

Violet Diode Laser in Time-Resolved Stored-Ion Spectroscopy

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Received May 1, 2003; accepted in revised form October 6, 2003

PACS Ref: 32.70.Cs, 42.62.Fi, 42.55.Px

Abstract

Lifetime measurements of the metastable $3d\ ^2D_{3/2}$ level in singly charged calcium were performed using the newly developed violet diode laser operated in an external cavity arrangement. The laser was employed on stored ions at the CRYRING facility to optically pump the resonance line at 397 nm. In combination with laser probing at 866 nm this detection scheme gave the possibility to record the lifetime of this level, which has a radiative lifetime of about 1 s.

1. Introduction

1.1. Violet diode lasers in spectroscopy

The spectroscopic applicability of diode lasers was largely increased after the introduction of GaN violet and blue cw diode lasers [1]. While offering the advantage of a low cost, robust, turn-key light source, these lasers have the drawback of needing an external tunable cavity to operate in a single longitudinal cavity mode. The output power of the initial violet/blue lasers commercially available was about 5 mW, although at present 30 mW diode lasers are also offered. First atomic physics experiments using such tunable violet/blue diode lasers concerned the second resonance lines of potassium at 404.5 and 404.8 nm [2]. Absorption, fluorescence and opto-galvanic spectroscopy were demonstrated. Ultrasensitive atomic detection employing two-tone frequency modulation spectroscopy was demonstrated on potassium and on the lead 405.8 nm line originating from a weakly populated metastable level [3]. Sum-frequency generation to the 253.7 nm mercury line was also demonstrated by mixing the light from the 404 nm and a 688 nm diode laser in a BBO crystal [4]. In low pressure samples the well-resolved spectral structures of mercury isotopes could be seen. Basic properties and spectral characteristics of violet/blue external cavity diode lasers (ECDLs) have been investigated in [5,6]. A further use of violet diode lasers is as excitation sources for laser-induced fluorescence. A very compact fibre-optic fluorosensor combining a violet diode laser with an integrated spectrometer has proved to be very useful, e.g., for early cancer detection [7].

In this paper, we report on the use of a violet ECDL for measurements in an ion storage ring of the lifetime of a metastable state in Ca^+ . The laser generates the wavelength corresponding to an electronic transition of the ion.

1.2. Lifetime measurements on metastable states in Ca^+

Recently, much interest in the atomic physics community has been focused on the metastable $3d\ ^2D_{3/2,5/2}$ levels in Ca^+ . This is partly due to the fact that the atomic system has a single electron outside a closed shell. Hence, selective studies of the interaction between the valence electron and the core could be performed, which makes it a good test system for theory [8,9]. The simple level structure permits laser cooling and extensive experimental studies have been done in ion traps. Such studies have different motivations. Besides the interest in testing theory, the Ca system is one of the candidates for constructing a new and more accurate frequency standard [10]. In addition, experimental work is in progress for using metastable Ca^+ in an ion trap as a quantum bit (qbit) [11].

Laser cooled ions in a trap could be used to determine the radiative lifetime of metastable levels. Studies of quantum jumps of one single ion [12] can be used to measure the lifetime of the $3d\ ^2D_{5/2}$ level. This was, so far, most accurately done by Barton *et al.* [13] (0.6% error bars) (earlier measurements are referred to in that article). The $J = 3/2$ level could, however, not be determined by this method. This level can be measured less accurately (with 5% uncertainty) by laser probing as was done at CRYRING previously [14].

Singly charged Sr has a homologous level structure with corresponding metastable levels; $4d\ ^2D_{3/2,5/2}$. The lifetimes of these levels have also been measured at CRYRING by the laser probing technique (LPT) [15]. For the $4d\ ^2D_{3/2}$ level also another method based on optical pumping (OP) was utilised at CRYRING [16]. In the latter method a laser excited essentially all ions into one specific metastable level ($4d\ ^2D_{3/2}$) and with the heavily populated level it was possible to observe the forbidden transition directly. It was found that this method improved the accuracy substantially (by a factor of five in this case).

