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Disentanglement of magnetic field mixing reveals the spontaneous *M*2 decay rate for a metastable level in Xe^+

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We have investigated the radiative decay of the metastable level $5d^4D_{7/2}$ in Xe^+. Theoretically we find the decay to be heavily dominated by an *M*2 transition and not by *M*1/*E*2 transitions. Lifetime measurements of $5d^4D_{7/2}$ in a storage ring are difficult since magnetic mixing of the metastable with a short-lived level quenches its population. Decay rates were determined at different magnetic field strengths (*B*) in order to allow a nonlinear extrapolation to $B=0$. The experimental lifetime of 2.4 ± 0.8 s was in agreement with the calculated value, but much smaller than previously estimated.

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It is well known that atomic ions may possess metastable excited levels that live very long. In singly charged ions the excitation energies are such that radiative transitions will appear in the optical wavelength region. Here the transitions will be completely dominated by electric dipole transitions (*E*1), while second-order effects (*M*1, *E*2) will be typically $10^{-5}-10^{-8}$ times smaller. This means that levels that are forbidden to decay by *E*1 transitions may have lifetimes of seconds or more due to the weakness of the higher-order contributions to the radiative decay. The longest lifetime that has been studied experimentally so far is a level in Yb^+ that has a lifetime of about 10 years [1,2]. For ions in metastable levels there are usually strong destructive processes that compete with the radiative decay. In particular, collisional quenching will be dominant or competitive unless vacuum is extremely good. Much experimental information on lifetimes of metastable levels has been gained in recent years by the use of storage devices such as ion traps and ion storage rings used in combination with lasers and supported by advances in vacuum technology.

Singly charged ions of noble gases possess metastable levels. The laser probing technique (LPT) developed at the ion storage ring CRYRING by our group was first applied to the metastable level $5d^4D_{7/2}$ in Xe^+ [3]. This level is prohibited to decay radiatively by an *E*1 transition to the ground level $^2P^o_{3/2}$ due to the selection rules for the angular momentum *J* (see Fig. 1). Its radiative decay is instead governed by higher multipole transitions. It was found that second-order terms could give rise to decay to the $6s^4P_{5/2}$ level that lies 0.3 eV below. Both *M*1 and *E*2 contribute to this decay and their total transition probability was found to correspond to a lifetime of 240 s [4]. The calculations of the present work give even smaller transition probabilities for these transitions (Table I).

In the original experiment an intriguing phenomenon was observed. When lifetime measurements were performed on the $5d^4D_{7/2}$ level in the odd xenon isotope $^{129}Xe^+$ that has hyperfine structure, it was found that the two different hyperfine states ($F=4$ and $F=3$) have drastically different lifetimes [3]. This effect could be explained by a hyperfine-induced mixing with the $5d^4D_{5/2}$ level that lies 5 meV below $^4D_{7/2}$. The hyperfine states with $F=3$ can mix and, since the radiative decay rate of $5d^4D_{5/2}$ is relatively high (about 10^7 s^{-1}), the $F=3$ sublevel of the $5d^4D_{7/2}$ level will "leak out" by this mixing—yielding a considerably shorter lifetime for the $F=3$ level than for $F=4$.

The even isotope $^{132}Xe^+$ has no nuclear spin and consequently no hyperfine interaction is present that can mix the levels. The ion beam, however, is confined in its circular

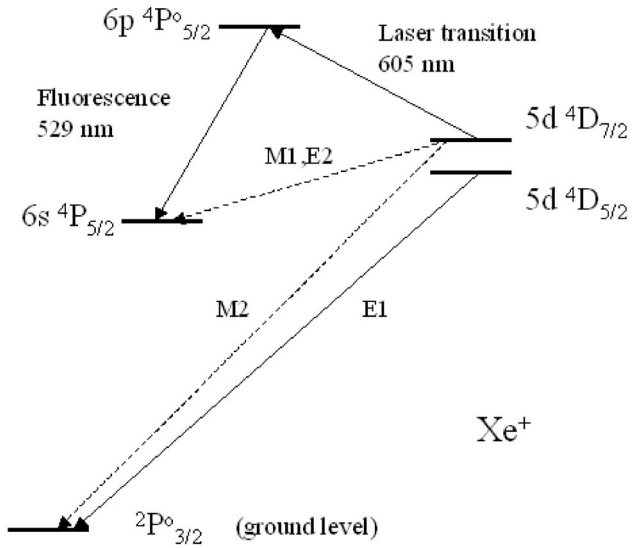

FIG. 1. Schematic energy-level diagram of Xe^+ showing the levels that are involved in the present experiment. The laser probing technique is applied by inducing the 605-nm transition and observing the fluorescence at 529 nm in order to study the decay of the metastable $5d\ ^4D_{7/2}$ level. The technique is described in detail in Refs. [7,8]. In the presence of a magnetic field the $5d\ ^4D_{5/2}$ is mixed with the $^4D_{7/2}$ level. This opens a new decay channel via an *E*1 transition.

