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Lifetime Measurements of Metastable States in Fe⁺

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The lifetime of two metastable levels in Fe⁺ has been measured by laser probing of a stored ion beam. In the dense spectrum of Fe⁺, the metastable levels $a\ ^6S_{5/2}$ and $b\ ^4D_{7/2}$ were selected and their lifetimes were determined to be 230 ± 30 and 530 ± 30 ms, respectively. The lifetimes are compared with previous theoretical results. Metastable lifetime measurements of Fe⁺ are of great importance for interpretation of spectra from astronomical objects. The present experiment opens for the possibilities to investigate lifetimes of metastable states in complex atomic ions, which have, so far, been unexplored.

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The electronic structure of Fe⁺ (denoted Fe II in spectroscopic literature) is one of the most complex of an atom. The half-filled d shell in the M shell gives rise to a vast number of energy levels. The laboratory spectrum of iron contains thousands of lines and it has been quite a challenge for spectroscopists to interpret and classify the various spectral lines. One of the authors of the present article (S. J.) has recently completed identification and assignment of 1000 levels in Fe⁺ [1], and the present investigation concerns two of them.

At the same time, iron is one of the most important elements to study experimentally due to its high cosmic abundance. All stars that have come to a developed phase show in general very rich iron spectra. The spectral lines contain important information about the plasma conditions in the astronomical object provided that they can be identified and that relevant atomic parameters, such as the transition probability, are known. For astrophysical emission line regions, the spectrum is often crowded by parity-forbidden Fe II lines, denoted [Fe II], and the plasma diagnostics depends on the knowledge about the atomic physics behind these lines [2].

The high density of lines in emission spectra of the neutral Fe atom and its lowly charged ions have made experimental studies of lifetimes and transition probabilities difficult since highly selective measurements are required. Thus, only comparatively few transition probabilities have been measured. Recently, an international collaboration, the FERRUM project, has been started with the aim of extending the database for transition probabilities in iron [3]. The present experiment is the first step to widen this project to also include forbidden transitions. Within another international collaboration denoted the IRON project [4], a wealth of theoretical data has been produced for collisional and radiative processes.

In low charge states of iron the open $3d$ shell gives rise to a great number of energy levels in the complex of low configurations $(3d + 4s)^k$, and most of them are metastable. In singly charged iron, there are 62 such metastable levels with long lifetimes ($\tau > 100$ ms), as they are “forbidden” to decay by electric dipole radiation (since they have the same parity as all lower lying states). In addition, there are levels with opposite parity of the same configuration complex with lifetimes in the ms and μ s regime. Thus, they have low transition rates even if they obey the selection rules for $E1$ transitions and LS coupling. The general difficulty in measuring lifetimes of metastable states becomes even more complicated with a line-rich spectrum, and that has until now hindered metastable lifetimes to be measured in Fe⁺. Recently, a technique using laser probing in an ion storage ring was introduced [5]. This method has the selectivity and sensitivity required for measurements in dense and weakly populated energy structures. In this Letter, we report on the first successful lifetime measurements of two metastable levels in Fe⁺.

The experiment was performed in the ion storage ring CRYRING [6] in the Manne Siegbahn Laboratory in Stockholm. The laser probing technique has recently been described in detail [7–9]; the experiment will be discussed only briefly here. Singly charged ions of iron were produced in a low-voltage electron-impact discharge ion source loaded with FeCl₂. The ions were accelerated to a kinetic energy of 40 keV, sent through an isotope separator, and were finally injected into the ring. The ions were stored at injection energy and a beam current of 0.5–1 μ A was obtained (corresponding to about 10^9 stored ions). Apart from some initial faster decay (that will be discussed later), the decay of the ion beam current was caused by neutralization of the ion beam in collisions with the rest gas (of about 10^{-11} Torr), that gave a lifetime of about 30 s.