As mentioned, only the lifetime of the $3d\ ^2D_{5/2}$ level has been very accurately determined in Ca. There are experimental indications that the lifetime difference between the two fine structure levels might be significantly larger than what theory predicts (i.e., 3%) [8]. Consequently, it is of great interest to improve the experimental lifetime accuracy substantially, in particular for the $3d\ ^2D_{3/2}$ level.

Due to these facts, our aim was to try to improve the measurements at CRYRING by applying OP to the two metastable levels in Ca^+ . This should be possible by the use of newly developed violet diode lasers that can pump the resonance lines in singly charged Ca.

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2. Experimental

2.1. Ion storage ring

Techniques for lifetime measurements of metastable levels have been developed at the ion storage ring CRYRING [17] at the Manne Siegbahn Laboratory in Stockholm. This ring consists of 12 straight sections and 12 bending magnets. The circumference is 51.6 m and the base pressure is below 10^{-11} mbar. The ring contains a number of components as a high-frequency acceleration stage and an electron cooler and it is equipped with an ion source for highly-charged ions (an electron beam ion source, EBIS) as well as a conventional source for production of singly charged ions. In the present experiment, singly charged ions were produced in a conventional discharge source by electron impact. It has been found that a fraction of the ions leave the source in metastable levels. However, this fraction is usually very small. The ions are accelerated from a high-voltage platform (usually at 40 kV). They are subsequently separated in a 90° bending magnet and only ions of one specific isotope are injected into the ring.

2.2. Laser probing technique

The LPT developed at CRYRING has been described in detail previously; see for example Ref. [18]. Here the population of a metastable level is investigated by inducing a laser transition from the metastable level up to a more highly excited level. This level will promptly decay since it has allowed decay modes. The fluorescence from this decay will give a measure of the population of the metastable level. If this is done in a collinear geometry the resolution will be high due to the kinematic compression effect [19] (i.e., the velocity distribution being narrowed by the acceleration). The laser light and the ion beam are merged over a distance of about 4 m and the laser light leaves the ring through a specially designed tandem Brewster window (for minimum back reflection) [20]. The interaction length should, however, be restricted to a shorter distance to achieve efficient fluorescence collection. This has been done by introducing a Doppler tuning device (DTD) that locally changes the ion beam velocity to yield a different Doppler shift of the laser light [20]. The DTD consists of a number of tubes that could be put at high voltage. The ion beam will pass axially through the tube and be accelerated or retarded depending on the voltage applied. For a few centimetres in the middle of the DTD the beam will have a fixed velocity different from its velocity in the rest of the section. The laser frequency is tuned into resonance for the ion velocity in the DTD and the fluorescence will be localized to a short distance inside the device. Using a lens system, the fluorescence is focussed on a photo multiplier tube (PMT) counting photons. A coloured glass filter in front of the PMT suppresses scattered laser light.

Since the laser probing gives information about the population of the metastable level, the lifetime of the level can be determined by probing the population at different times. With time zero defined as the time when the ions are injected into the ring, probing can be applied at a later occasion. This is done by controlling a mechanical shutter that opens to transmit laser light into the ring at a specific delay time. This delay time can be systematically varied in order to record the lifetime of the metastable level. It

should be noted that the probing technique is destructive since it implies quenching of the level. Consequently, only one time point can be obtained for each ion injection. The systematic variation of the delay time is controlled by a computer that is triggered by the control system of the ring in order to induce the timing correctly.

At a beam energy of 40 keV, which is usually used, ions will primarily be lost by neutralisation in collisions with atoms and molecules in the rest gas. Due to the procedure of sequential probing, for each ion injection into the ring it is important to monitor the number of ions, particularly the number of ions in the metastable level. The technique is based on the assumption that the beam intensity is constant for every injection. Methods have been developed to continuously monitor these quantities and to permit correction if variations would occur. In the experiments performed by LPT so far, the corrections have been negligible or small.

A tunable ring Ti:Sapphire laser (Coherent 899-29) pumped with an Ar ion laser (Innova 400-25) was used for probing the population of the metastable level in Ca^+ .

