Photorecombination studies of highly charged ions at the storage ring ESR: a progress report

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Abstract. In this contribution, we report on photorecombination measurements of highly charged ions (HCl) that were performed using the electron cooler of the heavy-ion storage ring ESR of the Gesellschaft für Schwerionenforschung (GSI) as a target of free electrons. The experimental technique of resonance reaction spectroscopy by means of dielectronic recombination (DR) was employed in two new ways:

The determination of isotope shifts in the DR resonance spectrum of few-electron heavy ions is a novel method with unique properties for investigations at the interface between electrons and the atomic nucleus, e.g., charge radii and hyperfine effects. First results from a pilot experiment with the two isotopes A=142 and A=150 of Li-like neodymium \(^{57}\text{Nd}^{5+}\) are discussed.

Until very recently, electron-ion collision experiments at high relative energies of more than a few keV were not accessible at the electron coolers of storage rings. Such high energies are needed to access DR processes that involve the K-shell electrons of very heavy ions. The novel approach of DR measurements with a stochastically (pre-)cooled ion beam resolves this problem. Results for the photorecombination of hydrogen-like \(^{91}\text{U}^{5+}\) in the energy range (63 keV - 74 keV) of the KLL-DR process are presented.

1. Introduction

The investigation of photorecombination (PR) of heavy highly charged ions (HCl) with free electrons at ion storage rings has experienced a remarkable development since the first experiments about 25 years ago [1, 2]. In particular, resonance reaction spectroscopy (RRS) by means of dielectronic recombination (DR), i.e. the high-resolution cross section measurement of the resonant channel of PR, has been developed into a versatile ‘tool’ for studying atomic structure and recombination dynamics. Among others, the list of RRS applications comprises such diverse items as precision studies of quantum electrodynamics (QED) in strong fields [3, 4], tests of fully relativistic collision theories [5, 6], the influence of external fields on the recombination process [7, 8], investigations of the interface between the atomic electrons and the nucleus [9, 10], or
the provision of absolute recombination rate coefficients relevant for astrophysics [11, 12]. The technique of RRS is particularly appealing for very heavy few-electron ions, as the experimental resolution is mainly determined by the ‘quality’ of the two beams, i.e. the temperatures $kT_\perp$ and $kT_{||}$ of the electron beam and the momentum spread $dp/p$ of the ion beam. It is thus independent of the nuclear charge $Z$ for narrow natural linewidths. QED and relativistic effects as well as contributions from the finite size of the atomic nucleus scale with large powers of $Z$. Presently, the ESR of GSI is the only storage ring that provides access to the fascinating field of RRS studies with the heaviest few-electron ions. Intense beams of stable and even unstable exotic species in virtually all charge states are available, up to the heaviest natural element, uranium. Two new series of experiments have recently been initiated at the ESR that represent major advances in the investigation of HCI: Firstly, isotope shift measurements of few-electron HCI employing RRS (section 2). Secondly, the technique of DR experiments with a stochastically cooled or pre-cooled ion beam has been developed. The most fundamental high-energy processes like the DR of H-like uranium U$^{91+}$ can now be accessed at the ESR (section 3).

PR, i.e., the capture of a free electron with immediate or subsequent emission of one or more photons can proceed either via a non-resonant channel, radiative recombination (RR), or a resonant pathway, dielectronic recombination (DR).

\[
e^- + A^{q+} \rightarrow (A^{(q-1)+})^{**} \downarrow A^{(q-1)+} + \text{photons.} \tag{1}
\]

In RRS the resonance condition of the first step of DR, the dielectronic capture (DC, time-reversed autoionization) of a free electron is utilized. In the DC process, an autoionizing doubly excited intermediate state is formed that can either re-ionize or alternatively decay via the emission of photons to below the autoionization threshold. In the latter case, the full two-step DR reaction is complete and a recombined ion is produced.

