunderstood. From [9], it follows that among the participants of the works, the observed „quantum jumps" became more widely known as„macroscopic quantum jumps", introduced in [10].

[3] Three of them later became Nobel laureates in Physics: H. Dehmelt (1989), K. Cohen-Tannoudji (1997), and D. Wineland (2012) [9].

The absence of critical references to the Bohr interpretation of jumps in [2] likely stems from the fact that papers [1–3] submitted for publication were discussed at the IQEC-86 conference in San Francisco before their June 1986 publication.[4] In [1,3], the effects were explained without invoking Bohr jumps, resolving the question at the conference, the proceedings of which have never been published. Therefore, in their subsequent article [5] submitted in August 1986, the authors of [2] no longer mention Bohr jump registration, interpreting the experiments [1–3] only as the quenching of resonance fluorescence consistent with ideas from 1980−1985 unrelated to the Bohr postulate. No corrections to [2] were published, possibly because its print [2] release was delayed to October. Consequently, many Russian physicists distant from the [1–3,8] topic still accept this artifact at face value and, based on [2], believe atomic transitions occur as jumps. Therefore, we demonstrate the illegitimacy of applying Bohr's quantum jump concept to interpret the [2] single-atom results.

Adding to the above experiments, three further studies published between 1986−1989 in other journals employed not only V-schemes with ion traps ([18] $^{24}Mg^{+}$) including a Mg cascade), but also Λ-cascade schemes with atomic beams [17,19] $^{138}Ba^{+}$). The results were interesting but aligned with the [8] concept.

Among post-1986 works, [7] stands out as a review, due to its many commented references on [8]. Particularly notable is that the article's title question„Do quantum jumps occur at certain times?" is linked by its authors [7] to the presence or absence of Bohr quantum jumps.

The authors of [7], highlighting [1], note that other single-atom experiments were often cited as proof of something suspiciously akin to Bohr jumps, and concerns about reviving Bohr's concept persisted. The article presents arguments and references alternative hypotheses. In their opinion, the concern was finally dispelled by [12,14,20], showing how the principle of atomic state superposition, alien to Bohr's old theory, aligns with the „quantum telegraph".

In their article, [7] also conclude that single-atom experiments do not prove quantum jumps in the old sense. They use a thought interference experiment to demonstrate this. However, their argument relies on Schrödinger's famous 1952 work [21], based on gradual optical transitions and recognizing interfering waves as the only reality.

3. Bohr Quantum Jumps in Single-Atom Spectroscopy as a Result of an Error in Experimental Data Interpretation

The final review article on 1980s experiments was published in 2015 [9]. Unlike [7], its authors were active participants in the experimental justification and implementation of the [8] concept during those years. Thanks to their historical overview in 2014, the true cause of the

[4] See the bibliography in [5], submitted in August before the publication of [2].

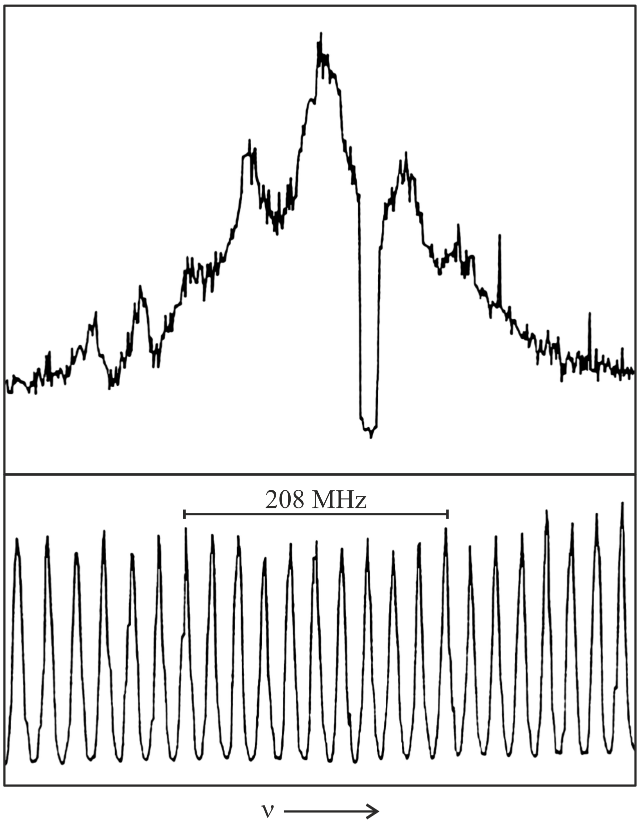

Figure 2. Early data (1984–1985) from the Hamburg University group ([22], Fig. 28). The laser frequency at 650 nm and time increase from left to right. Number of data points: 370, time per point: 1 s.

misconception by the authors of [2], who claimed Bohr jump registration, was uncovered.

One objective of [9] was to show„…how discoveries can be missed if unexpected, mysterious observations are ignored rather than investigated". Section 5.2 of that review considers one such case in the Hamburg research group [2,5]. Contacts between the authors of [9] and [2,5] during the preparation of the review revealed that the latter had registered quantum jumps prior to the works [1].[5] They cite their group's data in a dissertation fragment relating to 1984–1985 (Fig. 28 in [22]). Contacts between group members also revealed that Fig. 28 (from [22]) and Fig. 2 (from [2]) originate from the same experiment series but differ noticeably.

