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Studies of Lifetimes in an Ion Storage Ring Using Laser Technique

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Abstract. The laser-probing method for lifetime measurements of metastable levels, performed by applying the Fast Ion Beam Laser (FIBLAS) method to ions stored in a storage ring, has been developed by the Stockholm group. Recently, we have applied this method to lifetime measurements of close lying metastable levels. In this paper we discuss experimental studies of ions with complex structure and present the first experimentally obtained lifetimes of selected metastable levels in complex systems as Fe^+, Eu^+ and La^+.

1. Introduction

Lifetimes of metastable states, which are in the range of ms to s, find applications both in fundamental atomic structure studies and in relation to astrophysical observations. Laboratory lifetime investigations of highly forbidden transitions are usually difficult since the ions usually are exposed to rapid collisional quenching and special methods for trapping under ultra-high vacuum (UHV) conditions are needed. The long trapping time offered by a storage ring makes it a powerful tool for measurements of long-lived states. The laser-probing technique, recently developed by S. Mannervik and co-workers, which utilities the spectroscopic advantage of the FIBLAS method characterised by sub-Doppler resolution, has been applied to ions stored for a long time in a storage ring. The method has properties of high resolution and high sensitivity and can be used for lifetime measurements of metastable levels, with lifetimes from a few ms up to a few seconds. The results obtained in lifetime measurements of metastable levels in Xe^+, Ca^+ and Sr^+ and comparison with measurements performed in different traps were discussed in paper [1]. Here we discuss the application of the laser probing technique to complex systems for which high resolution and high sensitivity is needed. We present a summary of results obtained in recent investigations of complex systems as Fe^+, Eu^+ and La^+. The particular interest for selection of these ion species will be briefly discussed during the presentation of the obtained results.

2. Experiment

2.1. THE CRYRING FACILITY

The experiments were performed at the ion storage ring CRYRING at the Manne Siegbahn Laboratory in Stockholm [2]. In previous papers, the facility was described in more details [5, 7], and only a short overview is given here. A Nielsen type of hot-cathode, low-voltage electron impact ion source (MINIS) was used for ion production. The ion beam was extracted from the source by electrostatic electrodes at a maximum voltage of 40 kV. After mass separation by an analysing magnet and passage through a radio-frequency quadrupole (RFQ), ions were injected into the storage ring. The ion beam current of the stored ion beams, measured by a calibrated current transformer, was typically 0.5–1 μA. The storage time of singly charged ions at UHV condition in CRYRING (pressure below 10^{-11} mbar) is limited by neutralisation in collisions with particles of the rest gas. The lifetime of the stored ion beam was recorded by counting the number of neutral particles after one of the dipole magnets. The particle detector consisted of a BaF_2 scintillator and a PM tube [3]. The lifetime of the stored ion beam was estimated to be 30 s and 18 s for the Fe^+ and Eu^+ beam, respectively.

2.2. THE CONCEPT OF LASER-PROBING TECHNIQUE

Only a small fraction of all ions produced by the ion source and stored in the ring are in metastable levels. By laser pulses applied at different delay times after injection of ions one can investigate how many ions have remained in the metastable level. The laser frequency is chosen to correspond to a transition from the metastable level to some higher lying level while the number of emitted photons when this level decays is monitored. This prompt fluorescence is proportional to the population of the metastable state. The probing method is destructive and new ions must be injected before the next probe pulse can be applied. The laser pulse duration is chosen to let most of the metastable state population to be pumped out.

As spontaneous decay of the metastable state can occur anywhere in the ring, the detection efficiency for spontaneous decay will be small because the detector covers only a small part of the ring circumference. The high efficiency of the laser probing method is achieved by localising the interaction area of the ions in metastable state and the laser light in front of the detector. This is obtained by applying an additional acceleration of the ions by a Doppler Tuning Device (DTD) [4, 5] by which a local Doppler shift is obtained.

Laser light of the desired wavelength ranges was provided by a laser system consisting of a Coherent 699-29 Autoscan Ring Dye Laser pumped by a Coherent Innova 400-25 Argon Ion Laser. The laser light was transported into the storage ring by a set of mirrors and focussed to a spot diameter of a few millimetres at the interaction region. Since the laser light is merged with the ion beam in collinear geometry, the instrumental line widths of the recorded spectra will be improved by the kinematic compression effect [6].

