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OBSERVATION OF Pr^+ IONS IN PAUL TRAP

J. DEMBCZYŃSKI, D. STEFAŃSKA, G. SZAWIOLA, B. FURMANN,
E. STACHOWSKA, A. JAROSZ, B. ARCIMOWICZ, A. BUCZEK,
W. KOCZOROWSKI, A. KRZYKOWSKI, A. KAJOCH, M. ELANTKOWSKA,
J. RUCZKOWSKI AND W. KOWALKIEWICZ

Chair of Atomic Physics, Poznań Technical University
Piotrowo 3, 60-965 Poznań, Poland

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The first observation of Pr^+ ions stored in a Paul trap is reported. Initially the ions were observed by the electronic detection method, and further the laser induced fluorescence, following resonance absorption at a certain optical transition in Pr^+ ion, which is excited from the ground state, was recorded. Moreover, fluorescence signal following the excitation from a low-lying metastable state could be detected. The Paul trap system and some other parts of the experimental setup were constructed within the frame of this work and thus are briefly described in the present contribution.

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1. Introduction

A quadrupole radio-frequency ion trap, called also after its inventor a Paul trap [1, 2], is in general a versatile device for investigations concerning ions and it has found numerous applications over the past few decades. In particular, it constitutes a very useful experimental tool for spectroscopic investigations of a very high precision (however, until recently those had been restricted to the cases where the excitation followed from the ground state of the ion under study). Though the "effective temperature" of the ions in a Paul trap is very high (of the order of several thousand K, resulting from the characteristic oscillatory motion performed by the ions due to the rf electric field), which results in a large Doppler broadening of the optical transitions (of the order of several GHz), it is nevertheless possible to perform optical-spectroscopic measurements of the hyperfine structure or isotope shifts of the stored ion species in a Doppler-free manner with the use of pulsed or continuous two-step excitation method (as shown on the examples of Th^+ [3–5] and Hf^+ [6] ions). A step further towards high precision in the hyperfine structure measurements is achieved with the use of another two-step excitation method — double optical-rf resonance [7, 8], where the second step excites directly the magnetic-dipole rf-transitions between the hyperfine levels of a certain electronic state. Here the first-order Doppler effect is absent since the wavelength of the rf transitions considered is larger than the geometrical dimensions of the trap, the second-order Doppler effect is negligible and the natural line width is extremely small — the resolution under these circumstances can reach the Hz or even the mHz region. This method has successfully been applied in the case of Eu^+ [7] and Pb^+ [8] ions. Recently its application has been extended also to investigations of metastable levels (within the frame of a project supported by the Volkswagen Foundation [9, 10]).

All the above mentioned experiments were performed in German laboratories (with Eu^+ and Pb^+ at the University of Mainz and with Th^+ and Hf^+ in Karlsruhe Nuclear Research Center), but it has to be stressed that some members of our group were involved, or partially involved, in most of them. The present work reports the first observation of the stored Pr^+ ions, and — to our knowledge — simultaneously the first observation of the ions in a Paul trap ever performed in Poland.

Pr^+ ions in a Paul trap were observed both with the electronic detection method and with the method of laser induced fluorescence. The first method is simpler and, historically, much older (its idea has been introduced by Paul in his pioneer work [1] and described in more detail by one of his co-workers [2]). Unfortunately, under typical circumstances it gives only the information on the average mass of the stored ion species. The second method, however, can assure that the proper ion species are present in the trap, since no other ion sort can emit resonantly fluorescence light under strictly specified excitation conditions and exhibit a specified hyperfine structure pattern.

2. Operation principle of a Paul trap

The operation principle of a Paul trap has already been described in detail in numerous works by various authors [e.g. 1, 2, 11]. Here it will be presented only insofar, as necessary for understanding of the principle of the electronic detection (see Sec. 3) of the stored ions.

The storage quadrupole field inside the trap is obtained when a superposition of an alternate rf voltage (amplitude V_0, frequency Ω) and a constant voltage (U_0) is applied between the ring electrode and the two end-cap electrodes. The electrical parameters determine all the important aspects of the procedure of the storage of the ions in the trap:

- the stability of the solutions of the equations of motion (the operation point of the trap has to be located within the r, z-stable area in the stability diagram, where z is the symmetry axis of the trap and r can be an arbitrary axis in the $x-y$ plane), which is a prerequisite of the possibility of the ions' storage in the trap,
- the depth of the effective storage potential, which determines e.g. the trap storage capacity (the maximum possible number of stored ions) and the amplitude and frequency of the oscillatory motion of the ions (and thus e.g. the effective temperature).

