Towards a measurement of the $2s2p\ ^3P_0 \rightarrow 2s^2\ ^1S_0$
E1M1 two photon transition rate in Be-like xenon ions

D Bernhardt$^1$, C Brandau$^{1,2}$, C Kozuharov$^2$, A Müller$^1$, S Schippers$^1$, S Böhm$^1$, F Bosch$^2$, J Jacobi$^1$, S Kieslich$^1$, H Knopp$^1$, P H Mokler$^{1,3}$, F Nolden$^2$, W Shi$^1$, Z Stachura$^4$, M Steck$^2$ and T Stöhler$^{2,5}$

$^1$ Institut für Atom- und Molekülphysik, Justus-Liebig-Universität, 35392 Giessen, Germany
$^2$ GSI Helmholtzzentrum für Schwerionenforschung, 64291 Darmstadt, Germany
$^3$ Max-Planck-Institut für Kernphysik, 69117 Heidelberg, Germany
$^4$Instytut Fizyki Jądrowej, PL-31-342 Kraków, Poland
$^5$Physikalisches Institut, Universität Heidelberg, 69120 Heidelberg, Germany

E-mail: Dietrich.Bernhardt@iamp.physik.uni-giessen.de

Abstract. Dielectronic recombination (DR) of Be-like $^{136}$Xe$^{50+}$ was measured by employing the electron-ion merged-beam technique at the storage ring ESR of GSI in Darmstadt, Germany. We present results covering the center-of-mass energy range 0 – 4 eV. In addition to Rydberg series of resonances associated with ground-state intra L-shell excitations, resonances were found which are formed during DR of initially metastable $^{136}$Xe$^{50+}(2\ s^2\ 3\ P_0)$ parent ions. This state is the first excited state and is long lived because the J=0 $\rightarrow$ J=0 transition and the absence of nuclear spin make the rare E1M1 two-photon transition the lowest-order decay channel. Future measurements which will monitor the strength of the resonances as function of storage time may be exploited for a measurement of the E1M1 two photon transition rate.

Atoms and ions in metastable excited states with very small electromagnetic transition rates are promising systems for realizing ultrasprecise atomic clocks, for developing new diagnostic tools, for studying astrophysical media with respect to the competition of radiative and non-radiative processes, for realizing novel types of cold atomic gases, and for probing fundamental correlation effects in the bound states of few-electron systems. Atomic and ionic systems with two valence electrons in the $n^{10}$ principal shell have a $ns^2\ ^1S_0$ ground state. For $n = 2$ the first excited state above the ground level is the $2s2p\ ^3P_0$ state (figure 1). Due to zero total angular momentum in both levels a single-photon transition is forbidden except for the hyperfine-induced (HFI) decay that is possible if the atomic nucleus has non-zero nuclear spin [1]. Rates for the hyperfine-induced decay of $2s2p\ ^3P_0$ states in beryllium-like ions have been calculated [2–6] with state-of-the-art methods, and also first laboratory measurements [7] have been performed.

For ions with zero nuclear spin such as $^{136}$Xe$^{50+}$, the E1M1 two-photon transition is the lowest-order decay channel. In contrast to HFI transitions, there are no experimental results for E1M1 two-photon transitions in berylliumlike ions available yet [8]. So far, two-photon transitions have predominantly been studied in He-like ions [8, 9]. The only theoretical predictions of E1M1 $^3P_0 \rightarrow ^1S_0$ lifetimes $\tau_{E1M1}$ have been provided by Schmieder [10] for Z=12.
Figure 1. (color online) Simplified level diagram for berylliumlike $^{136}$Xe$^{50+}$. The $^{3}P_0 \rightarrow ^{1}S_0$ transition proceeds either via emission of two-photons (E1M1) or, in case of a nucleus with non-zero nuclear spin, decays within hyperfine-induced single-photon. For Xe$^{50+}$, all other depicted decay modes (E1: electric dipole transition; M2: magnetic quadrupole transition; IC: intercombination transition) have a much shorter lifetimes [13]. The level energies were taken from the work of Safronova et al [12]. The transition lifetimes are given by Cheng et al [4].

to 20 (figure 2) and by Laughlin [11], who presented his results in the form of a closed formula which can be written as:

$$\tau_{E1M1} = 3.1 \times 10^{18} \text{ s} \cdot Z^{-4} (E_{\gamma_1} + E_{\gamma_2})^{-5}$$  \hspace{1cm} (1)$$