In the present experiment we measured the radiative lifetime of two metastable levels, $3d^54s^2a^6S_{5/2}$ and $3d^6(^3D)4s b^4D_{7/2}$, at excitation energies 2.9 and 3.9 eV, respectively (where a, b, \dots refer to the first, second, ... level of an LS symmetry). As in previous lifetime measurements utilizing the laser probing technique, we were relying on the production of ions in the metastable state directly in the ion source. In Ca^+ [7] and Sr^+ [9], that have only two metastable levels each, the fraction of metastable ions was estimated to be of the order of 1%. Considering the large number of metastable levels present in Fe^+ , only a much smaller population could be expected for each state.

The laser probing technique allows the population of a metastable state to be monitored. The ions are laser excited by an allowed transition from the metastable state to a higher level, and the fluorescence from the prompt decay of the upper level is observed. The excitation schemes for Fe^+ are shown in Fig. 1. One of the strengths of this tech-

nique is the *high detection efficiency*. All metastable ions can, by collinear excitation and local Doppler tuning [8], be probed in front of the photodetector. The spontaneous radiative decay by a forbidden transition is slow and will cause lifetimes in the millisecond to second range. Since the revolution time for 40 keV Fe^+ ions in the storage ring with 51.6 m circumference is 0.14 ms, such long lifetimes will distribute the photon emission from the decay over the whole ring. Consequently, with the use of only one photodetector, the detection efficiency will be improved by more than a factor of 1000 by laser probing. In addition, the laser probing permits electronic gating of the detector signal so that detector background contributes only during the laser pulse and is thereby reduced substantially (typically by a factor of 10–100). High efficiency and background reduction make the method very suitable for studies of weak signals. In addition, isotope separation before ion injection into the ring and high spectral resolution obtained by collinear laser excitation give the required *high selectivity*.

Laser light was provided by a cw ring dye laser (Coherent 699-29) operated with DCM. As expected, the laser-induced fluorescence resonances from Fe^+ were very weak, in the worst case about 10^{-3} of the signal obtained for Ca^+ [7] at the same ion beam current. In Fig. 1, laser-induced fluorescence for the two transitions is shown. At best, a signal of almost $10\times$ the background could be obtained for the $a^6S_{5/2}$ level.

With laser frequency set at resonance, the metastable lifetime could be measured by laser probing. With laser exposure for 100 ms most of the metastable ions were pumped out of the metastable level and a relative measure of the population was obtained by the detected laser-induced fluorescence. A mechanical shutter was used to create the laser pulse. The shutter was synchronized with the control system of the storage ring, giving the opportunity to apply the laser light at a variable delay after ion injection into the ring. By sequential variation of the delay time, decay curves could be recorded. Only one data point per injection will be obtained by this method, and it is therefore of great importance to monitor the injected ion beam intensity and to correct for variations between different injections if such occurs. For this purpose, various normalization curves were recorded for every decay curve [7–9]. In the present experiment the conditions were so stable that there was no need for correction.

We have previously observed repopulation [7–9] of metastable states by collisional excitation during the time when the ion beam is stored in the ring. For Fe^+ such effects were not observed. This is partly explained by the fact that the signals were weak and a tail in the decay curve from repopulation could be covered by detector background. Another explanation can be that repopulation will distribute the ions on many different metastable states, implying that repopulation for one specific state is weak. Consequently, no correction for repopulation was made in