The stored ions perform a somewhat complicated motion, where two main oscillatory modes are superimposed: a micromotion following the driving rf field with a frequency Ω and a comparatively small amplitude, and a macromotion with a lower frequency $\omega_i = \frac{1}{2}\Omega\beta_i(U_0, V_0, \Omega)$ (where $i = r$, z and $0 \leq \beta_i \leq 1$) and a significantly larger amplitude.

The ions have to be generated inside the trap and they can be stored only in an environment free from alien particles, which might randomly influence the motion of the ions under consideration; thus the trap can operate only under ultra-high vacuum conditions. In order to make the storage process effective, it is, however, necessary, to influence the ions' motion in a controlled way, namely to slow them down; this is done with the help of a specially selected buffer gas.

3. Experimental setup

The overall experimental setup used for the observation of the stored Pr^+ ions is presented in Fig. 1. It consists of four basic subsystems:

- a Paul trap with an ultra-high-vacuum (UHV) system, an electrical system for generation of the storage quadrupole electric field, and two detection systems: an electronic detection system and an optical detection system,
- a laser system, consisting of a pump laser, a ring dye laser and its frequency control system, and a frequency marker, and a laser beam transport system,
- a frequency reference system, consisting of a hollow cathode discharge filled with praseodymium and the system for detection of the emitted fluorescence,
- a data acquisition system.

All of these systems are briefly described below.

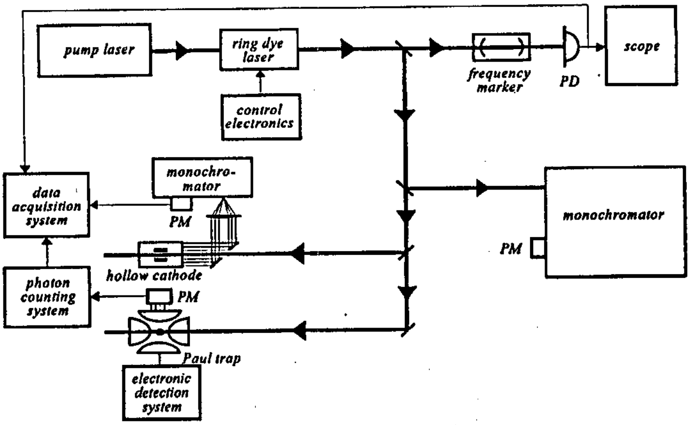

Fig. 1. The experimental setup used for observation of Pr^+ ions stored in a Paul trap.

3.1. Paul trap system

Our trap was manufactured in cooperation with Karlsruhe Nuclear Research Center. A schematic of the trap itself is presented in Fig. 2. The radius of the ring electrode is $r_0 = 1$ cm and the distance from the trap center to the top of either end cap electrode is $z_0 = r_0/\sqrt{2}$. The electrodes of a Paul trap have to be very precisely shaped hyperboloids of revolution in order to avoid, as far as possible, any distortions of the quadrupole electric field. In order to enable any investigations of the stored ions, however, all of the electrodes contain various kinds of holes. One of the end caps has two parallel slits (both 3 mm long), which are used to introduce the wires with the samples placed onto them into the trap (about 0.2–0.3 mm deep). Two circular apertures (of 3 mm diameter) located at the opposite sides of the ring electrode let the exciting laser beam into and out of the trap. At the trap center this beam intersects the ion cloud and the resulting induced fluorescence light is transmitted (to about 50%) through a special system of rectangular holes cut in the other end cap (referred therefore to as the "sieve" end cap).

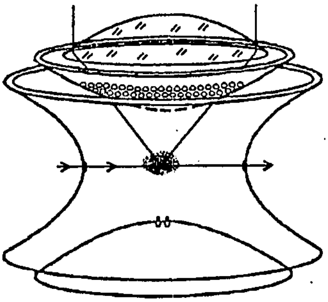

Fig. 2. The schematic of the Paul trap.

The UHV system consists of a UHV chamber and a pump system. The UHV chamber was constructed from standard UHV elements. The pump system is a two-step one; an oil rotational pump provides pre-vacuum (about 10^{-4} mbar) for the turbomolecular pump. The end vacuum is routinely about 10^{-9} mbar or slightly below. The buffer gas, so far being used for experiments with Pr^+ ions, was helium at a pressure range 10^{-3}–10^{-5} mbar (another elements are also considered for that purpose and their performance will systematically be examined in the near future).

