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Collisional excitation and destruction of metastable states in a stored ion beam

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A laser probing technique has been applied to a stored ion beam of Xe^+ to study collisional destruction of metastable states as well as collisional excitation by the residual gas in the storage ring. The results are of great importance for the evaluation of lifetime measurements of metastable states in storage rings. For the states studied in Xe^+, it was found that neither of the two processes caused systematic errors in the lifetime determination. It is also shown that the method can yield partial cross sections for collisional excitation. [S1050-2947(97)50808-0]

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In recent years ion storage rings have become an attractive tool for studies of atomic and molecular ions. Since the fast ion beam is stored for a long time, there is time both for manipulation of the ions and for observing and inducing weak processes. A number of review articles have appeared that describe advantages, technical progress, and experimental results (see, e.g., [1,2]). In general, the ion storage ring experiments permit isolation of specific processes under well-controlled conditions (regarding energies, isotopes, charge state, and initial quantum state). To achieve a long storage time very low residual gas pressure is required, since otherwise the ions will be lost by collisions. For the ion storage ring CRYRING in Stockholm where the present experiment was performed, the pressure is about 10 pTorr (H_2 equivalent) [3]. Even at this low pressure, however, collisions with the residual gas should be considered. The present investigation deals with such phenomena.

Lifetime measurements have recently been performed in ion storage rings for states both in positive [4,5] and negative atomic ions [6,7] as well as in molecular ions [8–10]. In these experiments the decay rates of specific states were studied. The basic problem here is how to monitor the specific state. Some of the experiments utilize the fact that the ion beam can only consist of one particular state. One example is the determination of the lifetime of the $2s^2 2p\,^4P$ state in Be^- [6]. In this case it is known that beryllium cannot bind an electron to the ground state and the $2s^2 2p\,^4P$ state is the only state that can be used to form an ion beam of Be^-. In the experiment performed by Balling *et al.* [6] the intensity decay of the stored ion beam of negative beryllium was monitored. This directly gave a measure of the lifetime of the metastable $2s^2 2p\,^4P$ state. If no quenching effects are present, the measured decay rate gives the natural lifetime of the state. There are in particular two quenching effects that have to be considered in detail: collisions with the rest gas in the storage ring and magnetic state mixing in the bending magnets. Both these effects were investigated by Balling *et al.* [6]. For positive ions there is always a stable ground state and the metastable states always constitute a minor fraction of the stored ion beam. Due to this fact and the fact that the energy spectra are more dense, methods for lifetime measurements require higher selectivity here. A specific problem is that loss of ions from the stored beam (mainly due to neutralization or stripping depending on the beam energy) might be strongly state dependent. Such an effect was clearly observed by Andersen *et al.* [11] for Fe^-. If the natural metastable lifetimes are comparable to the beam loss lifetime, such effects must be considered in detail.

Recently we have reported [12,13] on measurements of metastable lifetimes in the CRYRING by a laser probing technique. This method has many advantages, and in particular the strong selectivity permits studies of metastable states embedded in complicated level structures. As reported in [12,13], the method is even capable of resolving individual hyperfine states, which made it possible to observe a drastic hyperfine induced quenching for one particular hyperfine state of the $5d\ ^4D_{7/2}$ state in $^{129}Xe^+$. As concluded in the first paper [12], two effects have to be investigated in order to permit an accurate determination of the absolute value of the lifetime. First, influence from the magnetic field of the bending magnets (only certain states are sensitive to that) has to be studied. This was done in some more detail for $5d\ ^4D_{7/2}$ in [13]. Second, collisional destruction of the metastable state has to be investigated and, if needed, compensated for. The specific problem here is that this destruction could be quite different for the ground state compared to metastable ions. In a first preliminary experiment on the latter effects [13], we observed the following. The pressure in the ring was increased from 10 to 300 pTorr, approximately, by switching off the pumping and by heating the NEG pumps. A faster current intensity decay as well as a faster observed decay for the metastable state were consequently expected. For the current intensity the trend was in the expected direction, though the effect was only minor, but for the metastable state the trend was the opposite. This surprising observation implied that, instead of the expected increased destruction rate, a repopulation of the metastable state was caused by collisional excitation. Thus we were motivated to perform a separate investigation of this phenomenon, the result of which is reported in this Rapid Communication.

