The FERRUM project: an extremely long radiative lifetime in Ti II measured in an ion storage ring

Hartman, Henrik; Rostohar, D.; Derkatch, A.; Lundin, P.; Schef, P.; Johansson, Sveneric; Lundberg, Hans; Mannervik, S.; Norlin, L-O.; Royen, P.

Published in:
Journal of Physics B: Atomic, Molecular and Optical Physics

DOI:
10.1088/0953-4075/36/13/101

2003

The FERRUM project: an extremely long radiative lifetime in Ti II measured in an ion storage ring

2003 J. Phys. B: At. Mol. Opt. Phys. 36 L197

(http://iopscience.iop.org/0953-4075/36/13/101)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS B: ATOMIC, MOLECULAR AND OPTICAL PHYSICS

J. Phys. B: At. Mol. Opt. Phys. **36** (2003) L197–L202 PII: S0953-4075(03)62170-X

LETTER TO THE EDITOR

The FERRUM project: an extremely long radiative lifetime in Ti II measured in an ion storage ring

H Hartman[1], D Rostohar[2], A Derkatch[2], P Lundin[2], P Schef[2], S Johansson[1], H Lundberg[3], S Mannervik[2], L-O Norlin[4] and P Royen[2]

[1] Atomic Astrophysics, Lund Observatory, Lund University, Box 43, SE-22100 Lund, Sweden
[2] Physics Department, Stockholm University, AlbaNova University Centre, SE-10691 Stockholm, Sweden
[3] Department of Physics, Lund Institute of Technology, Box 118, SE-22100 Lund, Sweden
[4] Physics Department, Royal Institute of Technology, AlbaNova University Centre, SE-10691 Stockholm, Sweden

Received 9 April 2003, in final form 9 May 2003
Published 11 June 2003
Online at stacks.iop.org/JPhysB/36/L197

Abstract
We have extended the laser probing technique at the CRYRING storage ring to measurement of the extremely long lifetime (28 s) of the metastable $3d^2(^3P)4s\ b\ ^4P_{5/2}$ level in Ti II. The result obtained demonstrates the power of this method for investigation of such long-lived levels. This is the first experimental lifetime investigation of metastable states in Ti II.

Forbidden lines play a key role in the interpretation of spectra of diluted astrophysical plasmas. Such plasmas are often stellar gas ejecta and therefore associated with a central star. The diluted stellar radiation transfers energy to the plasma through excitation and ionization processes in the atoms and ions present. The photoelectrons produced are also heating the plasma. In general, the temperature is low and the nebular emission lines observed are either recombination lines or collisionally excited forbidden lines. Forbidden lines are transitions from metastable states, i.e. transitions disobeying the selection rules for electric dipole radiation. Typical lifetimes of metastable states are of the order of 1 s and they are in general destroyed in laboratory plasmas through collisions. However, in astrophysical plasmas the density is so low that the probability for collisions is very small, and the states decay by magnetic dipole (M1) or electric quadrupole (E2) radiation. Thus, forbidden lines are used in astrophysics as an indicator of density, and there is a need for experimental atomic data for these transitions.

In an ongoing international project on the mysterious star Eta Carinae [1] the Hubble Space Telescope (HST) has been used to study spatially resolved gas condensations close to the star. These gas condensations, or blobs, show distinct emission line spectra dominated by forbidden Fe II lines, [Fe II]. In a recent work within the FERRUM project [2] we have for the first time determined experimental Einstein coefficients, A-values, for a number of

[Fe II] lines [3], which will be used in future analyses. The laboratory work combines lifetime measurements of metastable states [4] at the storage ring CRYRING in Stockholm (Sweden) with branching fractions determined from theoretical and astrophysical data.

The spectrum of one specific gas blob in Eta Carinae differs remarkably from all others observed, indicating a completely different origin and strange ionization conditions. For example, it offers the first discovery of forbidden lines for strontium, [Sr II], in any astrophysical plasma [5] and the detection of numerous [Ti II] lines. Most transition group elements appear as singly ionized atoms, but Fe II is unusually weak. The dominance of [Ti II] in this blob spectrum suggests the use of these lines for temperature and density diagnostics. As there are no theoretical or experimental data on radiative lifetimes and transition probabilities available for metastable states and forbidden transitions in Ti II, we have started measurements in the same way as reported for [Fe II] above.