The electric supply system for the trap consists of a high voltage rf generator (for Pr^+ ions $V_0 \approx 300$ V at a frequency $\Omega = 2\pi 600$ kHz is routinely used) and a ramp generator, supplying a DC voltage ramp (for the electronic detection, see below) or a specified constant voltage (for Pr^+ ions $U_0 \approx 10$–20 V). This system is integrated with the electronic detection system.

The electronic detection of the stored ions can be performed by tuning their macromotion into the resonance with an external resonant circuit (driven by a generator); when the resonance occurs, the electric signal from the circuit is measurably damped. A resonant circuit for the frequency range 90 ± 4 kHz was constructed in our laboratory.

The optical detection system consists of the optical part (a condensor lens of $f = 27$ mm placed slightly above the "sieve" end cap, a sapphire window in the flange of the UHV chamber, an optical filter, selected according to the special application, and an imaging lens of $f = 100$ mm), and the electronic part (a photomultiplier and a photon-counting system).

3.2. Laser system

The laser system used consists of a tunable single-mode ring dye laser (coherent, modified version of the model CR 699-21), operating on the dye Stilbene 3, optically pumped with an Ar^+-laser, operating in all-lines-UV mode. Further elements of the system are the electronic control system of the dye laser, used for frequency stabilization and scanning, and the frequency marker (an evacuated, temperature stabilized confocal Fabry–Perot interferometer with $FSR = 150$ MHz), providing frequency scale for the recorded spectra.

Our present ring dye laser is a further modification of the already modified model CR 699-21, which has been described in detail in [12]. The present modification consists in a minor change of the resonator configuration (a smaller radius of curvature of one of the mirrors) and in application of still less selective intracavity elements (because of the low pump power available — maximum 1.5 W), which nevertheless assures a reasonably stable single-mode operation and allows the scan ranges of above 50 GHz to be achieved. These large scan ranges are very helpful in the case of Pr^+ ions, where the hyperfine structures in the optical transitions are particularly broad (for comparison: the maximum scan range in the standard model is 30 GHz and it can only be achieved with difficulty). The single-frequency stabilization is performed in the same way as in the standard model.

The system of the beam transport to the Paul trap consists of a few folding mirrors and three lenses located at specific distances over the whole optical path from the laser output mirror to the trap center. Two of the lenses collimate and initially form the beam and the last one, of $f = 75$ cm, placed in front of the entrance Brewster window of the chamber, focuses the beam in the trap center. From the calculations of the expected relative intensity of the fluorescence light a rather weak dependence on the focus diameter results, however, in order to reduce the scattered light intensity one has to clear the apertures in the ring electrode of the trap as far as possible, thus a possibly narrow beam over the whole trap area and in its vicinity is preferred. The transport system was constructed according to the calculations.

3.3. Frequency reference system

The frequency reference system is a system normally used for investigations in a hollow cathode discharge performed in our lab [e.g. 11]. The hollow cathode is filled with praseodymium, and the selected optical transition (see Sec. 4) in Pr^+ ion can be excited simultaneously in the Paul trap and in the hollow cathode. Since in the last case observation of laser induced fluorescence presents no problem, the recorded spectrum is the most certain frequency reference possible, available all the time during the experiment in the trap.

3.4. Data acquisition system

The data acquisition system used in this case is essentially the same as employed in all our previous experiments [e.g. 13]. It consists mainly of the PC with a card of analog-to-digital converters. The signals of the induced fluorescence in the hollow cathode and in the Paul trap are recorded in an alternate manner (i.e. in separate scans) and simultaneously the transmission signal of the frequency marker is registered.

4. Experimental results

As mentioned in Sec. 1, the Pr^+ ions stored in the Paul trap were observed by two independent methods: with the use of the electronic detection and with the use of the detection of laser induced fluorescence. The results obtained are presented below.

4.1. Electronic detection

The simplest way of tuning of the macromotion frequency of the stored ions to the resonance with the external circuit is to vary the constant voltage applied to the trap (provided the macromotion frequency under normal operating conditions is not much different from the resonance frequency of the circuit, so that the process does not violate the stability conditions or move the operating point too dramatically in the stability diagram). If the interactions between the ions in the cloud (Coulomb repulsion) and between the ions and the atoms (or molecules) of the buffer gas are neglected, only the ions of a certain mass, which satisfies the formula for the parameter β_i, can be detected at certain values of the electric parameters. However, these interactions cannot be neglected in practice; the former causes a shift of the constant voltage, where the resonance occurs [2, 9] (the shift amounts to a few V at the maximum cloud density of the order of 10^7 ions/cm^3) and the latter brings the resonances from various ion species present in the trap (if there are more than one) together, so that only the average mass can be determined [3]. This makes interpretation of the electronic detection rather difficult and somewhat uncertain.

