High Rydberg Resonances in Dielectronic Recombination of Pb^{79+}

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Dielectronic recombination resonances of Pb^{79+} associated with $2s_{1/2} \rightarrow 2p_{1/2}$ excitations were measured at the heavy-ion storage ring ESR at GSI. The fine structure of the energetically lowest resonance manifold $Pb^{78+}(1s^2 2p_{1/2} 20l_j)$ at around 18 eV could partially be resolved, and rate coefficients on an absolute scale were obtained. A comparison of the experimental data with results of a fully relativistic theoretical approach shows that high-angular-momentum components up to $j = 31/2$ significantly contribute to the total resonance strength demonstrating the necessity to revise the widespread notion of negligible high-angular-momentum contributions at least for very highly charged ions.

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During the last decade dielectronic recombination (DR) experiments at storage rings have proved to be a versatile alternative to conventional optical spectroscopy [1]. Besides the information on resonance energies DR measurements on an absolute scale also provide a sensitive tool for the investigation of radiative transition rates and autoionization rates, and hence the transition probabilities and amplitudes (e.g., [2,3]). The high energy resolution available in these experiments particularly at low electron-ion collision energies allows for the state-selective preparation and observation of autoionizing high Rydberg ions. With the present work we demonstrate a level of experimental energy resolution and accuracy that opens up new possibilities for precision spectroscopy of very highly charged ions allowing information to be obtained that can challenge state-of-the-art calculations of QED effects in strong fields [4] and is even promising to probe the nuclear size of isotopes including radioactive species with lifetimes as low as only minutes.

DR is the resonant two-step channel of photorecombination (PR), an electron-ion collision process in which a free electron is captured and the excess energy is carried away by one or more photons. In dielectronic capture (DC) which is the first step of DR, a bound electron is excited by the simultaneous radiationless capture of a free electron (time-reversed autoionization). DR is completed if the doubly excited intermediate state decays radiatively to a state below the autoionization threshold. For the $2s_{1/2} \rightarrow 2p_{1/2}$ core excitation of lithiumlike ions DR can be described by:

$$e^- + A^{q+}(1s^2 2s_{1/2}) \rightarrow A^{(q-1)+}(1s^2 2p_{1/2} nl_j)^{**} \rightarrow A^{(q-1)+} + \text{photons}. \quad (1)$$

As a consequence of energy conservation, DC is possible only for sufficiently large $n \geq n_{\min}$. For DR associated with the $2s_{1/2} \rightarrow 2p_{1/2}$ transition in Pb^{79+} ions $n_{\min} = 20$. Alternatively, PR can proceed via the nonresonant direct pathway of radiative recombination (RR).

In this Letter the focus is on the $Pb^{78+}(1s^2 2p_{1/2} 20l_j)$ doubly excited resonance manifold. In highly charged few-electron ions even very high Rydberg states, like the $n = 20$ resonance under investigation, exhibit a fine-structure splitting of a few eV, more than 7 orders of magnitude larger than in hydrogen. The DR cross section is mainly determined by the *slowest* of either the radiative or autoionizing decay of the doubly excited intermediate state. For a given isoelectronic sequence of ions the radiative rates scale strongly with the nuclear charge Z, whereas the autoionization rates are roughly independent of Z. In contrast to doubly excited states of light ions, in heavy ions the radiative rates exceed the autoionization decay rates, therefore heavy ions provide a particular sensitivity to the autoionization probabilities, and hence to the electron-electron interaction. Furthermore, very high angular momenta are potentially involved within each Rydberg manifold. With increasing orbital angular momentum quantum number j the mutual influence between the highly excited Rydberg electron and the lithiumlike core configuration becomes weaker. The resulting reduction of the autoionization rate is partially counterbalanced, however, by the increasing

statistical weight $2j + 1$. The assessment of the distribution of resonance strengths within a Rydberg manifold of resonances as well as the fine structure of the high Rydberg states are the subject of the present work.

Much of the motivation for a detailed investigation of high Rydberg states stems from an attempt to precisely determine the $2s_{1/2} - 2p_{1/2}$ excitation energy (Lamb shift) of very heavy lithiumlike ions by utilizing DR. By measuring the individual resonance positions of a multitude of $1s^2 2p_{1/2} n l_j$ resonances formed by DR it is possible to extrapolate the energies to the series limit ($n \to \infty$), and, hence, to obtain the excitation energy of the lithiumlike parent ion [5,6]. For that analysis the distribution of resonance strengths within each Rydberg manifold has to be known.