The experiment was performed at the ion storage ring CRYRING [14] at the Manne Siegbahn Laboratory (Stockholm). Singly charged xenon ions were extracted from an

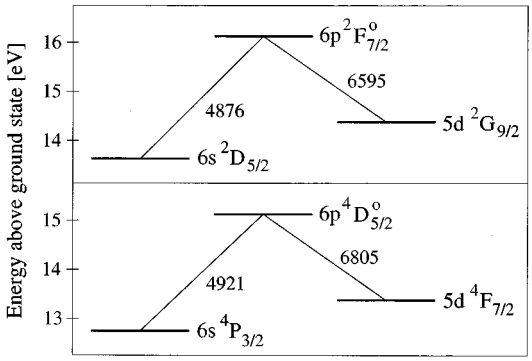

FIG. 1. Partial level diagrams showing the laser-induced transitions and corresponding fluorescence transitions utilized for the present experiment.

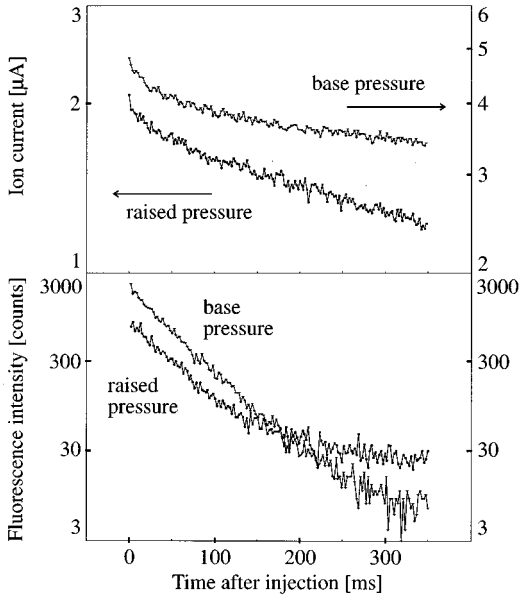

FIG. 2. Ion-beam current decay and metastable population decay for base pressure and raised pressure.

electron-impact ion source (MINIS) and accelerated to an energy of 40 keV. The xenon isotope $^{132}Xe^+$ was selected by a 90° bending magnet. About 10^{10} ions were injected and stored in the ring for each cycle. A small fraction of the beam (estimated to be in parts per thousand) consisted of ions in long-lived metastable states produced in the ion source. The metastable states had excitation energies between 12 and 14 eV. The circumference of the ring was 51.6 m, giving a revolution time of 0.22 ms for 40-keV Xe^+. The beam current was measured by a current transformer to give both the magnitude of the current and the intensity decay during the cycle.

The laser probing technique introduced for measurements of metastable lifetimes at ion storage rings has been described previously [12,13]. The technique utilizes the fact that the fluorescence intensity from a laser-induced transition from a metastable state is a measure of the metastable population. A cw ring dye laser pumped by an argon ion-laser was used to produce laser light in the desired wavelength region (650–680 nm), in order to excite the transitions $5d\ ^2G_{9/2}-6p\ ^2F^o_{7/2}$ and $5d\ ^4F_{7/2}-6p\ ^4D^o_{5/2}$; cf. Fig. 1 (the metastable states $5d\ ^2G_{9/2}$ and $5d\ ^4F_{7/2}$ were found not to be quenched by the magnetic mixing mentioned above). A photomultiplier observed the laser-induced fluorescence light through a colored glass filter, which blocked scattered laser light. Laser exposure of the stored ions was permitted by a mechanical shutter. An electronic circuit, triggered by the ion injection pulse of the CRYRING control system, could be adjusted to open the shutter at the desired delay time after ion injection and to keep the shutter open for a preset time interval.

The base pressure in the ring was 10 pTorr (H_2 equivalent) as measured by a coasting deuteron beam. About 90% of the residual gas consisted of hydrogen. At the low beam energy used for the xenon ion beam, stripping was negligible and the beam loss was expected to come from neutralization. Direct charge transfer from ground state H_2 to Xe^+ was expected to be weak [15], since that is an endoergic process, which also explains the tiny effect on the current intensity decay observed in the first test with vacuum variations mentioned above, since mainly H_2 contributed to the increase of the rest-gas pressure. Instead, the beam lifetime was primarily determined from resonant charge transfer by the $Xe^+ + Xe$ reaction. According to measurements by King and Latimer [15] the cross section for the latter process (about 2×10^{-15} cm^2) is at least ten times as large as the hydrogen reaction and the present experiment seems to indicate that the difference might be even larger. It was therefore assumed that the lifetime of the ion beam was mainly determined by the resonant $Xe^+ + Xe$ reaction. The xenon content in the residual gas, which was detected by a rest-gas analyzer when the ion beam was running, is due to the dumped xenon beam, which cannot be efficiently pumped. Thus the present investigation aimed at the study of both neutralization of metastable states and excitation of such states due to collisions in the rest gas.