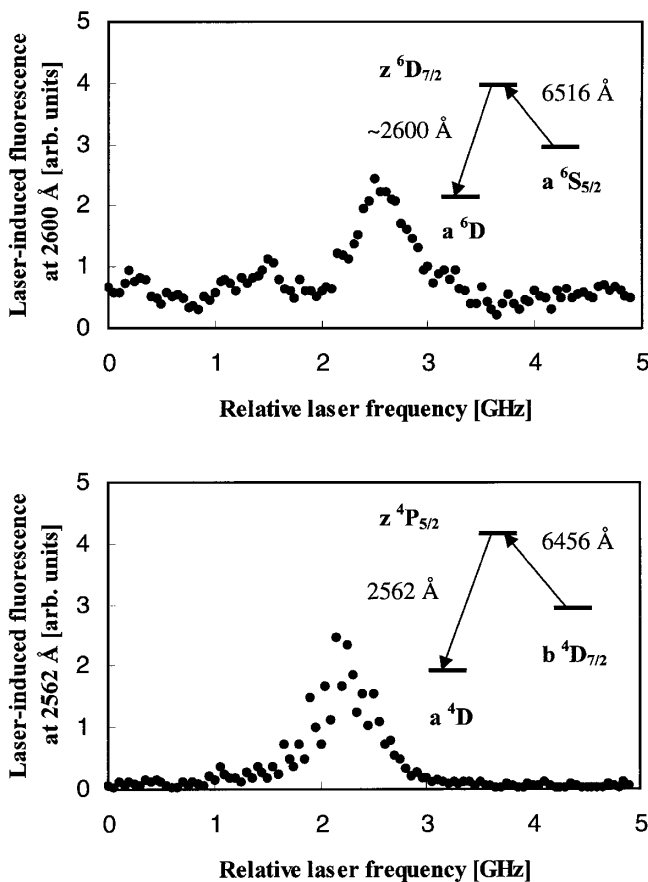


FIG. 1. Laser-induced fluorescence spectra obtained using the excitation schemes shown in the insets. The metastable $a^6S_{5/2}$ level was excited with laser light at 6516 Å and all the decay branches from the $z^6D_{7/2}$ to the fine structure levels of the a^6D term were simultaneously observed (at 2586, 2612, and 2631 Å). The $b^4D_{7/2}$ level was observed in a similar way as indicated in the figure. The larger scattering of the data points in the lower figure is due to another recording mode of the fluorescence than for the upper one.

the present experiment. Also collisional quenching of the metastable levels was weaker in Fe^+ than for Ca^+ [7] and Sr^+ [9]. This was investigated by raising the pressure in the ring in steps up to an average pressure of about $4\times$ the base level. The collisional effect was found to be small compared to the statistical error.

With the large number of ions injected into the ring, a minor nonexponential loss is present at the beginning of the ring cycle. This loss is presumably due to intrabeam scattering and beam optical effects, implying that the loss should be independent of the electronic state of Fe^+ . The ion beam intensity decrease was recorded simultaneously with the fluorescence decay. This curve was used to correct the lifetime curve of the metastable level for ion beam loss. The correction resulted typically in 10% longer lifetimes. This was the only correction of significance in the lifetime analysis.

For the weak decay curves obtained for the $a^6S_{5/2}$ level six data sets were recorded. From these curves a lifetime of 230 ± 30 ms was obtained. Only two curves were recorded for the stronger signal obtained from the $b^4D_{7/2}$ and the lifetime was determined to be 530 ± 30 ms. Examples of raw data are shown in Fig. 2.

Already in 1962 Garstang [10] had performed extensive calculations of probabilities for forbidden transitions in Fe^+ in order to support the interpretation of observations from peculiar stars. Garstang found that the $a^6S_{5/2}$ level mainly decays directly to the ground levels a^6D_J by electric quadrupole transitions ($E2$). Considering transition probabilities for all decay branches given in Garstang's calculation we derive a theoretical lifetime of 326 ms, which is more than 40% longer than our experimental value. For the $b^4D_{7/2}$ level, Garstang included decays to nine lower terms. The dominating decay branches were $E2$ transitions to a^4P_J (13%) and a^4F_J (62%) as well as $M1$ transitions to the ground levels a^6D_J (20%). The remaining 5% of the decay of the level is distributed over the levels of the

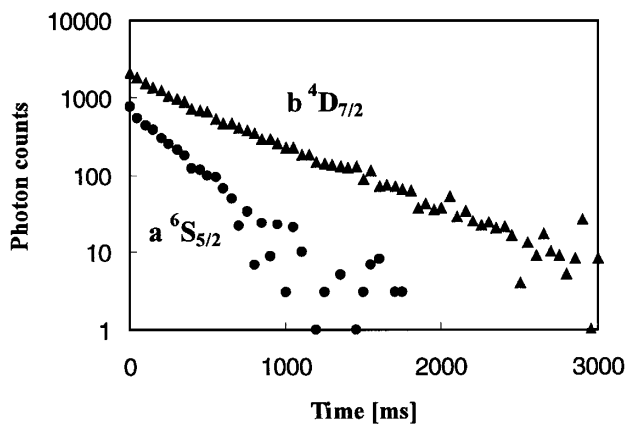


FIG. 2. Decay curves for the $a^6S_{5/2}$ and the $b^4D_{7/2}$ levels obtained by laser probing. Data collection time for these decay curves was about 7 hours each. Detector background has been subtracted before plotting.