2.3. Optical pumping

As mentioned above, in the previous experiments using LPT [14], the small fraction of ions excited to the metastable levels in the ion source was probed to determine the lifetimes. For Ca we have estimated this metastable fraction to be at most 1%. In that case infrared laser light (866, 854 nm) was applied and fluorescence in the violet wavelength region (397, 393 nm) was detected; see Fig. 1.

In OP the idea is to apply the laser light in the violet in order to transfer ground state ions to the metastable levels. In Sr^+ we were previously able to transfer most of the stored ions to one of the metastable levels [16]. With such a large population in the metastable level it was possible to detect the forbidden transition directly and to measure the lifetime of the metastable level by the use of a multichannel scaler (MCS). It turns out that systematic errors can be reduced thereby [16,18]. In particular, the contribution from collisional excitation—repopulation [18] is significantly smaller.

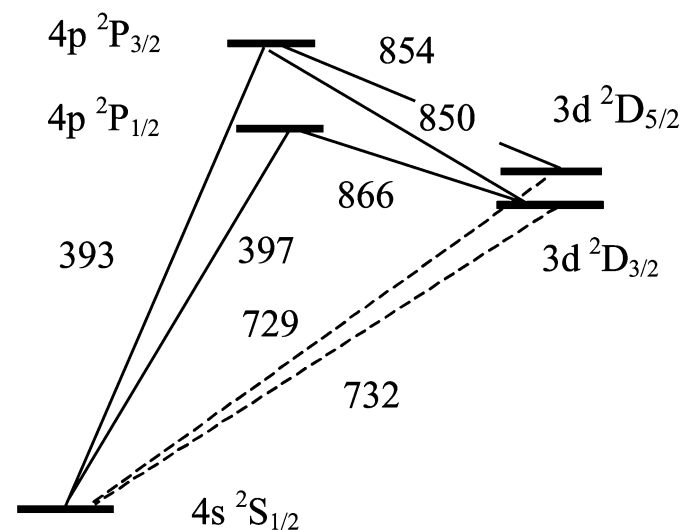


Fig. 1. Schematic level diagram for Ca^+ showing levels and transitions of interest for this work. Wavelengths are given in nm (in the rest frame). Dashed lines indicate forbidden transitions.

In the present experiment we wanted to extend the use of the violet diode laser by employing it for lifetime measurements at CRYRING. The laser should generate the wavelength corresponding to either of the two transitions at 397 or 393 nm (corrected for the Doppler shift). The longer wavelength should be used to populate the $3d\ ^2D_{3/2}$ (similarly as was done for Sr^+). The shorter wavelength will populate $3d\ ^2D_{5/2}$. As can be seen in Fig. 1, there is, however, also a weak decay branch from the upper $3p\ ^2P_{3/2}$ to the $3d\ ^2D_{3/2}$ level. To make the experiment clean an infrared repumper at 850 nm is needed to empty this state.

2.4. Diode laser setup

A violet diode laser acquired from Nichia Corporation (type NLHV500E) was used as the light source for OP. It has a nominal operating wavelength of 397 nm at room temperature and a maximum cw output power of 5 mW. The diode laser was mounted in a thermo-electrically cooled holder (Thorlabs TCLDM9) and operated with a low-noise laser diode driver (Melles Griot 06DLD103). The diode laser was operated just a few mA above the threshold current, of 36 mA, in order to avoid fast degradation of the laser chip. Coarse wavelength tuning of the laser was accomplished by changing the temperature of the laser capsule.

The free-running laser output is composed of multiple longitudinal modes, separated by approximately 0.05 nm. These can be seen in Fig. 2, which shows a spectrum of a similar blue diode laser measured with a high-resolution spectrometer. An external feedback cavity employing a