For RRS experiments, the ion species under investigation is injected into the storage ring and cooled down to minimize the beam size and energy spread. The circulating heavy ions interact with the co-propagating electron beam of an electron cooler or a cooler-like electron target. Recombination products are separated from the primary beam in the next bending magnet of the storage ring and are detected with 100% efficiency. Sequences of electron-ion collision energies are introduced by ramping the electron energy in a well defined and precise manner. The center-of-mass (c.m.) energy is deduced from the detuning voltage of the electron cooler with respect to cooling conditions (zero relative energy). If the c.m.-energy matches the resonance condition for DC an increased number of recombined ions is recorded. The spectroscopic information is then inferred from the resonance positions in the DR reaction cross section, and hence no photon energy measurement is necessary. Normalization of the number of recombined ions on the ion beam intensity and the electron density yields the PR rate coefficient on an absolute scale.

2. Isotope shift measurements of heavy Li-like ions by means of DR

With increasing nuclear charge the mutual overlap of the electronic wave function and the atomic nucleus plays an increasingly important role in the atomic structure. The contribution of the nuclear finite size to the electron binding energies rises with the 5th to the 6th power of the nuclear charge $Z$, thus exceeding the typical $Z^4$ scalings of relativistic or QED effects. On one hand, detailed knowledge about the nuclear size and partially also about its structure are essential ingredients for the clear interpretation of high-precision atomic-structure studies with respect to QED contributions. This is particularly true in view of the progress in experimental accuracy that has been lately achieved in the spectroscopy of H-like and Li-like uranium.
Isotope shift in the dielectronic recombination resonance spectrum of the two Li-like neodymium $^{A}\text{Nd}^{57+}$ isotopes with $A=142$ (grey line) and $A=150$ (black line). The initial dielectronic capture (time-inverse Auger process) populates intermediate doubly excited states, i.e., $e + \text{Nd}^{57+}(1s^22s_{1/2}) \rightarrow \text{Nd}^{56+}(1s^22p_{1/2}19\ell j)$.

On the other hand, the tightly bound electrons of HCI can be utilized as sensitive probes of the source of their atomic binding, the heavy nucleus. The use of DR of few-electron HCI for the investigation of charge radii and hyperfine effects is a novel alternative to the established methods like elastic electron scattering, the spectroscopy of muonic atoms, optical or X-ray spectroscopy (see e.g. [15, 16] and references therein). The individual approaches probe the nuclear charge distribution in a different manner and yield in many cases results for the charge radii that are not in good agreement with each other. For the generation of data compilations [15, 16, 17] considerable effort is being spent to produce a consistent set of rms values for the nuclear charge distribution. Isotope shift studies using DR have been discussed in the literature for a few years [18, 19, 20]. For nuclear charges $Z > 50$ a precision comparable to the existing methods can be expected. As a consequence of the high experimental resolution, of the large resonant atomic cross sections and with a detection efficiency close to unity, RRS is a highly sensitive method. DR experiments with as few as $10^3 - 10^4$ particles can be performed. Special emphasis should be given to the possibility to access unstable species like exotic isotopes or even the excited states of isomers if their lifetime is long enough to store and cool the ions ($\sim 1$ s).