Both depict resonance fluorescence graphs of Ba^+ ions at 494 nm, frequency-shifted -300 MHz below resonance center, against 650 nm laser frequency. However, in the experiment of Fig. 28 ([22])[6], only a single ion was involved, and the experiment was conducted under significantly lower laser field intensities. In contrast, Fig. 2 ([2]) shows the optogalvanic signal width, broadened by strong fields, reaching about 700 MHz at half maximum, as noted above. Exactly this circumstance allowed the authors to identify sharp structures caused by variations in the number of ions correlated in the fluorescence process. In [2] they write that the structure 300 MHz below the line center corresponds to a two-photon Stokes resonance $^3S_{1/2}–^2D_{3/2}$ (the left wing of the graph), and the sign of downward steps corresponds to transitions to level $^2D_{5/2}$. However, excessive intensity of laser fields exciting fluorescence prevents describing the experiment results within simple models of combination scattering (CS).

At the same time, from the graph of [22], reproduced here as Fig. 2, the width of the field background is about 200 MHz. This indicates a low intensity of the laser at $\lambda = 494$ nm insufficient to cause field broadening of the line or generate nonlinear structures around -300 MHz. The presence of nonlinear CS structures separated by about 70 MHz near the center of the 494 nm fluorescence line signals that the intensity of the $\lambda = 650$ nm scanning laser is enough to generate these structures.

Surprisingly, the signs of the Stokes and anti-Stokes wings of CS changed compared to when an intense 494 nm laser was used. Indeed, starting the scan, the detuning of the scanning laser frequency initiating CS from the line center is maximal (left wing) and decreases to zero at the line center. This corresponds to the anti-Stokes wing, while in [2] this frequency tuning region corresponded to the Stokes wing. After passing the line maximum, the detuning changes sign (right wing) and increases, remaining below the scanning laser detuning, corresponding to the Stokes wing, whereas in [2] the right wing was anti-Stokes. This constitutes a fundamental difference between the experiments [2] and [22]. In the first case, a weaker field was scanned at a fixed strong frequency, in the second, a stronger field was scanned at a fixed weak frequency.

The „unexpected" and misunderstood results of early experiments were presented in 2014 as the first experimental evidence of quantum jumps in [2], interpreted as Bohr jumps. However, they only indicate that even 30 years later the authors of [2] and [9] remained convinced that no other interpretation, except quantum jumps (even if not Bohr jumps), was possible for the early experiments of the Hamburg team (1984–1985). Yet, those experiments demonstrate a completely different effect.

This can be shown by considering the cleaner experiment Fig. 2 (with one barium ion and lower field intensities). The broad peak near the maximum of the contour in Fig. 2 reflects the stepwise transition through the real $6p\,^2P_{1/2}$ state. The deep narrow dip to the right corresponds to a combination transition through a virtual state shifted relative to $6p\,^2P_{1/2}$ so the stepwise and combination transitions are spaced and appear independently. This situation resembles that in Fig. 3.17.b of the monograph [23]. However, near exact resonance ($\Omega = 0$) they cannot be separated, and the situation is described by mutually dependent overlapping contours of different signs and widths (Fig. 3.17.a).

[5] Decades after the [2] publication, they no longer claim Bohr jump discovery but refer to macroscopic quantum jumps primarily registered in [1].

[6] This graph is reproduced as Fig. 2 in our paper.

Detailed descriptions of nonlinear interference interactions of stepwise and combination transitions are given in Chapter III, §8 of [23] and in Chapter 3 of [24]. This phenomenon is called the nonlinear interference effect (NLIE), and it has no relation to either macroscopic or Bohr quantum jumps. It is the third universal effect of nonlinear spectroscopy, manifesting in emission and absorption, complementing level splitting and saturation effects previously known. In this case, a specific modification was recorded.

Hence, the example selected by the authors of [9] to demonstrate „how discoveries can be missed“ was unsuccessful, as the erroneous spectral form explanation led to the conclusion of observing Bohr quantum jumps.

4. Conclusion

Thus, the original claim by the authors of [2] about observing Bohr jumps in single-atom trapped ion quantum jump studies was erroneous. It did not receive support from specialists addressing this problem at that time.

In the authors' next paper [2] on the subject ([5]), the claim no longer appeared, but as [9] shows, even in 2014 they believed they observed quantum jumps, albeit not Bohr jumps, in their early spectral studies. This belief was due to the incorrect interpretation of their earlier experiments conducted in 1984–1985 which actually had no relation to quantum jumps.

The phenomena were specific manifestations of nonlinear interference effects (NLIE). However, interpreting Fig. 2 of [2] with a simple NLIE model based on first nonlinear corrections was hindered by the excessive intensity of the fixed-frequency 494 nm laser initiating NLIE on the $6s^2S_{1/2}$–$6p^2P_{1/2}$ transition. Yet, the Fig. 2 graph of this article, from [22], obtained at lower fixed-frequency laser intensities, could be qualitatively explained by accounting for NLIE induced on the $5d^2D_{3/2}$–$6p^2P_{1/2}$ transition by a moderately intense 650 nm tuning laser.

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Conflict of interest

The authors declare that they have no conflict of interest.

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