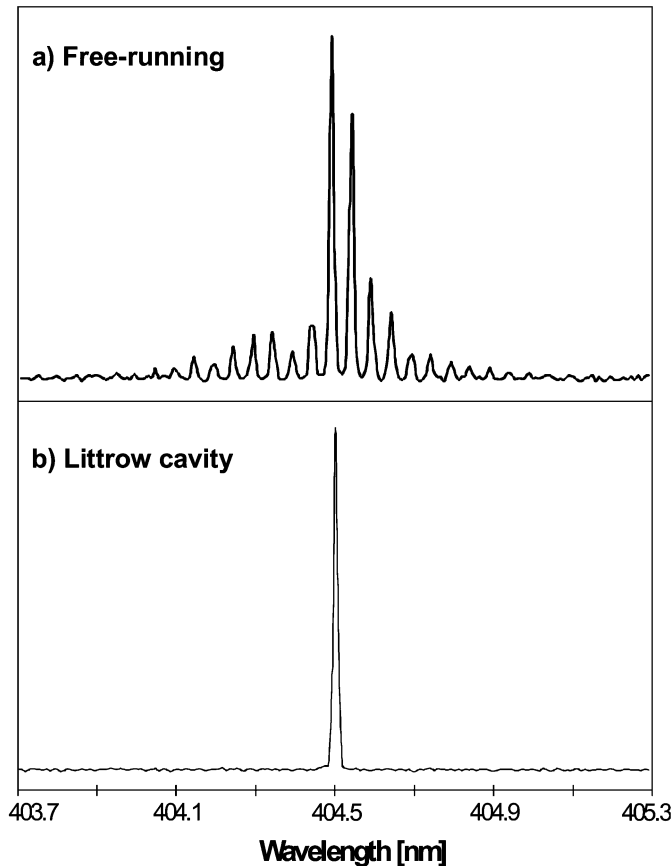


Fig. 2. Blue diode laser output spectra when operated with (a) and without (b) an external cavity.

diffraction grating in the Littrow configuration was used to ensure single-mode operation. This improved the spectral performance insuring efficient absorption of resonant radiation by the Ca^+ ions, since in the case of multimode behaviour only one oscillating mode at the time can interact with the ions and contribute to the optical pumping process. The external cavity was built using a Thorlabs system based on the diode laser holder, a piezo-electric mirror mount (KC1-PZ), a piezo-electric driver (MDT-690) and a 2400 l/mm grating (Edmund Scientific 43224). A moulded glass aspheric lens designed for a 780 nm wavelength (Geltech C230TM-A) was used to collimate the divergent output beam from the diode laser.

The experimental arrangements employed in the present experiments are shown in Fig. 3. The laser had the same direction as the stored ions, and both beams were overlapping over a distance of about 4 m inside the ring. While most of the diode laser light was transmitted through a window into the ion storage ring to excite the Ca^+ ions, a small part of the laser beam was deflected by a neutral density filter and sent to a wavemeter (Burleigh WA-4500) for continuous wavelength control. As part of the wavelength calibration process, absorption spectroscopy was employed in the wavelength range of interest using another Nichia blue diode laser tuned to the $4s\ ^2S_{1/2} - 5p\ ^2P_{3/2}$ transition in potassium at 404.8 nm. The cross section of the beam emerging from the external cavity was relatively large and had a non-Gaussian intensity distribution. This caused inefficient coupling into the 2-mm diameter input aperture of the wavemeter, as well as cumbersome adjustment of the wavemeter's internal etalons. The beam was therefore compressed with a two-lens telescope to a diameter of 1 mm that also homogenised the intensity profile. Light feedback from optical surfaces, that could cause an unstable laser frequency, was avoided by intentional misalignment of the telescope lenses. The cross section of the laser beam sent out from the external cavity was about 2×5 mm, but due to diffraction and aberrations in the collimating lens, it diverged to a size of 17×13 mm at a distance of 10 m, corresponding to the path length inside the ion storage ring.

The external cavity setup allowed for convenient continuous tuning of the laser output by changing the grating angle and the distance between the grating and the diode by means of the high accuracy piezo translators. The mode-hop-free continuous tuning range was 9 GHz,

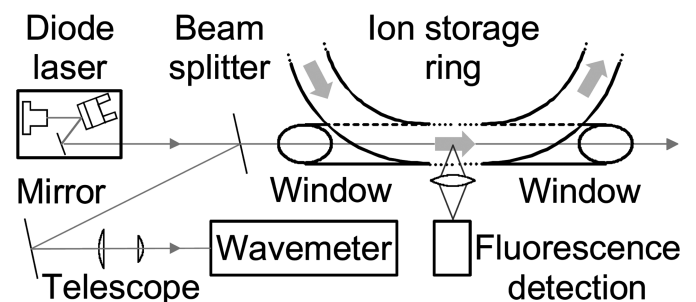


Fig. 3. Schematic illustration of the experimental setup, indicating the violet diode laser source, the wavelength calibration equipment, the ion storage ring, and the detection part. The infrared laser used for the probing is not included. This laser beam is also merged collinearly with the ion beam.