The collisional destruction of the metastable states in the beam is primarily expected to be due to neutralization, even if Penning ionization is also possible. In order to compare the neutralization of the ground state to the destruction of the metastable state, xenon gas was leaked into the injection line of the beam and the ion pumps were switched off. The residual gas content was monitored by a rest-gas analyzer and the total pressure was measured by vacuum gauges around the ring. With xenon gas let into the ring, a faster intensity decrease of ion beam current was observed, as can be seen in Fig 2. It was noted that, with the pumps switched off, other residual gas components (such as CH_4 and Ar) also grew stronger (as they were released from the pumps), but the measurement was still conclusive. The lifetime curves for the metastable levels recorded simultaneously by the laser probing technique, were, however, *not affected*, except for a higher intensity in the tail where contributions from collisional repopulation were observed. This repopulation contribution was treated in detail and subtracted (see further discussion below). For a typical case, the slope of the current curve increased by a factor of 2.7, while no significant change could be observed in the lifetime curve of the metastable levels, as can be seen in Fig. 2. From the determination of the slopes and their uncertainties we conclude that the *cross section for neutralization of the metastable levels is*

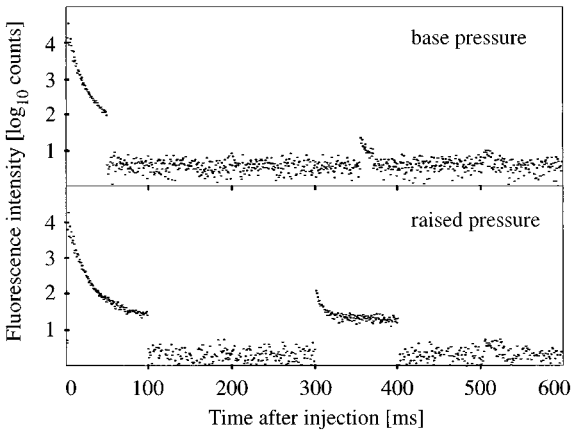

FIG. 3. Fluorescence intensity as a measure of the metastable population for base pressure and raised pressure. The first laser pulse quenched the initial metastable population and the second pulse revealed the collisonal repopulation. The very weak peak observed at 500 ms is due to a third laser pulse applied after the ion beam was dumped. This peak gives the background from scattered laser light.

significantly smaller than for the ground state. It should be noted that this result gives the *total* upper limit of metastable destruction, including both neutralization and collisonal deexcitation (i.e., Penning ionization of the rest gas). The results might be considered as natural, since the reaction between metastable xenon ions and neutral xenon is nonresonant, while the reaction between ground-state ions and neutral xenon is. Nevertheless, it is a very crucial observation as regards the possibility of performing accurate lifetime measurements. In our recent lifetime measurements we have now been able to determine millisecond lifetimes with about 2% accuracy [16], since this potential source of systematic errors could be ruled out.

To study the collisional repopulation a special method was introduced. For investigation of the weak repopulation process we wanted to avoid the metastables produced initially in the ion source. These states could be quenched by a laser pulse immediately after injection into the ring. With an ion beam of about 40 mm in diameter (with a velocity spread causing a Doppler width of about 500 MHz) and a laser beam with 10-mm diameter and 100–200-mW cw power, it was found that a laser pulse length of 100 ms was needed to quench the metastables by a factor of 1000 (Fig 3). The ions were then left circulating without interaction with the laser beam, but only with the residual gas. At a late part of the ring cycle the shutter was opened again and the actual metastable population could be detected. As can be seen in Fig. 3, even at the best vacuum a small repopulation is observed. In this run the quench pulse was only on for 50 ms, giving a quench factor of about 200, but with our measured lifetime (57 ms) [16] of this particular metastable level ($5d\ ^4F_{7/2}$), its population should have been reduced further by a factor of 200, due to spontaneous decay during the time until the second laser pulse was applied. The signal observed when the laser light is applied after 350 ms is, however, substantially stronger. Measurements were repeated when the pressure was raised. Then a significant repopulation was observed. This also confirmed the conclusion that the metastable lifetime curves recorded at ''high'' pressure were distorted by