Here $Z$ is the atomic number of the Be-like ion, and $E = E_{\gamma_1} + E_{\gamma_2}$ is the $^{3}P_0 \rightarrow ^{1}S_0$ transition energy shared by the two photons $\gamma_1$ and $\gamma_2$ given in eV. The theoretical E1M1-lifetimes obtained from equation 1 by using theoretical excitation energies $E$ [12] are shown by the (red) solid line in figure 2. For Xe$^{50+}$ one obtains a lifetime $\tau_{E1M1}$ of $\approx 30$ s. The predictions by Laughlin were made within a non-relativistic framework using LS-coupling which is not appropriate for heavy ions. In particular, equation 1 was derived under the assumption that the $2s2p \ ^{3}P_0$ and the $2s2p \ ^{3}P_1$ state are degenerate levels. Clearly, this assumption is inappropriate for Xe$^{50+}$ where the splitting ($E_{01}$) is $\approx 23$ eV [12] which is $\sim 20\%$ of the $2s2p \ ^{3}P_0$ excitation energy ($\approx 104$ eV) [12] (figure 1). Based on the formulas that were provided by Laughlin [11] we derived a more general formula for $\tau_{E1M1}$ which takes into account the splitting $E_{01}$, i.e.

$$\tau_{E1M1} = 5.2 \times 10^{17} \text{ s} \cdot Z^{-4} \left( \frac{2E_{01}^2 (3E^2 - 10EE_{01} + 10E_{01}^2) (E_{01} - E)^2 \log \left( \frac{E_{01}+E}{E_{01}-E} \right)}{2E_{01} - E} + \frac{1}{6} E \left( E^4 + 8E^3E_{01} - 68E^2E_{01}^2 + 120EE_{01}^3 - 60E_{01}^4 \right) \right)^{-1}$$  \hspace{1cm} (2)$$

It can be easily seen, that for $E_{01} \rightarrow 0$ equation 2 approaches equation 1. With the $2s2p \ ^{3}P_0$ and $2s2p \ ^{3}P_1$ level energies from the relativistic many-body–perturbation calculations of Safronova et al [12] equation 2 yields to the dashed curve given in figure 2. For $Z > 25$, this modification
increases the predicted lifetimes by factors ranging from 1.5 to 2.5. For Xe$^{50+}$ this leads to a lifetime prediction of 78 instead of 30 seconds. More reliable theoretical descriptions of E1M1 transitions in heavy Be-like systems, which take into account relativistic effects, are not available. For 2E1 two-photon transitions in helium-like ions from the excited $2^1S_0$ state to the $1^1S_0$ ground state the comparison of non-relativistic [14] and relativistic [15] treatments shows that for xenon relativistic 2E1 lifetimes are about 30% larger as non-relativistic 2E1 lifetimes [15].

To perform lifetime measurements at ion-storage rings the transition lifetime $\tau$ has to be longer than the time needed for ion beam preparation which is usually of the order of seconds. Moreover, $\tau$ should not significantly exceed the ion-beam storage lifetime $t_{\text{store}}$. The latter is affected by vacuum conditions including residual gas composition, by intra-beam scattering, and by additional ion loss processes due to collisions with the electron beam of the electron cooler. Some of the corresponding loss rates depend strongly on the ion velocity. In case of the heavy-ion storage ring ESR at GSI, which can store heavy ions with energies up to about 400 MeV/u, $t_{\text{store}}$ can be up to several hours [16]. The restrictions for lifetime measurements discussed above together with the available predictions for $^3P_0 \rightarrow 1^1S_0$ E1M1 transitions (figure 2) suggest a convenient time window for dielectronic recombination (DR) based decay studies at the ESR for ions with nuclear charge in the range $40 \lesssim Z \lesssim 70$.

DR is a resonant process, where the excitation of a bound electron by the incoming continuum electron is accompanied by the capture of this electron into a bound state $n_lj$. Thus, in recombination spectra, DR resonances are observed at discrete electron-ion collision energies. For an ion beam composed of two fractions such as the present case of $2s^2 \ 1^1S_0$ ground state ions and ions in the $2s2p \ 3P_0$ metastable state, the measured recombination spectrum is a superposition of two initial-state-specific spectra weighted with the fractional abundances of each component. In certain energy regions resonances from ground and metastable state are well separated. This allows to distinguish the two beam admixtures. Monitoring such resonances