A storage ring as a tool opens the possibility for lifetime investigations of metastable levels which have lifetimes longer than a few milliseconds. In principle the lifetime can be measured directly by observation of the forbidden transition in the stored ions. Detection efficiency will, however, be very low since the dimensions of the storage ring are large (the circumference of CRYRING is 51.6 m) and the slow radiative decay may happen anywhere in the ring. For lifetimes in the millisecond region such measurements are still possible and have been performed, for example, by Träbert and co-workers at the Heidelberg ring (TSR). Very recently they presented experimental lifetimes of metastable levels in highly ionized iron [6]. In these measurements good statistics were obtained, but it is clear that the resolution did not always permit selective studies of individual levels and composite decay curves were recorded with contributions from more than one level.

For lifetime measurements of complex atomic systems it is necessary to have a highly selective technique since such systems can have numerous closely spaced metastable levels. With the laser probing technique, which utilizes laser excitation of ions in a storage ring, such high selectivity is obtained. The technique is basically developed from the fast-ion-beam laser method. It uses the benefits of photoexcitation of ions by a narrow-linewidth laser and the kinetic compression [7] obtained by using fast-moving ions, which results in sub-Doppler resolution. This opens even the possibility of selective excitation of closely spaced states, e.g. hyperfine levels [8]. Laser probing also drastically improves the detection efficiency (by about four orders of magnitude compared to passive observation) and it can be applied to long lifetimes that cannot be studied by passive means. Recently the laser probing method was successfully applied to lifetime measurements of metastable levels in singly charged iron [3, 4] and in rare-earth elements [9]. The method has previously been explained in detail [10] and only a brief overview is given here. In this letter we present a successful extension of the laser probing method by reporting measurements of an extremely long lifetime of half a minute in Ti II.

Singly charged ions were produced by a low-voltage electron-impact discharge ion source loaded with $TiCl_4$. The ions were extracted from the source and accelerated to 40 keV. After selection of the Ti isotope of mass 48 amu, the ions were stored in the CRYRING storage ring without further acceleration. The beam current of stored ions was about 0.5 μA. Only a small proportion of all stored ions populate metastable levels. A laser pulse with matching wavelength excites the ions in a specific metastable state to a higher level, which decays by an allowed transition by emission of a photon. About 500 ms duration of the laser pulse was found to be long enough to pump out more than 90% of the ions from the metastable level. The number of ions in the specific metastable state is proportional to the number of emitted photons. Applying the laser pulse at different times after the injection of ions into CRYRING, the lifetime curve can be built point by point.

For the lifetime measurements it is important that the storage time of the ions is considerably longer than the radiative lifetime of the levels investigated. Neutralization, due to collisions with atoms and molecules of the rest gas, is the dominant loss process for singly charged ions at 40 keV. The neutralized ions (atoms) are not influenced by the magnetic field and will thus leave the ring and hit the BaF_2 detector mounted after one of the bending magnets [11]. By reading the detector continuously with a multi-channel scaler (MCS) card, information about the beam intensity decay is obtained. The beam lifetime (i.e. when the initial intensity has dropped by a factor of $1/e$) at base pressure was estimated to be 60 s. This lifetime reflects the neutralization of ions in the ground state. The present experiment is, however, focused on the lifetime of specific metastable levels. The contribution from collisional destruction in the decay of these levels is selectively studied as described below.

Besides a long storage time, the stability in the production of the metastable ions as well as the total number of stored ions delivered by the ion source are of crucial importance since the different points on the lifetime curve correspond to different ion injections into the ring. Special normalization procedures have been developed that permit normalization for such variations [10]. Such corrections are usually small or negligible.

The number of ions in the metastable level decreases with time due to radiative decay, but ions in the metastable level may also be lost because of neutralization and de-excitation (the 'Penning effect') in collisions with the residual gas. At the ring base pressure (below 8×10^{-12} mbar) the collisional destruction rate is generally low, but for very long radiative lifetimes it may contribute significantly. The collisional destruction rate has to be subtracted from the measured decay rate in order to obtain the radiative transition rate and the radiative lifetime. It can be determined by measuring the lifetime as a function of pressure. The interaction between the stored ions and the residual gas will also cause excitation processes, which are specifically studied by performing so-called repopulation measurements. In this case two laser pulses are applied in every ring cycle. Firstly, immediately after the ion injection a prompt laser pulse is applied, which will quench all metastable ions delivered by the ion source. Secondly, a subsequent laser pulse is emitted at variable delay time after the prompt pulse in order to probe the metastable ions produced by collisional excitation [10, 11].