corresponding to the free spectral range of the external cavity. This can be compared with the coarse, discontinuous tuning range of approximately 3 nm. The laser bandwidth was also narrowed down in the external cavity setup to an estimated value of approximately 5 MHz. The alignment of the external cavity was optimised by repetitive adjustments of the beam collimation, as well as the vertical and the horizontal angles of the grating, until the threshold current was reduced as much as possible and the continuous tuning range was maximised. Finally, the external-cavity-coupled laser was operated in a single longitudinal mode, and emitted an output power of about 1 mW.

3. Measurements and results

The wavelength of the violet diode laser was fine-tuned by changing the grating angle to the correct resonance line of the Ca^+ ions, with due consideration to the Doppler-shift induced by the coasting ions in the storage ring. To increase the frequency stability of the laser output, careful adjustment of the temperature and operating current was performed. Readjustments of the operating parameters were required during the measurements, but typical values of the operating current and the temperature were 39.5 mA and 25 °C, respectively.

The single mode operation of the laser was verified by using the interference fringes generated by the two internal etalons of the wavemeter. The wavelength stability of the diode laser during the measurements was also ensured by judicious mechanical and thermal isolation of the setup. The external cavity was encapsulated inside a metal cover, shielding it from external magnetic fields and air turbulence. These measures limited the drift to less than 1 GHz per hour, a drift that could be controlled by adjusting the piezo translators of the external cavity.

In order to tune the laser frequency of the violet diode laser into resonance with an atomic transition it was necessary to obtain a response signal that indicates that excitation was achieved. For lifetime measurements at CRYRING the resonance is usually observed by laser-induced fluorescence from another decay channel at a wavelength different from that of the laser. The laser frequency is scanned by specially tailored electronics that synchronize the scanning with the ring cycles, and fluorescence from the ion beam is detected by a photomultiplier.

The problem in the present situation was that the infrared fluorescence (Fig. 1) was weak. The strongest decay channel of the upper state was back to the ground state. In the previous Sr^+ experiment it was possible to detect fluorescence at the same wavelength as was applied by the exciting laser. In the present experiment this was, however, not possible. A weak halo originating from diffuse light inside the diode laser capsule, as well as light scattered by the input and output Brewster windows of the ion storage ring, caused a background of stray laser light that blocked the possibility to detect the fluorescence simultaneously.

To solve this fundamental issue, another method was developed based on laser probing, which could be applied when the violet diode laser was blocked by the shutter. The

basic idea is to use laser probing to obtain a response signal instead of detecting prompt fluorescence. This was done in the following way: First infrared laser light tuned to the 866 nm transition quenched the initial population of the $3d\ ^2D_{3/2}$ level (from the ion source). This was done to more easily detect the OP induced by the violet diode laser. The next step was to apply the violet laser in order to excite the specific level. As a third step the infrared laser was turned on again in order to detect if excitation was achieved. By this method high sensitivity was obtained. A typical data set is shown in Fig. 4.

Tuning the violet laser into resonance could then be done by recording the fluorescence intensity of the second peak as a function of laser frequency of the violet laser. In laser probing experiments the laser frequency is tuned to fulfil the resonance criterion only in the central electrode of the DTD (see Fig. 5) [18,20]. Thereby, the resonance is only achieved over a distance of a few cm and the laser-induced fluorescence will be restricted to a region in front of the PM tube. For OP by the violet diode laser it is, however, important to have longer interaction lengths to get high efficiency. Consequently it was essential to find the laser wavelength that gives resonant excitation in the whole interaction section (with a length of about 4 m) and not only in the DTD. As mentioned above the ions are accelerated to 40 keV before injection into the ion storage ring, where the ions are stored. The middle electrode of the DTD is held at a potential of -2 kV, which causes the ions to accelerate to a total energy of 42 keV at which the ions travel about 100 nanoseconds (about 5 cm) before they are retarded as they leave the middle electrode. Since the laser light is directed collinearly and along with the ion beam, the ion will experience a Doppler shift to the red. The red shift will be larger the higher the velocity. At the present wavelength (397 nm) the Doppler shift is 0.58 nm for 40 keV and for 42 keV the shift will increase by 15 pm. For a fixed laser wavelength, the ions will experience a laser scan of 15 pm as they travel through the common ring section. Since the laser light is red shifted by the Doppler effect it is necessary to blue tune the laser light relative the resonance wavelength for ions at rest.