---

**Figure 2.** (color online) Non-relativistic lifetime predictions for $1s^22s2p \ 3P_0 \rightarrow 1s^22s^2 \ 1S_0$ E1M1 two-photon transitions. Open circles represent calculations by Schmieder [10] for Z=12 to 20, the solid line results from equation (1) using LS-coupling according to Laughlin [11]. The dashed line shows the present modification of Laughlin’s calculations taking into account the splitting between the $^3P_0$ and $^3P_1$ energy levels [12]. For details see text.
Recombination rate coeff. (10^{-10} \text{ cm}^3 \text{ s}^{-1})

\text{Electron-ion collision energy (eV)}

\begin{align*}
\text{Xe}^{50+} 2s2p^3P_0 + e^- &\rightarrow \text{Xe}^{49+} 2p^23P_1 8_j \\
\text{Xe}^{50+} 2s^21S_0 + e^- &\rightarrow \text{Xe}^{49+} 2s2p^1P_1 8_j
\end{align*}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{recombination_spectrum.png}
\caption{(color online) Measured \textsuperscript{136}Xe\textsuperscript{50+} recombination spectrum. Resonances of metastable parent ions (dark shaded, red in the online version) appear on top of the groundstate parent ion resonances (white). The two sets of vertical bars indicate expected energies of DR resonances \text{Xe}^{50+}(2s2p^3P_0) + e^- \rightarrow \text{Xe}^{49+}(2p^23P_1 8_j) (upper red) and \text{Xe}^{50+}(2s^21S_0) + e^- \rightarrow \text{Xe}^{49+}(2s2p^1P_1 8_j) (lower blue). The expected energies were calculated by using core-excitation energies \cite{12} and Dirac binding energies (equation \ref{eq:dirac_binding}). In the displayed energy range the experimental energy spread is \sim100 \text{ meV}.}
\end{figure}

in the appropriate energy region allows one to perform sensitive lifetime measurements for the minor beam component even if its fraction is only on the percent level \cite{7}.

In the present experiment, DR of berylliumlike \textsuperscript{136}Xe\textsuperscript{50+} was investigated using the ESR cooling device alternatingly as an electron cooler to prepare a cooled ion beam and as a free-electron target \cite{18}. In addition to \text{Xe}^{50+} ions in the ground state, a fraction of the beam was in the \text{2s2p}^3P_0 metastable state populated during beam preparation in the stripper target passed by the accelerated xenon ions. The resulting metastable \text{Xe}^{50+} beam component is sufficiently long-lived to survive the initial cooling in the ESR. Figure 3 shows the measured DR spectrum of \textsuperscript{136}Xe\textsuperscript{50+} which is dominated by resonances from ions that are initially in the \text{2s}^21S_0 ground state. In addition, four resonances associated with \text{Xe}^{50+}(2s2p^3P_0) + e^- \rightarrow \text{Xe}^{49+}(2p^23P_1 8_j) dielectronic capture into states with \textit{j}= 9/2, 11/2, 13/2 and 15/2 are clearly visible between 1.15 and 2.0 eV. Additional resonances with \textit{j}=5/2 and 7/2 are expected at 0.08 and 0.8 eV. However these were not observed since the much stronger ground state resonances dominate the DR spectrum in the 0 – 1.1 eV energy range. The expected resonance energies \textit{E}, shown by vertical bars in figure 3, were calculated by using core-excitation energies \textit{E}_{\text{ex}} of Safronova \textit{et al} \cite{12} and hydrogen-like Dirac binding energies \textit{E}_B(\textit{n}, \textit{j}) for the 8\textsubscript{j} Rydberg electron \cite{19} as follows:

\textit{E} = \textit{E}_{\text{ex}} - \textit{E}_B(\textit{n}, \textit{j}) = \textit{E}_{\text{ex}} - m_e c^2 \cdot \left( 1 + \frac{Z \cdot \alpha}{n - (j + \frac{1}{2}) + \sqrt{(j + \frac{1}{2})^2 - (Z \alpha)^2}} \right)^{-1/2} + m_e c^2 \quad (3)