Monochromatic laser light was provided by a tunable ring dye laser (Coherent 699-29, Autoscan) operated with Stilbene 3 and pumped by an argon ion laser (Innova 400-25). The cw laser light was transported and introduced into the ring by a system of mirrors and merged with the ion beam collinearly. A beam expander was used for focusing. Laser pulses of suitable duration were produced with a mechanical shutter. To improve the detection efficiency of the fluorescence signal an additional acceleration of the ions by a Doppler tuning device [11] was performed in order to localize the region of laser excitation and the prompt fluorescence signal in front of a photomultiplier. The photomultiplier was equipped with a UG11 glass filter to decrease detector background; the glass filter was chosen to be transparent for the fluorescence light while the laser light was blocked. A special program controls the laser pulses of suitable duration and different delays for subsequent injections as well as the recording of the signals from the detectors. Sufficient data were collected (typically a running time of a few hours) to obtain reasonable statistics for the determination of the lifetime.

The lifetime of the $3d^2(^3P)4s\ b\ ^4P_{5/2}$ level turned out to be much longer than any other lifetime measured with the laser probing method at CRYRING (previously the longest lifetime (5.2 s) was measured in La^+ [12]). Thus, the analysis of the data recorded for this level required special caution and it was not obvious whether it would be possible to correct for the competing excitation and de-excitation mechanisms contributing to the measured lifetime of this long-lived state. The measured repopulation of the level was significant and subtraction of the repopulation curve from the decay curve changed the lifetime by more than 30% (see figure 1).

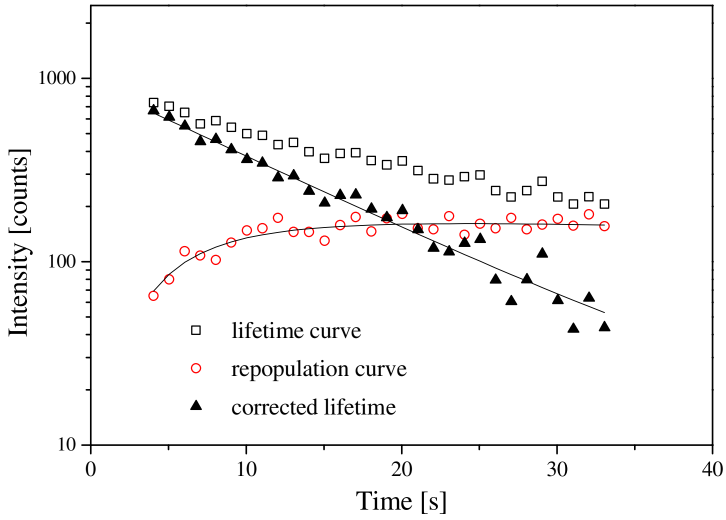

Figure 1. Fluorescence decay and repopulation curves for the $3d^2(^3P)4s\ b\ ^4P_{5/2}$ level in Ti II. The final lifetime curve ('corrected lifetime') obtained after subtraction of the repopulation is also shown. To average the statistical scatter, the repopulation curve was fitted by a double-exponential function ($f(t) = a_1 e^{-a_2 t} - a_3 e^{-a_4 t} + a_5$) before subtraction. The fitted functions for the repopulation and the final decay curve are included in the figure.

Since the repopulation curve was recorded separately, a serious systematic error will appear if the repopulation curve is not properly normalized to the lifetime curve. In a previous work on lifetimes of the metastable levels in Ca II the measurements were performed during storage times that were eight times longer than the radiative lifetime of the metastable levels [13]. In that case the population reached equilibrium through the two balancing effects, decay and repopulation. However, for very long radiative lifetimes, as in the present case, it is not possible to follow the decay until equilibrium is reached, and consequently the normalization procedure is delicate. In an alternative measurement procedure [14], however, it was possible to record the tails of the repopulation and lifetime curves in the same measurement. This novel procedure provided a reliable normalization, without which no conclusive result could have been obtained. The final error estimation includes the sensitivity for this normalization.