TABLE I. Transitions involved in the decay of the $5s^25p^4(^3P_2)5d[3]_{7/2}$ level and the MCDF transition rates. The wavelengths and wave numbers are calculated values. The levels are denoted in *jK* coupling and the corresponding *LS* notation is given within parentheses.

Transition	Type	λ (nm)	σ (cm^{-1})	A[a] (s^{-1})
$5s^25p^4(^3P_2)5d[3]_{7/2}(^4D_{7/2})$-$5s^25p^5\ ^2P^o{}_{3/2}$	$M2$	109.0	91740	4.30(−1)
- $5s^25p^5\ ^2P^o{}_{3/2}$	$E3$	109.0	91740	3.05(−5)/2.87(−5)
- $5s^25p^5\ ^2P^o{}_{1/2}$	$E3$	122.5	81627	1.66(−5)/1.58(−5)
- $5s^25p^46s[2]_{5/2}(^4P_{5/2})$	$M1$	4468.3	2238	2.85(−3)
- $5s^25p^46s[2]_{5/2}(^4P_{5/2})$	$E2$	4468.3	2238	1.60(−4)/3.46(−4)
- $5s^25p^46s[2]_{3/2}(^2P_{3/2})$	$E2$	23419.2	427	6.12(−9)/1.85(−8)
- $5s^25p^4(^3P_2)5d[2]_{5/2}(^4D_{5/2})$	$M1$	90909.1	110	2.66(−5)
- $5s^25p^4(^3P_2)5d[2]_{5/2}(^4D_{5/2})$	$E2$	90909.1	110	1.39(−9)/5.32(−12)

[a]a(b) stands for $a\times10^b$. a(b)/c(d) means Babushkin value/Coulomb value.

orbit by magnetic fields. These will cause a Zeeman mixing between the $5d^4D_{5/2}$ and the $5d^4D_{7/2}$ levels that will quench the population of the latter level. In the original measurements the observed lifetime was about 1000 times shorter than the calculated value mentioned above [3].

In a recent experiment [5] on the homologous levels in Kr^+ we have measured the radiative lifetime of the $4d^4D_{7/2}$ level. The experimental result (0.57 s) was shorter than expected and our relativistic multiconfigurational Dirac-Fock (MCDF) calculations unexpectedly revealed that the third-order $M2$ transition strongly dominated over all other decay modes [5]. The situation for the $5d^4D_{7/2}$ level in Xe^+ is similar and we realized that the $M2$ decay channel in xenon may be strong as well. Thus we decided to perform a detailed MCDF calculation of the radiative decay of this level. Experimentally, the magnetic mixing contribution to the decay could be determined by lifetime measurements performed at different magnetic fields in the bending magnets of the storage ring.

The experiment was performed in the ion storage ring CRYRING [6] at the Manne Siegbahn Laboratory in Stockholm. Ions were produced in a discharge source. A certain fraction of the extracted ions were in the metastable level and this part was used for the lifetime measurement. Ions were accelerated to energies between 5 and 50 keV. Before injection into the ring, the beam was isotope separated and the $^{132}Xe^+$ isotope was used for the experiment. The lifetime was measured by the LPT (Fig. 1), which has been described in detail previously [7,8].

The magnetic field of the bending magnets in the ring is adjusted to close the orbit of ions of the particular energy to which they have been accelerated. The method to vary the magnetic field that is present during the lifetime measurement is consequently to use different beam energies. This was done by using different voltages on the injection platform—in this case from 5 to 50 kV.

The data collected were corrected for collisional destruction and repopulation [7–9]. For the lifetimes measured here, the corrections were relatively small ($<10\%$). The instrumental losses, however, required great caution. In order to disentangle the magnetic mixing effect, the measurements had to be performed at different beam energies (i.e., different magnetic fields). When we lowered the beam energy from the normal value of 40 keV, as was required for the present experiment, increasing problems arose as regards parameter settings, beam intensity, and diagnostics. Several measurements, including both the level decay by LPT and the particle loss as recorded by a particle detector connected to multichannel scaler (MCS), were performed. The magnitude of the correction for instrumental loss increased the lower the beam energy was and these corrections gave the dominating contribution to the uncertainty of the field-free radiative decay rate.