Here we present results from a combined experimental and theoretical study of high Rydberg resonances in the DR of lithiumlike lead (Pb^{79+}). In particular, detailed information about the fine structure and the contributions of individual $1s^2 2p_{1/2} 20 l_j$ sublevels to the total resonance strength is provided. In the experiment surprisingly large contributions from states with high angular momenta were found with the peak in the distribution of resonance strengths at about $j = 17/2$. This finding contrasts the general notion of negligible high-angular-momentum contributions and particularly contradicts previous theoretical calculations [7].

In order to clarify this discrepancy we performed new calculations that included Auger and radiative rates of states with high angular momenta. The calculations are based on a fully relativistic treatment within the isolated resonances (IRA) and independent processes approximation (IPA). In the IPA the cross section $\sigma^{\mathrm{DR}}_{i\to f}$ for DR can be written as a product of the cross section $\sigma^{\mathrm{DC}}_{i\to d}$ for DC and the fluorescence yield $\omega_{d\to f}$ of the intermediate doubly excited state d. DC is related to autoionization by the principle of detailed balance, and hence

$$\sigma^{\mathrm{DR}}_{i\to f}(E) = \frac{\pi^2}{E}\,\frac{g_d}{2g_i}\,\mathcal{L}(E_r, E, \Gamma^{\mathrm{tot}}_d)\Gamma^{\mathrm{Aug}}_{d\to i}\omega_{d\to f}\,. \qquad (2)$$

Here E is the energy of the free electron, E_r the resonance energy, g_i and g_d are the statistical weights of initial and intermediate states, $\Gamma^{\mathrm{Aug}}_{d\to i}$ is the partial Auger width with respect to the initial state i, and Γ^{tot}_d is the total width of level d. The Lorentzian $\mathcal{L}(E_r, E, \Gamma^{\mathrm{tot}}_d)$ describes the shape of the cross section in the vicinity of the resonance. Auger rates were obtained using correlated relativistic wave functions including usual Coulomb and the generalized Breit interaction. Resonance energies and radiative transition rates were calculated with the multi configuration Dirac-Fock (MCDF) code GRASP [8,9] which takes into account the extended size of the atomic nucleus, Breit interaction, and QED contributions. The charge distribution of the nucleus was modeled with a Fermi distribution with a rms radius of 5.51 fm. The fully relativistic calculations directly include angular momenta up to $j \le 21/2$. Higher angular momenta, which contribute on a level of 10% were obtained by extrapolation of the values from $j \le 21/2$. Further details of the theoretical method can be found elsewhere [10–12].

The experiments were performed at the heavy-ion storage ring ESR at GSI in Darmstadt. Lithiumlike Pb^{79+} ions with an energy of 97.5 MeV/u were injected into the ESR and cooled such that the ion beam acquired a diameter of approximately 1 mm and a longitudinal momentum spread of $\delta p/p < 5 \times 10^{-5}$. In addition to electron cooling the ESR cooler also served as a target of free electrons. Typical ion currents of up to 600 μA corresponding to 4×10^7 stored ions at the beginning of a measurement cycle were accumulated in the ring. At a ring vacuum of a few 10^{-9} Pa the lifetime of the ion beam is mainly determined by electron-ion recombination in the cooler. At an electron current of $I_e = 80$ mA the lifetime $T_{1/2}$ of the ion beam was about 1000 s. Nonzero relative energies between electrons and ions were introduced by applying voltages to cylindrical drift tubes (1.94 m length) surrounding the straight overlap region (2.5 m) of the two beams. With the aid of fast power supplies the potential of these drift tubes can be switched within 2 ms to any desired value between -5 kV and $+5$ kV. With zero drift tube potential and a cooling voltage of $U_c = 53.51$ kV a center-of-mass energy range between 0 and about 100 eV could be covered. After the injection of the ions into the ring the beam was cooled for about 20–30 s before the data acquisition was started. Every 4 ms the data acquisition was read out and all relevant experimental parameters were stored on tape. A measurement cycle consisted of 7 subcycles with 236 nonzero voltage steps of 40 ms duration and a cooling interval of 20 ms after each step. An additional extended cooling period of 1 s was applied after each subcycle. The intermittent cooling preserves identical experimental conditions for every measurement, minimizes drag effects by the friction force exerted on the ion beam by the electron beam, defines zero relative energy, and can be used as an additional check for normalization. The complete cycle of 1652 steps was repeated 4 to 10 times before the ring was refilled. Recombined ions were counted with nearly 100% efficiency by a fast plastic scintillation counter located behind the next dipole bending magnet downstream from the electron cooler. Further details of the measurement procedure at the electron cooler of the ESR can be found in [13,14]. Normalization of the number of recombined ions R to the electron density n_e and the number of stored ions N_i yields the experimental rate coefficient α_{exp} on an absolute scale

$$\alpha_{\mathrm{exp}} = \frac{C\gamma^2 R}{n_e N_i L_{\mathrm{eff}}}\,, \qquad (3)$$

where $C = 108.36$ m is the circumference of the ESR, γ the relativistic Lorentz factor of the ions, and L_{eff} is the effective beam overlap length which depends on the chosen measurement potential [13,14]. In the energy region of the