In order to estimate the collisional destruction rate, which is pressure dependent, the lifetime measurements were made at four different pressures (base pressure; five-, seven- and ten-times-increased pressure; see figure 2). The pressure was changed locally by heating one of the NEG (non-evaporative getter) pumps. The average increase of the pressure cannot be measured by vacuum gauges but it can be obtained by comparing the slope of the ion beam lifetime curves, which is proportional to the pressure. At higher pressure the repopulation will also increase. The fitted repopulation curves were subtracted from the lifetime curves and the resulting curves were fitted further by a single-exponential function. From the decay rates plotted versus relative pressure (the Stern–Vollmer plot) the contribution from collisional destruction was estimated to be 44% at base pressure (see figure 3). After extrapolation to zero pressure we obtained a value of 28 ± 10 s for the radiative lifetime of the metastable $3d^2(^3P)4s\ b\ ^4P_{5/2}$ level in Ti II. The large uncertainty is mainly due to the correction for repopulation as well as estimates of the relative pressure. The magnitude of the error bars was deduced from the sensitivity of the final value on the corrections.

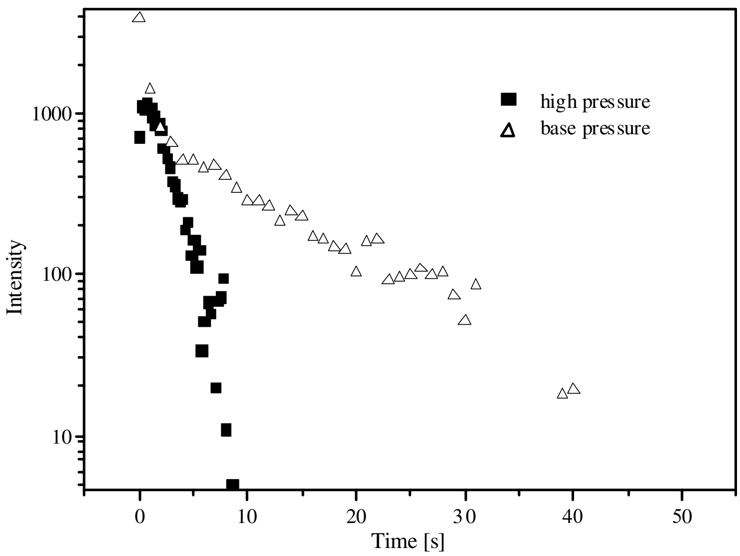

Figure 2. Decay curves obtained at base and ten-times-increased pressure in the storage ring (after correction for repopulation). The decay rate is drastically changed due to pressure effects (collisional quenching and repopulation).

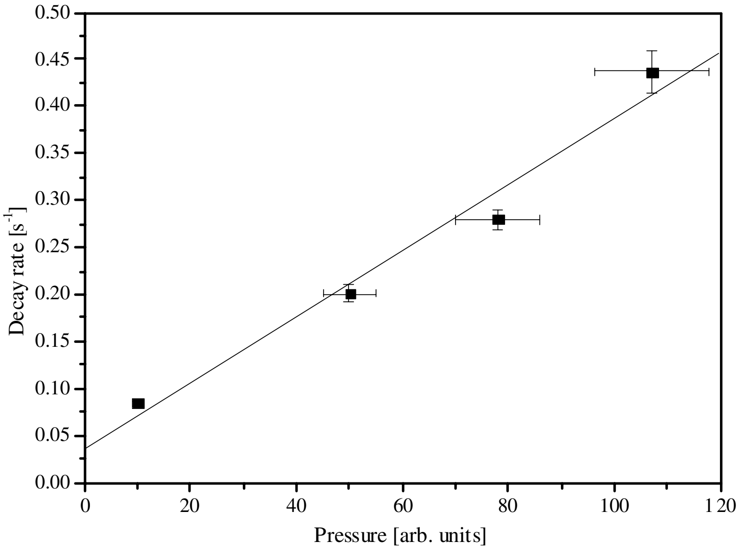

Figure 3. A Stern–Vollmer plot showing the measured decay rate versus the pressure in the storage ring. The pressure given is an average pressure in arbitrary units with the base pressure set at 10 (the absolute pressure being about 10 pTorr). The intercept of the fitted line at zero pressure is the decay rate (0.036 s^{-1}) without contributions from collisional destruction, i.e. the radiative decay rate. This intercept is independent of the scaling of the abscissa.