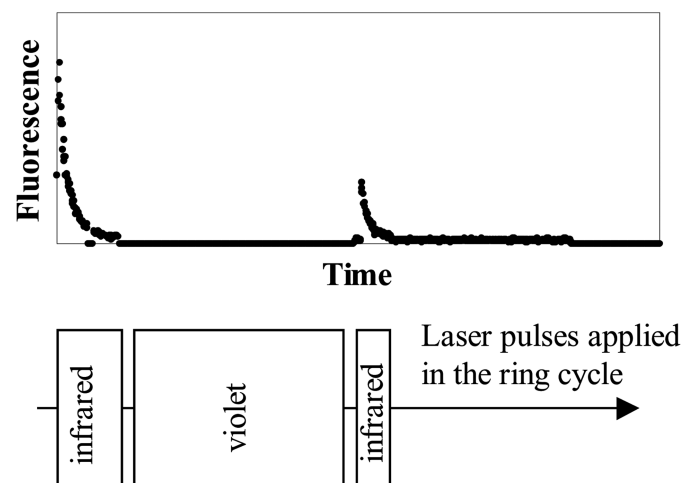


Fig. 4. Experimental multiscale curve showing the detected fluorescence. The initial population of the metastable level was pumped out by a 200 ms infrared laser pulse. Ground-state ions were then pumped to the metastable level by a 750 ms pulse of violet laser light. Finally, a second infrared pulse (100 ms) was applied to detect successful excitation.

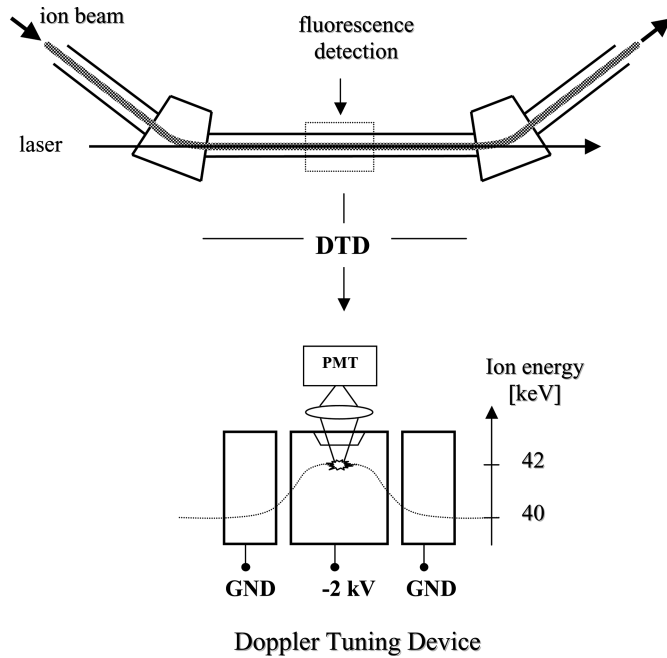


Fig. 5. Schematic figure of the beamline section containing the Doppler Tuning Device (DTD) used for localising the excitation in laser probing. The centre cylinder is put on -2 kV and the dashed curve shows schematically the kinetic energy. The potential difference will cause acceleration and the corresponding Doppler shifted laser wavelength will vary at different positions along the beam axis inside the DTD.