A first experimental attempt to study DR isotope shifts has been made at the TSR storage ring with the 3 isotopes $A=194$, $A=195$ and $A=198$ of $^{A}\text{Pt}^{48+}$ [9]. Due to poor counting statistics no shift in the resonance positions could be determined. However, another pronounced difference in DR data of the even isotopes compared to the ones of the odd isotope $A=195$ was found. In the DR spectrum of $^{195}\text{Pt}^{48+}$ several DR resonances were absent although they were present in the spectra of $^{194}\text{Pt}^{48+}$ and of $^{198}\text{Pt}^{48+}$. This strong ‘isotope’ effect was attributed to hyperfine quenching of metastable states in the Zn-like ions [9]. In a recent measurement at the CRYRING in Stockholm, for the isotope pair $^{207}\text{Pb}^{53+} - ^{208}\text{Pb}^{53+}$ of Cu-like lead, shifts of low-energy DR resonances caused by magnetic hyperfine interaction were observed. The corresponding hyperfine
constant could be extracted from the data [10]. Few-electron HCI—like the heavy Li-like ions under consideration in this report—have an inherent advantage over heavy atoms or moderately charged ions. The physical process under investigation is not masked by the many-body character of the electrons, allowing clear, unambiguous conclusions to be drawn. Furthermore, nuclear size effects scale proportional to $n^{-3}$ and contribute significantly to the $\Delta n = 0$ intrashell excitations in Li-like ions. For instance, for the $2p_{1/2} - 2s_{1/2}$ transition in Li-like $^{U^{59+}}$ the finite size of the nucleus causes about a 12% contribution to the transition energy [3]. Here, we present results from a pilot isotope shift experiment with the two stable Li-like neodymium isotopes $^{A}$Nd$^{57+}$ with $A=142$ and $A=150$. More details about the experimental procedure for RRS measurements at the storage ESR can be found in [3, 6], and further information about the choice of neodymium as the primary candidate for this first studies are available in [20]. In DR, isotope shifts do not lead to a shift of a single resonance but to a shift of a whole resonance pattern. In the present case of $^{A}$Nd$^{57+}$, the resonant capture of the free electron is associated with $2s \rightarrow 2p_{1/2}$ and $2s \rightarrow 2p_{3/2}$ core excitations and leads to Rydberg series of Nd$^{56+}$ $(1s^22p_{1/2}n\ell_j)$ configurations with $n \geq 18$, and of Nd$^{56+}$ $(1s^22p_{3/2}n\ell_j)$ configurations with $n \geq 8$, respectively. The isotope shift is mediated by the strong change of the overlap of the $2s$ wave function with the nucleus. All resonances belonging to the same core excitation exhibit the same isotope shift since the loosely bound Rydberg electron is a mere spectator that does not contribute to the energy shift. In Figure 1 the DR spectra of the two isotopes $^{142}$Nd$^{57+}$ and $^{150}$Nd$^{57+}$ in the narrow energy range (14.9 eV to 17.3 eV) of the resonance group Nd$^{56+}$ $(1s^22p_{1/2}19\ell_j)$ are shown. An energy shift of about $\sim 40$ meV is clearly visible demonstrating nicely the sensitivity of DR to isotope effects. The data for the spectra in Figure 1 were recorded within about 1 hour of data taking for each isotope with less than $10^7$ particles stored in the ring. For both isotopes experimental data have been obtained from the energetically lowest resonance group Nd$^{56+}$ $(1s^22p_{1/2}18\ell_j)$ at around 0.7 eV to 3.0 eV up to an energy of about 42 eV. This energy range provides a multitude of resonance structures that all exhibit a corresponding energy shift. Due to the simple electronic structure in Li-like ions the isotope shift is mainly determined by the nuclear size effect. The change in the rms charge radius will be calculated with the formulas in [21]. As both isotopes are even-even nuclids, no nuclear spin and no magnetic hyperfine splitting is present. Other corrections like changes in QED contributions, normal mass shift or many-body effects like specific mass shift are small and can be accounted for. These new data on the isotope shift of heavy few-electron ions are a promising starting point for further experiments with stable and unstable HCI. Candidates of special interest that exploit this new technique are lanthanides or the isotopes of the heaviest naturally occurring element, uranium.

3. Photorecombination studies of heavy K-shell ions

In very heavy HCI, like the one-electron uranium $^{U^{91+}}$ system, the tightly bound electrons are exposed to extreme electromagnetic fields of the heavy nuclei. Thus, the investigation of HClCs in this strong field limit provides the ultimate testing ground for our understanding of the fundamental interaction QED. A widely unexplored field is the influence of relativistic and QED corrections on the dynamics of atomic reactions. An optimal tool to address this question is again DR [22]. A detailed analysis of resonance strengths, linewidths and lineshapes provides access to relativistic and QED contributions to Auger and radiative matrix elements [23], to interference patterns [24] or even to QED effects in overlapping resonances [25]. For H-like and He-like ions, the energetically lowest DR process proceeds via a capture of the free electron into the L-shell and a $\Delta n = 1$ excitation K→L of the bound electron. Accordingly, the minimum resonance energy of H-like ions (KLL) is about half the binding energy of the K-shell ion, i.e., more than 64 keV in the case of $^{U^{91+}}$. The standard technique for DR measurements at the ESR [3, 6] of alternately switching the electron cooler within short time intervals (typically a
Figure 2. Preliminary experimental data for the PR of U$^{91+}$ in the energy range of the KLL-DR resonances (■) and fully relativistic DR-MCDF calculations convoluted with a 120 eV Gaussian (——). The experimental data are shifted by 200 eV. The energy shift is mainly caused by the space charge potential of the electron beam and will be corrected for in the final analysis.