More lifetime measurements on Ti II are in progress, which will be reported in a coming article. Since there are no theoretical data on forbidden lines in Ti II we have also performed calculations of transition probabilities for both E2 and M1 transitions using the Cowan code package [15]. The E2 transitions were in most cases stronger than the M1 transitions. Theoretical lifetimes are derived from the transition probabilities. Preliminary results predict a lifetime of 12 s for the $3d^2(^3P)4s$ b $^4P_{5/2}$ level to be compared with the new experimental value of 28 s. The forthcoming paper will also include an analysis of the [Ti II] lines appearing in the peculiar blob in Eta Carinae mentioned in the introduction.

In the present work we show that the range of lifetimes that can be studied with the laser probing technique in an ion storage ring is further extended and covers 5 ms to 30 s. That range covers the lifetimes of a large fraction of metastable states in complex spectra. It has previously been demonstrated that the inherent spectral resolution permits investigations of ions with complex structure. In the present experiment the radiative lifetime of the $3d^2(^3P)4s$ b $^4P_{5/2}$ level has been determined as 28 ± 10 s. To the best of our knowledge this is the first lifetime measurement of a metastable level in Ti II.

We are grateful to the staff of the CRYRING facility and appreciate their efforts to provide an intense and stable ion beam. This work is supported by grants (SJ, SM) from the Swedish Research Council (VR).

References

[1] Gull T, Johansson S and Davidson K 2001 *Eta Carinae and Other Mysterious Stars: the Hidden Opportunities of Emission Line Spectroscopy (ASP Conf. Ser. 242)* (Chelsea, MI: Sheridan Books)

[2] Sikström C M, Schultz-Johanning M, Kock M, Li Z-S, Nilsson H, Johansson S, Lundberg H and Raassen A J J 1999 *J. Phys. B: At. Mol. Opt. Phys.* **32** 5687
Johansson S *et al* 2002 *Phys. Scr.* T **100** 71

[3] Hartman H *et al* 2003 *Astron. Astrophys.* **397** 1143

[4] Rostohar D, Derkatch A, Hartman H, Johansson S, Lundberg H, Mannervik S, Norlin L-O, Royen P and Schmitt A 2001 *Phys. Rev. Lett.* **86** 1466

[5] Zethson T, Gull T R, Hartman H, Johansson S, Davidson K and Ishibashi K 2001 *Astron. J.* **122** 322

[6] Träbert E, Calamai A G, Gwinner G G, Knystaustas E J, Pinnington E H and Wolf A 2003 *J. Phys. B: At. Mol. Opt. Phys.* **36** 1129

[7] Kaufman S L 1976 *Opt. Commun.* **17** 309

[8] Mannervik S, Broström L, Lidberg J, Norlin L-O and Royen P 1996 *Phys. Rev. Lett.* **76** 3675

[9] Rostohar D, Andersson K, Derkatch A, Hartman H, Mannervik S, Norlin L-O, Royen P, Schmitt A and Tordoir X 2001 *Phys. Scr.* **64** 1

[10] Mannervik S 2002 *Phys. Scr.* T **100** 81

[11] Lidberg J, Al-Khalili A, Norlin L-O, Royen P, Tordoir X and Mannervik S 1999 *Nucl. Instrum. Methods* B **152** 157

[12] Derkatch A, Ilynsky L, Mannervik S, Norlin L-O, Rostohar D, Royen P, Schef P and Biémont E 2002 *Phys. Rev.* A **65** 062508

[13] Lidberg J, Al-Khalili A, Norlin L-O, Royen P, Tordoir X and Mannervik S 1999 *J. Phys. B: At. Mol. Opt. Phys.* **32** 757

[14] Derkatch A, Mannervik S, Norlin L-O, Rostohar D, Royen P and Schef P 2003 at press

[15] Cowan R D 1981 *The Theory of Atomic Structure and Spectra* (Berkeley, CA: University of California Press)