In Fig. 6, the experimental results of the laser wavelength tuning procedure are given. The figure shows the laser wavelength of the violet diode laser on the abscissa and the response signal obtained by laser probing on the ordinate. As can be seen from the figure, OP to the metastable level starts at the wavelength 396.363 nm . This reflects excitation of the fastest ions in the middle of the DTD. As the laser wavelength is tuned redder, ions in the field gap (at energies between 40 and 42 keV) are excited. Finally, the wavelength reaches a value where the ions at 40 keV are excited. This is seen as a sharp strong peak corresponding to an efficient pumping process due to the long interaction length. It can easily be confirmed that the total structure corresponds to a Doppler shift difference between 40 to 42 keV ions. The strong peak was scanned separately in order to optimise the wavelength setting and the resonance wavelength was found to be 396.3777 nm for 40 keV ions.

The original idea was that the violet laser should be used to heavily populate the specific metastable level. From such a high population it was expected that direct observation of the forbidden decay should be possible and that the lifetime could be measured with a multichannel scaler (MCS). This was done in Sr^+ previously [16]. It was found, however, that the signal-to-background ratio was not sufficient to obtain a high-quality signal, partly due to the lower transition rate in Ca^+ (a factor of 2.5 lower than for Sr).

Instead it was possible to combine OP and laser probing for lifetime measurements. The main idea is then to populate the specific level when the beam is stored. Thus it is possible to measure the lifetime later in the cycle when the initial non-exponential beam loss has faded away [18]. In our laser probing measurements the initial instrumental loss processes in the ring cycle limits the accuracy of the experiment [14] to 5% . With the new method this could be avoided.

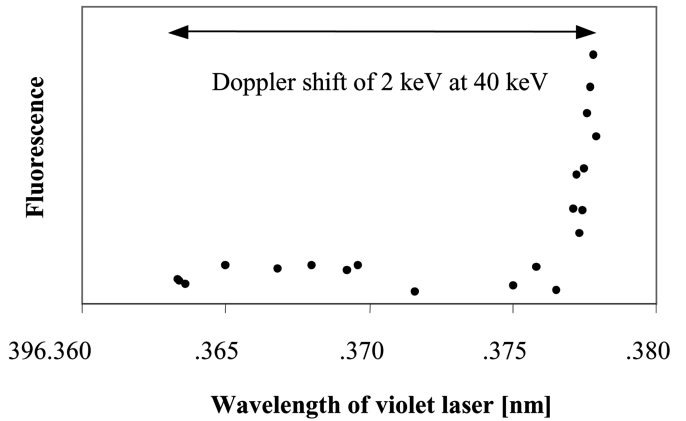


Fig. 6. Experimental investigation, according to the scheme in Fig. 4, of optical pumping efficiency as function of the wavelength of the violet diode laser. The shortest wavelength corresponds to excitation in the middle of the DTD (highest ion velocity). Here pumping is weak due to short effective overlap between the beams. The signal becomes strong when the laser wavelength reaches the value corresponding to resonance at injection energy (40 keV), for which the overlap distance is 4 m .

The Doppler-broadened Ca^+ ion line has a linewidth of around 500 MHz reflecting the velocity spread of the beam (and field penetration into the centre electrode). It means that the spectrally narrow diode laser light would only interact with part of the ions in the storage ring, burning a hole in their velocity distribution. This was prevented by scrambling the frequency of the diode laser light over approximately 600 MHz , by applying a current ramp of 8 Hz on one of the piezo-electric translators controlling the tilt of the external cavity grating.

Although the time allocated for this experiment by the Manne Siegbahn Laboratory (a Swedish national facility) was limited, we could, after combining optical pumping and laser probing, convincingly demonstrate the potential of the new method. One example is given in Fig. 7. The upper curve shows the decay of the metastable $3d\ ^2D_{3/2}$ level. The decay of the level was studied during 10 s . The early part of the curve shows the decay of the ions that left the ion source in the metastable level. After 5 s the violet laser is turned on, which is seen as an increased intensity due to scattered laser light. During this time ground state ions are optically pumped to the metastable level and the population increases by a factor of 20 in this level. By laser probing, the decay of this new fraction is recorded. It should be noted that although the intensity of the second curve is not higher on an absolute curve, but since the number of ions in the ring is reduced after 5 s , the relative fraction of ions in the metastable level is at least twice as high as what is obtained directly from the ion source. This can be deduced by comparing the probe curve with the curve just below which shows the number of ions stored in the ring. During the present experiment only the $3d\ ^2D_{3/2}$ level was investigated. In the previous experiment based on laser probing only [14], the lifetime was determined to be 1.17 ± 0.05 . With the limited amount of data obtained so far the accuracy could not yet be further improved.