$Pb^{78+}(1s^2 2p_{1/2} 20l_j)$ resonances (13–19 eV) we obtained $L_{\text{eff}} = 1.45$ m. The systematical uncertainty of the measured rate coefficient is estimated to be $\pm 15\%$. The main contributions to this uncertainty arise from the determination of the electron density ($\pm 10\%$), the ion current ($\pm 5\%$), and the effective length $L_{\text{eff}}(\pm 5\%)$. A first order energy calibration accurate to about $\pm 2\%$ can be obtained by calculating the relative energy scale from the ion energy and the applied drifttube voltages. An improved energy calibration has been performed by using the extrapolation method mentioned above and is the subject of a forthcoming publication [15]. After application of this method the remaining uncertainties in the present resonance positions are below 0.2 eV.

Experimental and theoretical DR rate coefficients $\alpha = \langle v\sigma \rangle$ in the energy range of the $Pb^{78+}(1s^2 2p_{1/2} 20l_j)$ Rydberg resonance are displayed in Fig. 1 together with the previous theoretical results of Mitnik *et al.* [7]. In order to account for the experimental response function our theoretical cross sections have been energy averaged with an anisotropic Maxwell-Boltzmann distribution [16] with parameters $k_B T_{||} = 0.2$ meV and $k_B T_{\perp} = 120$ meV. For the comparison with the present rate coefficients the cross sections calculated by Mitnik *et al.* [7] were multiplied with the relative velocity v_{rel}. This is justified since the previous calculations already included an experimental energy spread of 0.3 eV (FWHM). The $Pb^{78+}(1s^2 2p_{1/2} 20l_j)$ resonance manifold is split into groups of resonances with an identical angular momentum quantum number j of the Rydberg electron. Each of these groups consists of four individual configurations which result from the coupling of the two $l = j \pm 1/2$ components of the Rydberg electron with the angular momentum of the $2p_{1/2}$ core electron to the total angular momentum J of the doubly excited state. The Rydberg resonance group covers an energy range of about 4 eV. This energy splitting is well reproduced by both calculations. In order to investigate the mutual influence of core electrons and the Rydberg electron on the splitting, the resonance energies have also been calculated in a relativistic hydrogenic approximation (Dirac binding energies) of the Rydberg electron, assuming a series limit of $E(2s_{1/2} - 2p_{1/2}) = 230.68$ eV [17]. The results of this calculation are shown in Fig. 1(b). The strongest influence of the core is expected for $j = 1/2$ and hence it is interesting to see that even in this extreme case the electron-core interaction results in an additional shift of only about 0.5 eV. Furthermore, the size of radiative (QED) contributions to the binding energy of the Rydberg electron has been estimated from the self-energy of a $n = 20$ electron. The self-energy which is about 90% of the total QED contribution is $E_{SE}(20s_{1/2}) \approx 34$ meV and rapidly decreases to $E_{SE}(20g_{9/2}) \approx 0.2$ meV for higher angular momenta [18–20].

Besides the energy splitting also the shape and the absolute value of the rate coefficient—experiment and theory are both on independently absolute scales—are