other six terms (see Fig. 3). Here a radiative lifetime of 618 ms was obtained, which is 17% larger than the experimental result.

Nussbaumer *et al.* [11] have studied sextet transitions in Fe^+ . Their transition probabilities for $E2$ transitions between the $a^6S_{5/2}$ and the ground levels a^6D_J give a lifetime of 235 ms, in excellent agreement with our experimental result. Recently, within the IRON project, Quinet *et al.* [12] performed extensive calculations of transition probabilities of forbidden lines in Fe^+ . They have used two different methods for calculations, the SUPERSTRUCTURE (SST) code and relativistic Hartree-Fock (HFR). Both methods are generally in fairly good agreement with each other. For the $a^6S_{5/2}$ level we derive a lifetime of 262 ms (SST) and 220 ms (HFR) from the theoretical transition probabilities. For the $b^4D_{7/2}$ level they get 567 ms by SST and 500 ms by HFR. These new calculations are thus in good agreement with our experimental values. With the present level of experimental accuracy it is not possible to judge if one of the two methods is more accurate than the other. We also note that although the lifetime values derived from the data by Garstang [10] and Quinet [12] *et al.* are not in very large disagreement, there are very large differences in transition probabilities for some weak decay branches (in a few cases by a factor of more than 5).

As noted frequently in the literature, radiative decay rates and collisional excitation rates for metastable levels are of major importance when using astrophysical spectra to derive the electron temperature and density in an object. For Fe II, astronomers have had until now to rely on theoretical data for this analysis. The line-rich spectra and dense level structure of many iron group elements, that have obstructed lifetime measurements, also result not only in a significant configuration interaction but also in numerous cases of accidental level mixing, which are in general

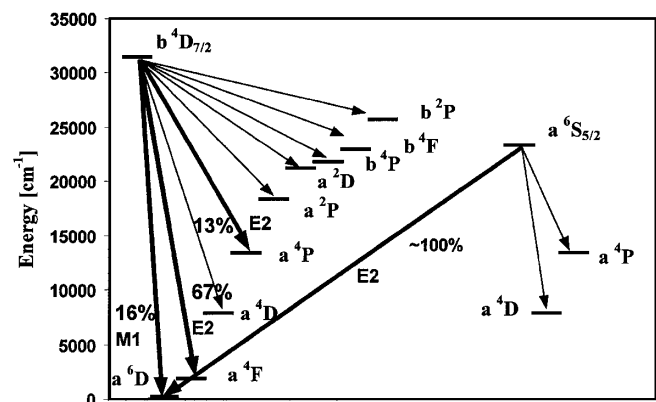


FIG. 3. Decay modes of the metastable $a^6S_{5/2}$ and $b^4D_{7/2}$ levels as included in the calculation of Garstang [9]. The more recent work by Quinet *et al.* [11] does not explicitly give all of the weakest decay branches. The branching fractions of the strongest channels as calculated by Quinet *et al.* are given in the figure. They deviate slightly from the results of Garstang (cf. the fractions mentioned in the text).

very difficult to predict theoretically. For the metastable levels, even a small mixing may cause large effects on the higher order radiative transition probabilities (i.e., for the forbidden transitions).

We conclude that the present experiment has proven that the laser probing technique has the selectivity and sensitivity to permit measurements of lifetimes of weakly populated metastable levels in dense spectra. The measurements performed here in Fe^+ are of great importance for astrophysical applications. We also find that the most recent calculations yield lifetimes in very good agreement with the experiment.

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