The calculated energies coincide roughly with measured resonance positions. Thus, we conclude that DR resonances associated with the initially metastable \textsuperscript{136}Xe\textsuperscript{50+}(2s2p^3P_0) ions have unambiguously observed.
For the near future, we envisage an ESR experiment using such resonances for a precise measurement of the lifetime of the $2s2p^3 P_0$ metastable state in Be-like ions. Techniques will be used similar to those developed and successfully employed at the Heidelberg storage ring TSR for the lifetime determination of the $1s2s^2 S_1$ states in the helium-like ions $C^{4+}$ [22] and $Li^+$ [23] and, more recently, for the measurement of the hyperfine-induced $3P_0 \rightarrow 1S_0$ transition in Be-like $^{47}Ti^{18+}$ [7]. Based on the available predictions for the E1M1 transition rate in Be-like ions (equation 2) we expect that accurate measurements of two-photon transition rates in heavy Be-like ions at the ESR should be feasible in the range $Z \approx 40$ to $Z \approx 70$. A more reliable theoretical description of E1M1 transitions in heavy Be-like systems, which take into account, e.g., j-j-coupling and relativistic effects would be very helpful for identifying the most suitable ions.

The authors thank S. Fritzsche, F. Fratini, G. Gribakin, Z. Harman, V. P. Shevelko, and A. Surzhykov for fruitful discussions and gratefully acknowledge the excellent support by the GSI accelerator and the ESR crew. This work was supported in part by the German reearch-funding agency DFG under contract number SCHI378/8-1 and by the German federal research ministry (BMBF) under contract number 06GI9111.

References

[1] Johnson W 2011 Can. J. Phys. 89 429–437
[2] Marques J P, Parente F and Indelicato P 1993 Phys. Rev. A 47 929–935
[3] Brage T, Judge P G, Aboussaid A, Godefroid M R, Jönnson P, Ynnerman A, Fischer C F and Leckrone D S 1998 Astrophys. J. 500 507
[4] Cheng K T, Chen H H and Johnson W R 2008 Phys. Rev. A 77 052504
[5] Andersson M, Zou Y, Hutton R and Brage T 2009 Phys. Rev. A 79 032501
[6] Li J and Dong C 2010 Plasma Sci. Technol. 12 364
[7] Schippers S, Schmidt E W, Bernhardt D, Yu D, Müller A, Lestinsky M, Orlov D A, Grieser M, Repnow R and Wolf A 2007 Phys. Rev. Lett. 98 033001
[8] Mokler P H and Dunford R W 2004 Phys. Scr. 69 C1
[9] Trotsenko S, Kumar A, Volotka A V, Banaś D, Beyer H F, Bräuning H, Fritzsche S, Gumberidze A, Hagmann S, Hess S, Jagodziński P, Kozhuharov C, Reuschl R, Salem S, Simon A, Spillmann U, Trassinelli M, Tribedi L C, Weber G, Winters D and Stöhlker T 2010 Phys. Rev. Lett. 104 033001
[10] Schmiede R W 1973 Phys. Lett. A 7 145–1468
[11] Laughlin C 1980 Phys. Lett. A 75 199 − 200
[12] Safonova M S, Johnson W R and Safonova U I 1996 Phys. Rev. A 53 4036–4053
[13] Ralchenko Y V and Vainshtein L A 1995 Adv. At. Mol. Phys. 29 147–154
[14] Brandau C, Kozhuharov C, Müller A, Shi W, Schippers S, Bartsch T, Böhm S, Bömje C, Hoffknecht A, Knopp H, Grün N, Scheid W, Steih T, Bosch F, Franke A, Mokler P H, Nolden F, Steck M, Stöhlker T and Stachura Z 2003 Phys. Rev. Lett. 91 073202
[15] Hoffknecht A, Brandau C, Bartsch T, Böhm S, Knopp H, Schippers S, Müller A, Kozhuharov C, Beckert K, Bosch F, Franke A, Krämer A, Mokler P H, Steck M, Stöhlker T and Stachura Z 2001 Phys. Rev. A 63 012502
[16] Shi W, Böhm S, Bömje C, Brandau C, Hoffknecht A, Kieslich S, Schippers S, Müller A, Kozhuharov C, Bosch F, Franke A, Mokler P H, Steck M, Stöhlker T and Stachura Z 2001 Eur. Phys. J. D 15 145–154
[17] Schmidt H T, Forck P, Grieser M, Habs D, Kenntner J, Miersch G, Repnow R, Schramm U, Schüssler T, Schwalm D and Wolf A 1994 Phys. Rev. Lett. 72 1616–1619
[18] Saghir A A, Linkemann J, Schmitt M, Schwalm D, Wolf A, Bartsch T, Hoffknecht A, Müller A, Graham W G, Price A D, Badnell N R, Gorczyca T W and Tanis J A 1999 Phys. Rev. A 60 R3350–R3352