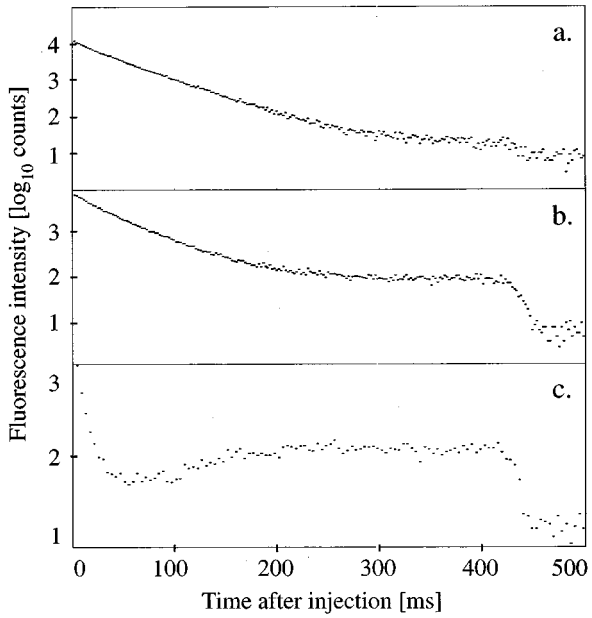

FIG. 4. (a) Lifetime curve for the $^2G_{9/2}$ level of Xe II recorded at base pressure. (b) Lifetime curve for the same level recorded at raised pressure. The high level in the tail is due to collisional repopulation. (c) A 50-ms laser pulse is applied immediately after injection of ions into the ring for every ring cycle. A second probe pulse of 20 ms is applied at a variable time after injection in order to monitor the repopulation process.

collision-induced repopulation of the metastable state.

The method used for lifetime measurements (i.e., a measurement of laser-induced fluorescence as a function of delay time) could also be utilized to monitor the time dependence of the repopulation process. A prompt laser pulse quenched the initial metastable population and then a new probe pulse in the same ring cycle was applied at a variable delay time. As can be seen in Fig. 4, the collisional excitation repopulates the metastable level up to a specific population, where a balance between repopulation and spontaneous decay occurs. This level depends on the ratio between the spontaneous decay rate and the excitation cross section. As concluded above, other destruction mechanisms will have negligible contributions compared to the spontaneous decay. Thus we can estimate the excitation cross section by the expression $\sigma=(A/vn)(N_e/N_g)$ (where A is the decay rate, v is the beam velocity, n is the residual gas density, N_e is the number of ions in the metastable excited state, and N_g is the number of ions in the ground state), which contains only measurable quantities. One of the main difficulties here is that the residual gas is a mixture of different molecules and atoms. Another is how to determine the detection efficiency of the metastables. A crude estimate of this efficiency is 0.1%. Using this value we find that the excitation cross section is between 10^{-19} and 10^{-18} cm^2.

Collisional excitations of vibrational and rotational states have been discussed in connection with molecular ion beams used for studies of dissociative recombination. In a paper by Forck *et al.* [17], the experimental results seemed to indicate collisional excitation of low-lying vibrational states. This kind of observation has, however, not been confirmed by more recent studies [18]. On the contrary, Heupel *et al.* have in a laser-based measurement on CH^+ [19] not been able to

see any trace of collisional excitation. The repopulation of metastable states observed in the present experiment on the xenon ion is a weak process. If similar effects were to be present in molecular ions, they would not affect the results of the dissociative recombination performed so far.

The present experiment deals with a fundamental issue regarding the use of ion storage rings for atomic and molecular physics, i.e., how clean are experiments using these facilities? Is the decay of metastable states in positive and negative ions as observed in storage rings primarily determined by spontaneous decay, or are there destructive processes occurring due to collision with the rest gas? The present results show selectively and quantitatively that such collisions are of minor importance for the states focused on here and that the storage rings possess the capacity to yield reliable results. The technique presented here also has the potential to be a powerful method for selective studies of partial cross sections with the use of an internal gas target, for which the target could be more well-defined in content and pressure. Such a device will be installed in CRYRING in the near future.

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