A change of more than 170 kV in the laboratory frame is required for reaching the 64 keV collision energy in the c.m.-frame. From these numbers the necessity to decouple the ion beam cooling from the measurement process is obvious. Therefore, the U$^{91+}$ primary beam was injected into the ESR and stochastically pre-cooled [26] to a momentum spread of $dp/p \approx 5 \times 10^{-4}$, and the electron cooler was used as a target for free electrons only. After pre-cooling at an energy of about 400 MeV/u, the ions were decelerated to a 0.8% lower momentum, i.e., to a more central orbit for the measurement. The latter step was necessary as the ion optical settings of the storage ring required for stochastic cooling did not allow simultaneous stochastic cooling of the primary beam and collection of the recombined U$^{90+}$ ions. A moderate RF bunching amplitude was applied during the measurement to counter-balance energy-losses in the residual gas of the storage ring and in the electron cooler. The cooler was set to potentials of 38.85 kV and 35.77 kV equivalent to c.m.-energies of 64.8 keV and 70.6 keV. Around these center values fast energy scans (53 ms per measurement point) were performed by decelerating or accelerating the electrons using drift tubes in the beam overlap region of the cooler. With this procedure we could measure the PR rate coefficient in the desired energy range of the KLL-DR resonances of U$^{91+}$ (Fig. 2). The total of 10 doubly excited U$^{90+}(2\ell j \ell j')$ states are split into 3 groups of fine structure components with $j = 1/2, j' = 1/2$, with $j = 3/2, j' = 1/2$ and with $j = 3/2, j' = 3/2$. The smooth background in the spectrum is predominantly caused by contributions of the non-resonant RR process. The experimental data are displayed together with our fully relativistic DR-MCDF calculation including Breit interaction. Details on the theoretical approach can be found in [23, 24]. For an easier comparison the theoretical cross section is convoluted with a 120 eV Gaussian, and RR contributions have been added. The preliminary experimental data are normalized to the theory and shifted by 200 eV. A more refined data analysis will yield experimental energies as well as rate coefficients on scales independent from theory, thus...
allowing for a detailed and quantitative comparison. It is worth noting that the experimental resolution achieved with this novel technique is already close to the natural linewidths of ~50 eV of the doubly excited states.

4. Conclusion
With the new method of DR measurements with a stochastically (pre-)cooled beam the available collision energy range is expanded towards higher energies. RRS studies at storage rings can now be performed in an unprecedented large dynamic energy span ranging basically from ‘zero’ relative electron-ion collision energy up to ~100 keV in the c.m. frame. The high efficiency of RRS facilitates experiments with exotic low intensity beams or processes with low cross sections. As a consequence of the co-propagating set-up at the electron cooler a remarkably high energy resolution is achieved. Structures in the resonant cross section can be resolved on a level below $10^{-2} - 10^{-1}$ eV at low electron-ion collision energies and on a level of ~100 eV at 75 keV. Already the present installations at the storage ring ESR of GSI provide a unique experimental environment for RRS studies of HCI. Particularly appealing are the future prospects at GSI.

As part of the extension of the existing accelerator complex to a “Facility for Antiproton and Ion Research” (FAIR) the successor of the ESR, the NESR, will be equipped with a dedicated ‘ultra-cold’ electron target that operates independently of any beam cooling tasks and that is optimized with respect to an electron beam as monoenergetic as possible [27]. It can be expected that these new installations, the technological advances, the combination with other new in-ring equipment and the increased beam intensities of stable and unstable species will boost resolution and versatility of the future studies.

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