The laser-induced fluorescence was observed through a mesh in one of the electrodes in the DTD and focussed by a lens system on a single photon counting photomultiplier equipped with a coloured glass filter to block scattered laser light. Typical background rates including scattered laser light are 10 s^{-1}.

2.3. EXPERIMENTAL PROCEDURE

The data points in the decay curve, recorded during the lifetime measurements correspond to different ring cycles. Thus the measurements would be sensitive to a situation where the injected intensity of ions may vary from injection to injection. Also the production of ions in the metastable level by the ion source may vary. During the lifetime measurement five different curves are stored to get full information about possible instabilities (see Figure 1). The points in the *lifetime raw data* curve correspond to the fluorescence counts read in the time window at different delay from ion injection in different ring cycles. The width of the time window, i.e. the time when the shutter is open, has been chosen to assure that more

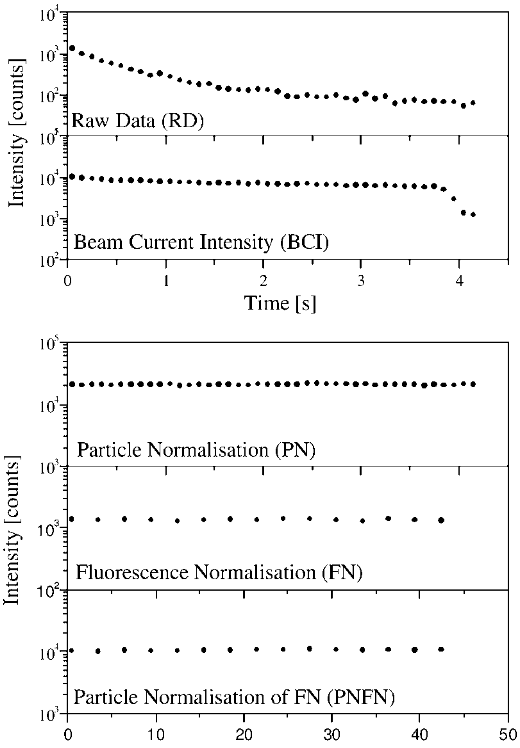

Figure 1. A typical data set for lifetime measurements by the laser probing technique.

than 90% of ions in metastable state are pumped out. The *beam current intensity* in the corresponding time window shows the decay of the stored ion beam current. The *lifetime raw data* and *beam current intensity* curves give information about the lifetime of the selected metastable level and the shape of ion beam intensity decay. Information about the stability of the production and storage of all ions, and in particular, the ions in metastable state are given by the next three curves. The *particle normalisation* curve is obtained by reading the number of counts from the BaF_2 detector for every ring cycle during a fixed externally gated time window of a few hundred milliseconds. The position of this window is chosen after inspection of the ion beam current curve for the particular stored beam. To monitor the stability in production of ions in metastable state, laser induced fluorescence at a *fixed* time delay is recorded every fourth cycle. This signal is stored in the fourth curve. The corresponding particle normalisation is also recorded. It should be noted that flat *particle normalisation* and *fluorescence normalisation* curves show that the production of ions and ions in metastable state by the ion source was stable.

The number of stored ions in metastable state can be changed by collisional interaction with the residual gas due to excitation, deexcitation and neutralisation [7]. Besides injection instabilities, these processes could also be sources of systematic errors in the determination of radiative lifetimes. In order to subtract contributions from ions in metastable state produced in collision processes, a separate repopulation curve is determined [5]. This curve is recorded in a similar way as the decay curve. In the beginning of every ring cycle, the ions in the metastable state produced by ion source are, however, quenched by a laser pulse (typically a hundred milliseconds long). A second laser pulse will be applied with different delays from the injection in order to investigate how many ions in metastable state are created by collisional interaction. The contribution from repopulation can subsequently be accounted for by subtracting the repopulation curve from the lifetime curve.