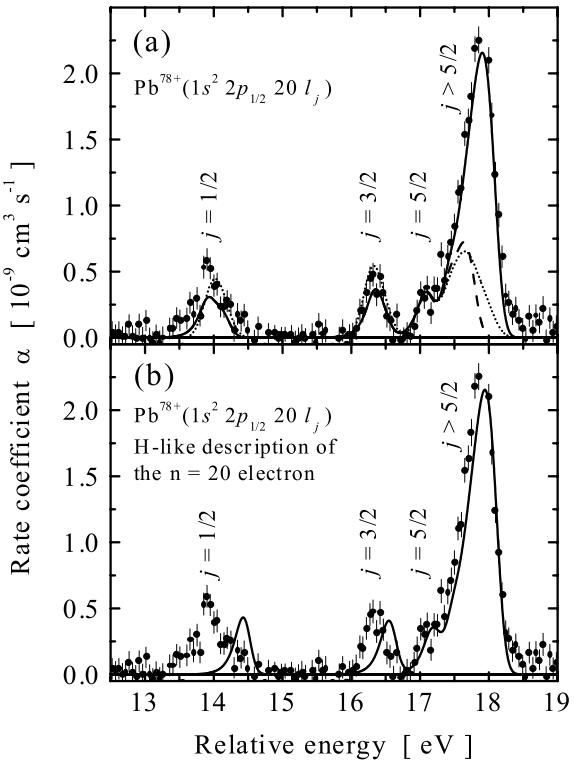

FIG. 1. Measured and calculated DR rate coefficients α for the $Pb^{78+}(1s^2 2p_{1/2} 20l_j)$ resonance group. The experimental error bars denote statistical errors only. In (a) the experimental data (full circles) are compared to our fully relativistic calculations including states up to $j \leq 31/2$ (full line) and states up to $j \leq 11/2$ only (dashed line). The theoretical cross sections have been convoluted with an anisotropic Maxwell-Boltzmann velocity distribution of the electrons ($k_B T_{||} = 0.2$ meV and $k_B T_{\perp} = 120$ meV). The dotted line is the calculation of Mitnik *et al.* (assumed resolution in their paper is 0.3 eV FWHM). Our theory has been shifted by -0.65 eV and the theory of Mitnik *et al.* by -0.3 eV to match the experimental peak positions. Both theories are known to have uncertainties of ≈ 1 eV in absolute peak positions mainly due to the approximations used to calculate the QED contributions. Panel (b) of the figure shows experimental and theoretical data, but with the theoretical resonance energies calculated in a relativistic hydrogenic approximation (Dirac binding energies) of the $n = 20$ electron.

well ex-plained by our calculations. The main part of the total resonance strength $S = \int \sigma(E)\, dE$ stems from high angular momentum components and is accumulated in the experimentally not resolved sum peak with $j > 5/2$. For low angular momenta the calculations of Mitnik *et al.* are in good agreement with our theoretical and experimental data, which is no surprise; as for $j \leq 11/2$ both calculations are based on a similar approach. For higher angular momentum components, however, Mitnik *et al.* used a different calculational procedure [7]: Auger rates were obtained in a semirelativistic approach and the radiative rates were approximated by nonrelativistic hydrogenlike dipole transitions. The resulting DR rate coefficients of Mitnik *et al.* show a rapid decrease for $j > 11/2$ yielding

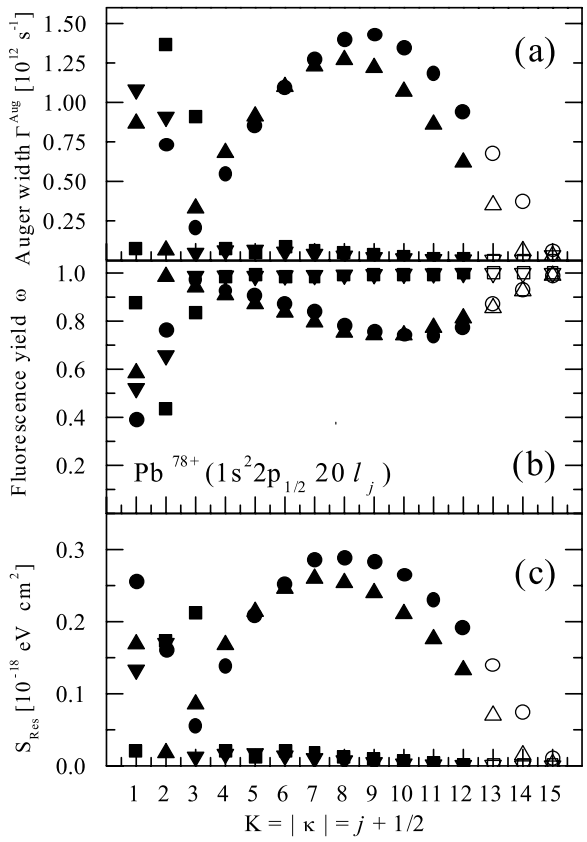

FIG. 2. Details of the DR calculations. (a) Auger widths $\Gamma^{\mathrm{Aug}} = g_d/2g_i\Gamma^{\mathrm{Aug}}_{d\to i}$, (b) fluorescence yields $\omega_{d\to f}$, and (c) resonance strengths $S = \int \sigma(E)\,dE$ in dependence of the relativistic quantum number $K = |\kappa| = j + 1/2$ of the Rydberg electron. For each j four couplings of the $1s^2 2p_{1/2}$ core and the Rydberg electron are possible: $J = l = j - 1/2$ (squares), $J = l + 1 = j + 1/2$ (circles), $J = l = j + 1/2$ (triangle up), and $J = l - 1 = j - 1/2$ (triangle down). Values for $j > 21/2$ have been extrapolated from $j \leq 21/2$ and are indicated by open symbols.

at least a factor of 5 less resonance strength in the high-j accumulation peak compared to our results.