The levels in Xe^+ are in general better described in *jK* coupling than in *LS* coupling [10]. The levels discussed in this Communication are, however, strongly dominated by the LS term given here and we will continue to discuss the levels by these labels for simplicity, except for this part where the relativistic calculations are discussed in some detail. The *jK* notation for the metastable $5d^4D_{7/2}$ level is $5s^25p^4(^3P_2)5d[3]_{7/2}$.

The MCDF method [11] was used to compute the radiative lifetime of the $5s^25p^4(^3P_2)5d[3]_{7/2}$ level. In addition to the $M1$ and $E2$ channels, also decay to the ground state $5s^25p^5{}^2P^o{}_{3/2}$ via $M2$ and $E3$ transitions were calculated, since it was expected that, in particular, the $M2$ transition could be strong as it was for Kr^+ [5].

For the calculations we used the General-purpose Relativistic Atomic Structure Package (GRASP92) [12] that implements the MCDF method to model the lowest-energy levels of Xe^+. They were done in four steps. In the first step, we included only the two configuration state functions (CSF) of the $5s^25p^5$ configuration and performed an average level (AL) [12] optimization of the $1s$-$4d$ core, $5s$, and $5p$ orbitals. The second step consisted in considering 20 CSF belonging to the configurations $5s^25p^5$, $5s^25p^46s$ $J=3/2,5/2$, and $5s^25p^45d$ $J=5/2,7/2$. In the third step, a list of 1294 CSF was generated by single electron excitations from the above-mentioned configurations into the {$4f$, ns, np, nd, nf, $n=5$-6} active set of orbitals. In both steps, the new added orbitals were optimized minimizing the lowest six levels using the extended optimized level (EOL) option [12]. The final

step consisted of extending the CSF list of the third step to 14814 CSF by merging it with one generated similarly to as in the third step, but by double electron promotions keeping a filled $5s$. A relativistic configuration interaction (RCI) calculation [12] was eventually performed using the orbitals of the previous step. Transverse Breit and QED interactions have been included.

The eight transitions involved in the decay of the $5s^2 5p^4(^3P_2)5d[3]_{7/2}$ level are given in Table I, and, apparently, the dominant decay channel is the $M2$ transition. For the $E2$ contributions, both the Coulomb and Babushkin gauges were considered. The contribution of all the decay channels other than the $M2$ transition to this rate is less than 1% and they can be neglected in the estimation of the lifetime. We obtain a lifetime of 2.32 s using the theoretical energy differences. Inserting the experimental excitation energy of the $M2$ transition in the calculations would decrease the lifetime by 18%, leading to a corrected value of 1.91 s.

The stored ion beam spends a large part (about 15%) of the storage time in the magnetic field of the bending magnets. Here the field will mix the $^4D_{7/2}$ level with $^4D_{5/2}$ and thereby open an additional decay channel. When the field is raised from 0.1 to 0.3 T, the decay rate increases by a factor of 6 with a nonlinear dependence.

In the field region, we can express the mixed wave function Ψ_j as follows:

$$|\Psi_j\rangle = |^4D_{7/2}\rangle + \xi B|^4D_{5/2}\rangle. \tag{1}$$

B is the magnetic field and ξ is essentially the off-diagonal matrix element reflecting the field mixing of the two levels divided by the energy difference between the unperturbed levels ($\xi B \ll 1$). The transition probability is to a large extent determined by the matrix element of the operator for the radiative transition, here symbolically denoted $\boldsymbol{R}$, which can be expressed as a series expansion of multipoles,

$$A_{ik} \propto |\langle\Psi_i|R|\Psi_j\rangle|^2 \approx |\langle\Psi_i|E_1|\Psi_j\rangle + \langle\Psi_i|M1;E2|\Psi_j\rangle + \langle\Psi_i|M2;E3|\Psi_j\rangle + \ldots|^2, \tag{2}$$

where Ψ_j is given above in Eq. (2) and Ψ_i is $^2P^o{}_{3/2}$. From the selection rules and the results of the calculations given above, we see that there are mainly two contributions to the part of the transition probability given in Eq. (2),

$$\Rightarrow A_{ik} \propto |\xi B\langle^2P^o_{3/2}|E1|^4D_{5/2}\rangle + \langle^2P^o_{3/2}|M2|^4D_{7/2}\rangle|^2. \tag{3}$$

In the ion storage ring it is not possible to avoid the first term, since a confining magnetic field is always present. From the dependence of the decay rate on the magnetic field, an extrapolation to $B=0$ can be done in order to determine the decay rate at field-free conditions. The relation between the kinetic energy (E_{kin}) and the magnetic field (B) is

$$E_{\mathrm{kin}} = \frac{q^2r^2}{2m}B^2, \tag{4}$$

where q is the charge of the ion, r is the bending radius, and m is the ion mass. If the measured decay rates are plotted versus beam energy (Fig. 2), the dependence follows a linear trend. The slope is very steep and we see that the decay is for all measured points dominated by the field-induced decay.