To distinguish between radiative decay and depopulation (deexcitation and neutralisation) due to the collisions with atoms and molecules of the residual gas, the lifetime measurement was also performed as a function of rest gas pressure. Plotting the decay rate of the metastable level versus relative pressure (Stern–Vollmer plot), the radiative decay will be obtained by extrapolation to zero pressure.

The ion beam decay curve can be recorded by feeding the signal from the particle detector into a multichannel scaler (MCS) triggered at ion injection. This curve usually shows the existence of initial loss of ions, which is much faster than later in the cycle where ions are lost mainly due to the neutralisation. To reduce the influence of this initial loss, the measurement should be started with a delay after injection. This delay time was typically chosen to be 50–100 ms. Longer delay costs loss in the number of ions in metastable state. Corrections for the remaining effect will be obtained in the data analysis.

2.4. LIFETIME MEASUREMENTS FOR COMPLEX IONS BY LASER-PROBING TECHNIQUE

In our lifetime measurements the ions are excited into the metastable states directly in the ion source. The fraction of ions in metastable state produced in the ion source is difficult to determine and it can only be roughly estimated to be less than 1%. In the case of Fe^+, this fraction of ions in metastable state will be distributed over 62 possible metastable levels. This fact explains that the signals in the Fe^+ measurements were weak and that, for example, the $3d^5\,4s^2a^6S_{5/2}$ level in the best case gave a signal-to-noise ratio of 10 [9]. To illustrate the weakness of this particular signal, Figure 2 shows a comparison of the resonance signals obtained for Ca^+ and Fe^+, respectively. The high detection efficiency of laser-probing method, however, is more than a factor of 10^3 higher than for passive observation of spontaneous decay from the metastable level, so lifetime investigations of metastable levels in Fe^+ has still been successful.

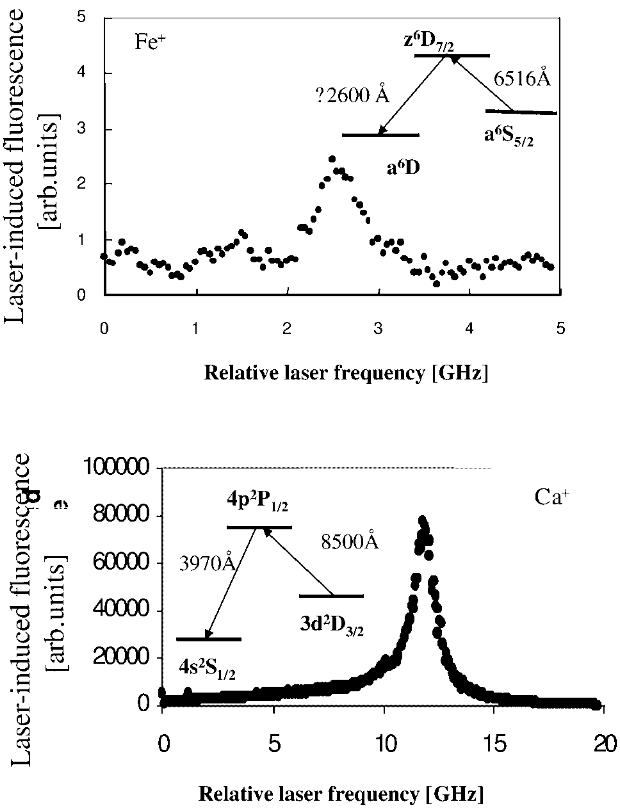

Figure 2. Laser-induced fluorescence spectra obtained using the excitation schemes shown in insets. The signal was 1000 time stronger in the Ca^+ case compared with the signal obtained in the Fe^+ case.

No repopulation was observed for the levels studied in Fe^+ [9, 10] and Eu^+ [11]. The explanation could be that, because of unfavourable signal-to-noise ratio, the repopulation cannot be resolved from the background. Also, the repopulation, as a process induced by collisional excitation with the rest gas, is not selective and in the case of Fe^+ it will be spread over all 62 metastable levels. In the lifetime investigation of the a^1G_4 and b^1D_2 in La^+ [12], repopulation was observed and metastable ions produced by collisional quenching were subtracted. Here it was found that the repopulation of the two levels was different.