4. Discussion and conclusions

We have demonstrated the possibility to use a newly developed violet/blue diode laser for lifetime measurements

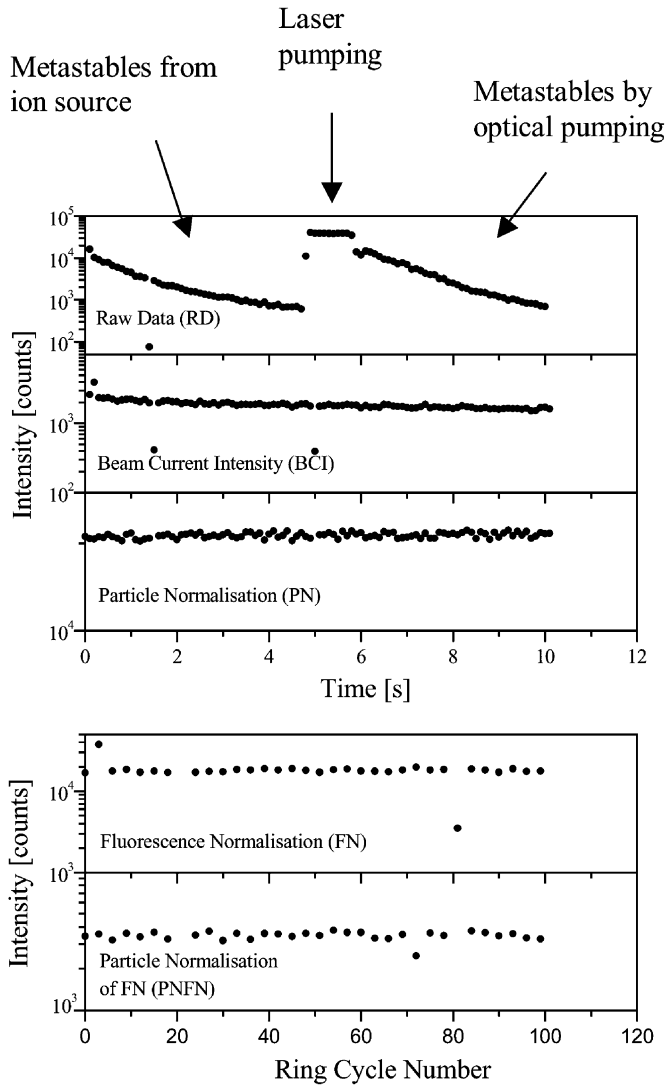


Fig. 7. Demonstration of the possibility to combine optical pumping and laser probing. It is clearly seen in the upper graph that the second lifetime curve, measured by laser probing after optical pumping, is “cleaner” (more “straight-lined” in logarithmic scale) than the first. The upper graph shows the population variation of the metastable levels. The next graph shows the number of stored ions as function of time. The three lower curves give the number of injected ions cycle-to-cycle as well as number of injected ions in the metastable levels. For stable conditions all these three curves should be “flat”.

at CRYRING. A new variation of lifetime measurements has been implemented, which combines OP with laser probing.

The main advantage with the new method is that if the metastable levels can be populated at a later time in the ring cycle, the transient beam loss phenomena have died

out [14,18]. This is clearly seen in Fig. 7, where the second decay curve (from 6 s and on) is straighter in the logarithmic scale. Obviously correction for instrumental beam loss will be of minor importance here.

Another advantage with OP is that the influence of repopulation [18] on the decay curve will be limited.

It seems likely that the accuracy could be pushed down to the 1% level, which would make it possible to give an answer to the question whether the lifetime difference between the $3d\ ^2D_{3/2}$ and the $3d\ ^2D_{5/2}$ level is larger than theory presently predicts.

Acknowledgements

This work was supported by grants from the Swedish Research Council. Excellent support from the staff of MSL is acknowledged.

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