The signal from the resonant circuit is applied between the two end caps, i.e. along the z-axis, so it is the frequency ω_z that has to be tuned to the resonance. The intensity of the signal applied cannot exceed a few mV, otherwise the oscillations of the ions brought into resonance might become too strong and the ions could not be stored any longer. The signal from the trap is amplified in a lock-in amplifier and displayed on the digital storage scope.

We obtained resonance conditions for a wide range of V_0 values and at the pressures of the buffer gas helium ranging over about two orders of magnitude. A dependence of the signal amplitude and the storage time on both of the parameters mentioned was observed; systematic investigations are in progress. We also undertook systematic studies aiming at the optimization of the storage parameters (operation point of the trap).

A typical signal of the electronic detection of the stored ions is shown in Fig. 3.

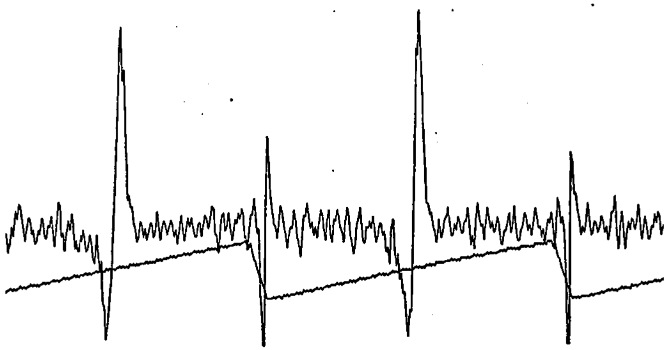

Fig. 3. Electronic detection signal of the stored Pr$^+$ ions.

4.2. Laser induced fluorescence

One of the most important criteria in the choice of the optical transition to be investigated in the Paul trap (on the basis of the tables available for Pr$^+$ ion [14, 15]) was that the lower level has to be the ground level (or, possibly, it might also be a low-lying metastable level [9, 10]); the other was the transition probability. We first considered two possible transitions within the spectral range covered by the dye Rhodamine 6G, but since no induced fluorescence signal from Pr$^+$ ions could be detected in the hollow cathode discharge at the specified wavelengths, we decided for the much less convenient spectroscopy in the blue-violet spectral region (covered by the dye Stilbene 3). Several transitions were examined in the hollow cathode and the most promising of them (from the ground state) was chosen for the experiment in the Paul trap:

- excitation:

$$\underset{E=0.00\ \mathrm{cm}^{-1}}{4f^36s\ ^5I_4}\quad \underset{\substack{\lambda=422.5\ \mathrm{nm}\\(3800)}}{\longrightarrow}\quad \underset{E=23660.2\ \mathrm{cm}^{-1}}{4f^36p\ ^5I_4}$$

- detection (induced fluorescence):

$$\underset{E=23660.2\ \mathrm{cm}^{-1}}{4f^36p\ ^5I_4}\quad \underset{\substack{\lambda=511.0\ \mathrm{nm}\\(360)}}{\longrightarrow}\quad \underset{E=4097.6\ \mathrm{cm}^{-1}}{4f^35d\ ^5K_5.}$$

In order to reduce the part of the background resulting from the scattered laser light and simultaneously maintain the transmission of the fluorescence light at a possibly high level, a color glass filter with particularly favorable transmission parameters ($T(\lambda = 422.5\ \mathrm{nm}) \approx 10^{-10}\%$, $T(\lambda = 511.0\ \mathrm{nm}) \approx 50\%$) was selected. The scattered light background, however, was detectable, which proves the high sensitivity of the photon counting system.