Collisional destruction rates were extracted from lifetime measurements performed at the base pressure (10^{-11} mbar) and at raised average pressure. Since at raised pressure two counteracting processes are enhanced: the collisional destruction and repopulation, the estimation of the collisional deexcitation rate can be difficult. For the metastable levels in Fe^+ collisional destruction was only observed for the $3d^6(^3H)4s\ b^2H_{11/2}$ level. Using a Stern–Vollmer plot this rate was estimated to be 0.028 s^{-1} at base pressure [10]. Inclusion of this effect changed the extracted radiative lifetime of this level by 10%. Due to the fact that the investigated levels have similar energy, the observed collision effect was assumed to be approximately the same and was used to subtract the collision destruction effect for lifetimes of all other levels in Fe^+. For the investigated levels in Eu^+ no significant extra contribution from collisional destruction was observed as the average pressure in the ring was increased by 50%. The contribution from collisional destruction for the a^1G_4 level in La^+ was estimated to be 2%. Similarly as in the case of Fe^+, this observed collision rate was used to subtract the collision destruction effect for the b^1D_2 level in La^+.

As mentioned above the ion beam decay curve usually exhibits an initial loss of ions, which has a slope much steeper than the later part of the particle curve where the slope is determined by the neutralisation rate. The strength of this initial peak has been found to depend on the ion beam intensity but also on the specific settings of ring parameters. The ion beam current of stored La^+ ions was about 20 times lower than the corresponding beam current for measurements with Eu^+ and Fe^+ and the transient peak was not visible here (see Figure 3). According to this fact we conclude that the intensity and the slope of transient part can be changed by changing the ring settings and decreasing the beam intensity. Since the extra loss is of instrumental character it will be equal for both ground state ions and ions in the metastable state. A way to compensate for this extra loss is to divide the fluorescence decay curve with this extra decay component extracted from the ion beam current intensity curve (curve No. 2 at Figure 1). This procedure has been utilised in the analysis of Eu^+ and Fe^+ and the correction resulted in an increase of the lifetime up to 10–20%.

Since the probing procedure is sequential, normalisation to the number of ions in metastable state produced in the ion source should be performed. The production of ions in metastable state was very stable in experiments performed with Fe^+ and Eu^+ ions, and the effect of this normalisation was negligible. For La^+, however,

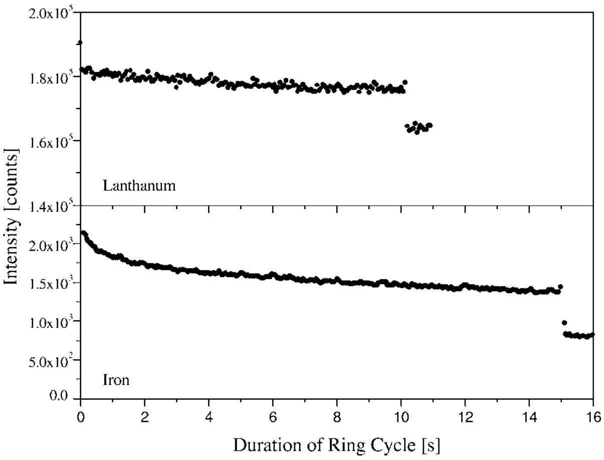

Figure 3. Comparison of the ion beam current curves for the La^+ and Fe^+ ion beams obtained with the BaF_2 detector. The data analysis in the La^+ case was started from time corresponding to second point on the curve.

the production of ions in metastable state was decreasing slightly during the time while the total production of ions was stable (see Figure 4). Here normalisation to the *fluorescence normalisation* curve gave a significant (but small) correction to the decay rate of the two investigated levels.

3. Results for complex ions

3.1. LIFETIME MEASUREMENT OF METASTABLE LEVELS IN Fe^+

High cosmic abundance makes iron very important for astronomical investigations. Therefore Johansson recently started an international project for collection of radiative decay rate data denoted as FERRUM [8]. In collaboration with his group we have made the first experimental investigation of decay rates for the $3d^5\,4s^2 a^6 S_{5/2}$, $3d^6(^3D)4s\ b^4D_{7/2}$, $3d^6(^3G)4s\ a^4G_{9/2}$ and $3d^6(^3H)4s\ b^2H_{11/2}$ levels in Fe^+. More details about these particular measurements are reported in [9] and [10] and here we only summarise the results.