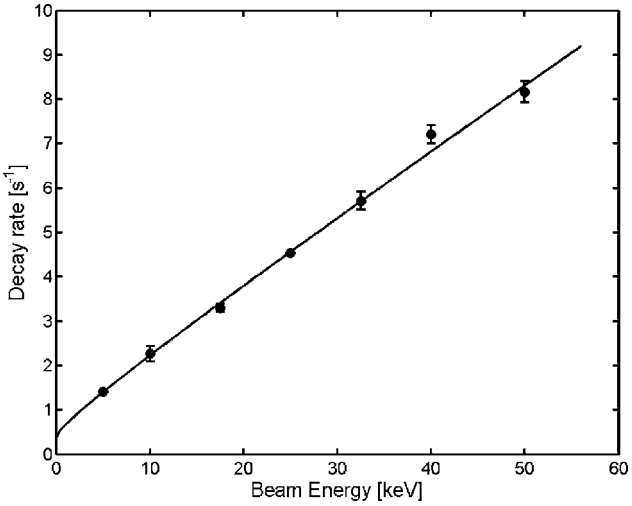

FIG. 2. Measured decay rate as a function of beam energy (proportional to B^2) and the fitted function that yields the radiative decay rate for field-free conditions as described in the text. Note the curvature of the fitted function at low beam energy.

From the linear behavior we understand that the term of Eq. (3), which is quadratic in B dominates in this region. For lower field strengths, the term that is linear in B [the cross term of Eq. (3)] will grow in importance and give rise to a deviation from the linear behavior in Fig. 2. As mentioned above, there are serious problems in storing the beam for low energies and we were not able to measure at lower energies than 5 keV. However, since the linear and the quadratic terms of Eq. (3) are linked it is still possible to determine the shape of the curve at low energies in order to extrapolate the decay rate to $B=0$.

Using the relation in Eqs. (3)–(5), we can express the radiative decay rate Γ as a function of ion-beam energy as follows:

$$\Gamma = \Gamma_{7/2} + k\left(2\zeta\sqrt{\Gamma_{7/2}E} + \zeta^2E\right)$$

$$\zeta \propto \xi\sqrt{\Gamma_{5/2}}, \tag{5}$$

where k is a fixed "efficiency" factor (i.e., the fraction of the beam path that is inside a magnetic field times the fraction of the magnetic sublevels that can mix). Γ_J are the natural radiative decay rates of the ^4D_J levels under field-free conditions. We see that the decay rate ($\Gamma_{7/2}$) of the $^4D_{7/2}$ level and the parameter ζ can be obtained from a nonlinear fit of the function in Eq. (5) to the data presented in Fig. 2.

The lowest data point is for $B \approx 0.1$ T. From the measured curve and the expressions above we find that the linear term (in B) does not become important until $B < 0.05$ T. The extrapolated experimental value for $B=0$ is a decay rate of 0.42 s^{-1} that corresponds to a radiative lifetime of 2.4 ± 0.8 s. The curvature at low B values and the lack of data points in this region cause the uncertainty to be large. We note that the agreement with the theoretical result is good. If the linear term in B incorrectly had been ignored and a linear function had been fitted to the points of Fig. 2, a much shorter lifetime had been extracted (1.6 s), and even more so if the lowest-

energy point had been neglected. The fitted value of the parameter ζ yields a value of $\Gamma_{5/2}$, which is consistent with the calculations.

Studies of metastable negative ions (Be^- [13] and He^- [14]) have been performed in the ion storage ring ASTRID in Aarhus. In these cases magnetic mixing also had to be considered, but the corrections that had to be introduced were small compared to what has to be done in the present work.

In conclusion, we have found that the radiative decay of the $^4D_{7/2}$ level in Xe^+ is determined to more than 99% by the $M2$ transition to ground level, similarly to the situation in Kr^+. The extremely strong magnetic-field-induced quenching observed in the present case is extraordinary. We have found that both the linear and quadratic terms of the field-induced decay had to be considered in order to determine the unperturbed radiative decay rate. This required a large set of data points for measured decay rates at different magnetic field strengths. Our experimental and theoretical results are in good agreement.

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