The laser induced fluorescence signal observed in the above scheme is presented in Fig. 4. The lower trace is the reference trsignal observed in the hollow cathode (Doppler broadening of 850 ± 50 MHz) and the upper trace was recorded

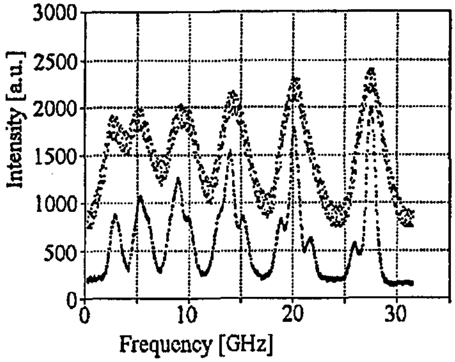

Fig. 4. Laser induced fluorescence of the optical transition at $\lambda = 422.5$ nm in Pr^+ ion (excitation from the ground state) with the frequency scale; upper trace — signal from the Paul trap, lower trace — reference signal from the hollow cathode.

in the Paul trap. In the latter case the signal is not yet very strong and thus the signal-to-noise ratio is lower than in the fluorescence signal from the hollow cathode; however, all the main groups of hfs components are clearly visible and it is the Doppler broadening (of about 2.2 ± 0.1 GHz) and not the signal-to-noise ratio which limits the resolution of the obtained spectrum. Actually no increase in the signal could be obtained by the increase in the exciting laser power (above the initial value of about 1 mW) and the shape of the signal suggested saturation; only a significant decrease in the power (by the factor of about 50) resulted in a distinct intensity decrease in the weaker hfs component groups, so that the observed intensity pattern became consistent with expectations. This saturation of the signal at a relatively low level (of about 20 μW) suggests that the actual number of Pr^+ ions present in the trap at the moment of recording could have been rather small, or the "quenching" via the buffer gas could have been somewhat ineffective (the probability of the decay into the lowest metastable level, located at 441.95 cm^{-1}, is rather high and a noble gas like helium can hardly resonantly receive an energy in this range). In such a case a considerable improvement can be expected, when the storage conditions are optimized. Nevertheless, a signal such as the one recorded is already sufficient as the first excitation step in a double optical-rf resonance experiment.

Recently we have managed to detect a laser induced fluorescence signal following the excitation from the already mentioned metastable level at the following transition:

$$\underset{E\,=\,441.95\ \mathrm{cm}^{-1}}{4f^3 6s\ {}^5I_5} \quad \underset{\substack{\lambda\,=\,430.6\ \mathrm{nm}\\(1500)}}{\longrightarrow} \quad \underset{E\,=\,23660.2\mathrm{cm}^{-1}}{4f^3 6p\ {}^5I_4}$$

(since the upper level is the same as in the previous case, the detection scheme remained unchanged). The result is presented in Fig. 5 as the upper trace, with the lower trace being again the reference signal from the hollow cathode. In this case the signal-to-noise ratio was rather poor at the beginning, but an about twofold increase was achieved by increasing the length of the interaction zone — the exiting

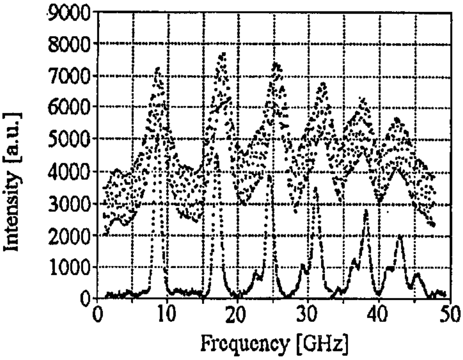

Fig. 5. Laser induced fluorescence of the optical transition at $\lambda = 430.6$ nm in Pr^+ ion (excitation from the metastable state of an energy $E = 441.95$ cm^{-1}) with the frequency scale; upper trace — signal from the Paul trap, lower trace — reference signal from the hollow cathode.

laser beam was reflected back into the trap and thus passed twice through the ion cloud. In this case the saturation of the signal disappeared at a higher power than in the case of the transition from the ground state — already at the value of several hundreds of μW the intensity pattern resembled the expected one. Some further improvement of the signal-to-noise ratio could be expected when phase-sensitive detection was applied and the optimization of the storage conditions (which is in progress) may cause some increase in the signal.

5. Conclusions

In the present contribution the first observation of Pr^+ ions in a Paul trap (and, to our knowledge, simultaneously the first observation of stored ions performed in a Polish laboratory) was reported. Beside the ground state of Pr^+ ion also a low-lying metastable state was observed (which is the second observation of a metastable state of an ion in a Paul trap ever performed, the first one being that of Eu^+ ions [9, 10]). The results are preliminary and further investigations aiming at the optimization of the experimental conditions are in progress. A double optical-rf resonance experiment on the stored Pr^+ ions is planned, which can provide very precise measurements of the hyperfine splittings of the levels of Pr^+ and, further, give important information about the higher electromagnetic moments of the ^{141}Pr nucleus.

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