Details of our calculation show (see Fig. 2) that the maximum contribution is from states with angular momentum $j = 17/2$ and that even higher j have to be taken into account. This situation drastically differs from circumstances in lighter lithiumlike ions such as, e.g., Ar^{15+} where for $n = 20$ the highest contributions are from $l = 2$ and decrease for higher angular momenta [21]. For high values of j our calculated resonance strengths of the two states with $J = j - 1/2$ are more than an order of magnitude smaller than those with $J = j + 1/2$. The doubly excited intermediate state decays mainly via the radiative deexcitation of the Rydberg electron while the radiative transition $2p_{1/2} \to 2s_{1/2}$ is typically 2 orders of magnitude slower. In contrast to low-Z ions the Auger rate is smaller than or close to the radiative rate and hence the fluorescence yield $\omega_{d\to f}$ is typically 0.5 to 1.0.

In conclusion, DR of a very heavy lithiumlike ion with the captured electron in a high Rydberg state has been investigated experimentally and theoretically. Contributions from very high angular momenta must be included and considered in a fully relativistic framework in order to understand the dynamics involved in the DR process. The data presented here could only be obtained due to the vast progress that has been accomplished in cooling efficiency, counting statistics, and energy resolution since the first DR measurements of very heavy few electron ions at the ESR [22]. Our results demonstrate experimental capability to perform precision spectroscopy of very highly charged ions without a photon in sight. The energy resolution obtained here paves the road to the determination of the Lamb shift of lithiumlike ions with a precision that will challenge state-of-the-art theory for QED in strong fields.

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[1] E. Lindroth *et al.*, Phys. Rev. Lett. **86**, 5027 (2001).
[2] P. H. Mokler and T. Stöhlker, Adv. At. Mol. Opt. Phys. **37**, 297 (1996).
[3] A. Müller and A. Wolf, in *Accelerator Based Atomic Physics Techniques and Applications*, edited by J. C. Austin and S. Shafroth (AIP Press, Woodbury, New York, 1997).
[4] V. M. Shabaev, Phys. Rep. **356**, 119 (2002).
[5] W. Spies *et al.*, Nucl. Instrum. Methods B **98**, 158 (1995).
[6] C. Brandau *et al.*, Hyperfine Interact. **114**, 45 (1998).
[7] D. M. Mitnik, M. S. Pindzola, and N. R. Badnell, Phys. Rev. A **61**, 022705 (2000).
[8] K. G. Dyall *et al.*, Comput. Phys. Commun. **55**, 425 (1989).
[9] F. A. Parpia, C. F. Fischer, and I. P. Grant, Comput. Phys. Commun. **94**, 249 (1996).
[10] P. Zimmerer, N. Grün, and W. Scheid, Phys. Lett. **148**, 457 (1990).
[11] P. Zimmerer, N. Grün, and W. Scheid, J. Phys. B **24**, 2633 (1991).
[12] M. Zimmermann, N. Grün, and W. Scheid, J. Phys. B **30**, 5259 (1997).
[13] A. Hoffknecht *et al.*, Phys. Rev. A **63**, 012702 (2000).
[14] W. Shi *et al.*, Eur. Phys. J. D **15**, 145 (2001).
[15] C. Brandau *et al.* (to be published).
[16] G. Kilgus *et al.*, Phys. Rev. A **46**, 5730 (1992).
[17] V. A. Yerokhin *et al.*, Phys. Rev. A **64**, 032109 (2001).
[18] É.-O. Le Bigot, P. Indelicato, and P. J. Mohr, Phys. Rev. A **64**, 052508 (2001).
[19] P. J. Mohr and Y.-K. Kim, Phys. Rev. A **45**, 2727 (1992).
[20] V. A. Yerokhin and V. M. Shabaev, Phys. Rev. A **60**, 800 (1999).
[21] J.-G. Wang, T. Kato, and I. Murakami, Phys. Rev. A **60**, 2104 (1999).
[22] W. Spies *et al.*, Phys. Rev. Lett. **69**, 2768 (1992).