The lifetime of the $3d^5 4s^2 a^6 S_{5/2}$, $3d^6(^3D)4s\ b^4D_{7/2}$, the $3d^6(^3G)4s\ a^4G_{9/2}$ and $3d^6(^3H)4s\ b^2H_{11/2}$ levels were measured by probing with the laser wavelengths locked to 6516, 6456, 5426 and 5536 Å, respectively. After data analysis, which was made in several steps, the lifetimes were determined to be 230 (30) ms, 530 (30) ms, 670 (20) ms and 3.8 (0.3) s, respectively. As discussed in the previous section, the high density of metastable levels and the low population of each state, was a strong challenge for the laser probing technique.

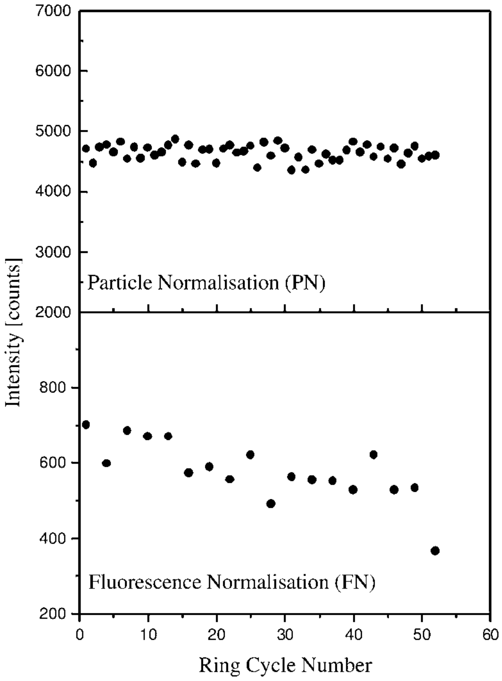

Figure 4. The *Particle Normalisation* and *Fluorescence Normalisation* curve recorded in the lifetime investigations of metastable levels in La$^+$. The production of La$^+$ metastable ions was decreasing during the time while the production of ions was stable.

3.2. LIFETIME MEASUREMENT OF METASTABLE LEVELS IN Eu$^+$

Singly charged europium has a very complex atomic structure due to the open f-shell. Many possible energy levels and crowed optical spectra make rare earth elements very complicated for experimental and theoretical analysis. No experimental or theoretical investigations of forbidden transitions have been reported until now.

The first experimental lifetime investigation of metastable levels in Eu$^+$ was done by applying the laser probing method. The lifetime of the a^9D_J ($J = 2$–5) levels were all found to be around 1 s. The procedure of data analysis and error estimation, as well as more experimental details, were discussed in [11].

3.3. LIFETIME MEASUREMENT OF METASTABLE LEVELS IN La$^+$

Lanthanum, with atomic number 57, is the first of the rare earth in the lanthanide series and its electronic structure is less difficult to handle in theoretical investigations as compared with europium, which is positioned in the middle of the series. This was our motivation for performing lifetime measurements of the a^1G_4 and b^1D_2 levels in La$^+$ [12]. As an integrated part of this investigation, calculations were performed by E. Biémont. The experimentally obtained lifetimes of the a^1G_4

and b^1D_2 metastable levels are 5.2 (0.2) s and 2.1 (0.3) s, respectively and corresponding theoretical results were 5.09 s and 2.28 s. The lifetime of the a^1G_4 level is the longest lifetime measured so far by the laser-probing technique in the ring.

4. Summary

In this paper we have discussed the laser probing technique of a stored ion beam as a method for experimental determination of lifetimes of metastable levels in ions with complex electronic structure. We have presented recent results obtained in lifetime investigations of metastable levels in Fe^+, Eu^+ and La^+. The results prove that the laser probing technique is a powerful tool for lifetime studies of ions with complex atomic structure.

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