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Breit interaction in dielectronic recombination of hydrogenlike uranium

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Absolute rate coefficients for dielectronic recombination (DR) of H-like U^{91+} ions have been measured. The electron-ion merged-beam technique at a heavy-ion storage ring was employed using a stochastically cooled ion beam. Thereby, the previously accessible electron-ion collision energies could be greatly extended to the range 63–90 keV. High-resolution DR spectra were measured covering all KLL and KLM resonances. For the resonance strengths, excellent agreement between relativistic theory and experiment is found only if the Breit contribution to the electron-electron interaction is included in the calculations. For the $KL_{1/2}L_{1/2}$ and $KL_{1/2}M_{1/2}$ groups the Breit contribution amounts to 44% of their total resonance strengths.

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Very highly charged few-electron ions are prominent objects for studies of fundamental atomic structure and, in particular, of relativistic and quantum electrodynamic (QED) effects. As a consequence of their scaling with high powers of the nuclear charge Z, contributions that are tiny in ordinary atoms are strongly magnified for high Z. But, still, these quantities, including the relativistic corrections to the electron-electron interaction, cause relatively small changes of the level energies of highly charged ions [1]. However, relativity and QED strongly alter the dynamics of a heavy atomic system, i.e., transition rates, angular distributions, and polarization effects. A strong Z dependence is naturally expected for quantities governed by electron-nucleus interactions (Z/r). In the static Coulomb case the electron-electron interaction is independent of Z ($1/r$). However, in the relativistic domain, quantities related to the electron-electron interaction can feature strong scaling with the nuclear charge. Such quantities are Auger transition rates. For KLL Auger rates in high-Z He-like ions Z^4 scalings have been predicted to be caused by the Breit interaction [2]. With increasing Z the Breit contribution to the Auger rates of some autoionizing configurations finally exceeds that of the Coulomb repulsion and dominates the transition rate. In its original form the Breit interaction for two moving charges is derived from classical electrodynamics using the correspondence principle and comprises contributions from current-current interaction and retardation corrections [3]. In terms of QED, the generalized Breit interaction (GBI) denotes the exchange of a transverse virtual photon of frequency ω between the electrons. In the limit for $\omega \to 0$ one obtains the standard Breit operator [3].

The present study scrutinizes the electron-electron interaction in the strongest possible atomic fields, i.e., the most appealing case for a decisive benchmark of the GBI. Dielectronic recombination (DR) resonance strengths of H-like U^{91+} ions were measured with high-energy resolution and on an absolute scale at the heavy-ion storage ring ESR in Darmstadt, Germany. The initial one-electron ion with no additional spectator electrons represents the most elementary case and, in combination with our independent absolute data, allows for unambiguous interpretation. In order to reach the required high electron-ion collision energies of 64–90 keV in a merged-beams experiment an alternative experimental approach was pursued: The ion beam was cooled stochastically, and the ESR electron cooler was exclusively used as a target of free electrons. In contrast, in earlier recombination measurements at GSI, the electron cooler was used alternatingly between beam cooling and electron-target operation, which restricted accessible energies in the center-of-mass frame to $E_{CM} \lesssim 1$ keV (e.g., Ref. [4]). Excellent experimental resolution was achieved facilitating detailed analysis of the Breit contribution for individual configurations. In support of the experimental work multiconfiguration Dirac-Fock (MCDF) calculations including the GBI were carried out.

In DR the initially free electron excites an electron that is bound to the primary ion. In this resonant dielectronic capture (DC), i.e., the time inverse of the Auger process, an autoionizing intermediate state is formed. DR is completed if the ion is stabilized by radiative decay to below the autoionization threshold. The resonance strength $S = \int \sigma_{DR}(E)\, dE$ for DR is proportional to the product of Auger rate A_a (formation by DC) and fluorescence yield γ. Hence, the DR process possesses a particular sensitivity to the Auger rates. In high-Z few-electron ions photon

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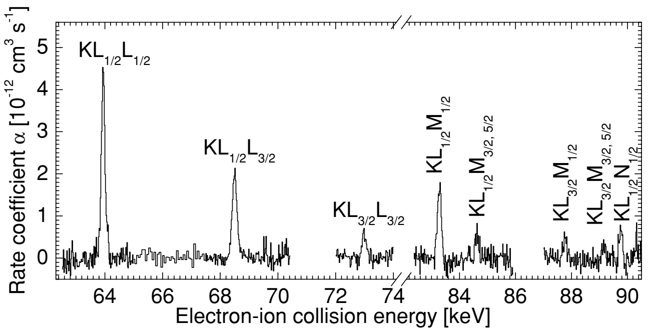

FIG. 1. Measured U^{91+} merged-beams DR rate coefficient. The data were obtained in several individual runs. For display, effects from the drift tube used for the energy scan and the beam merging have been deconvolved.

emission typically is very fast ($\gamma \sim 1$), and consequently $\sigma_{DR} \propto A_a$.

In several theoretical studies various aspects of U^{91+} DR have been treated. For example, in Ref. [5] the interference between DR and nonresonant radiative recombination (RR) was investigated, and in Ref. [6] the interference effects between overlapping DR resonances. The contribution of the GBI to the DR resonance strengths of heavy ions was calculated in Refs. [7–9] and included in a recent calculation of dielectronic recombination of U^{91+} by Andreev *et al.* [10]. Resonant transfer and excitation (RTE) of H-like uranium U^{91+} colliding with H_2 [11] and of He-like uranium U^{90+} colliding with H_2 [12] or a carbon foil [13] were previously studied in beam-target experiments. RTE is a process closely related to DR but with the captured electron initially bound to an atom or molecule. Even with H_2, the lightest available collision partner, the associated Compton profile leads to a substantial broadening of the individual resonances [11]. In both experiments the angular distribution of emitted x-ray photons was determined and compared with fully relativistic calculations including Breit interaction [13,14]. KLL-DR of few-electron heavy ions was observed at electron-beam ion traps (e.g., Refs. [15–17]). The main focus of Refs. [15,16] was the investigation of DR-RR interferences by means of x-ray spectroscopy, and hence no absolute cross sections were measured. In their experiment on KLL-DR of the Li-like iodine, holmium, and bismuth ions, Nakamura *et al.* [17] explored the importance of GBI to the DR process in a comparison of DR resonance strength ratios. Despite these experimental efforts no KLX-DR ($X = L, M, \ldots$) measurements have been performed on an absolute scale yet.

In the present work absolute DR resonance strengths have been determined on an absolute scale with an accuracy of 13% and resonance energies with an accuracy of 0.05%. Our different method enabled us to measure all groups of U^{91+} KLL and KLM-DR resonances with a resolution close to the natural line width (cf. Figs. 1–3). The measured data are compared to state-of-the art MCDF calculations of KLL and KLM-DR that include GBI. Table I lists the calculated resonance parameters for all 10 KLL-DR resonances of U^{91+}. The corresponding resonance spectra are shown in Fig. 2. The present MCDF resonance energies agree well with results from other theoretical methods that are also employed as a

TABLE I. Calculated U^{91+} KLL DR resonance energies E_{res}, natural line widths Γ, and resonance strengths S. For details about the estimation of uncertainties see Ref. [18].

	Coulomb + Breit			Coulomb	
Resonance	E_{res} (eV)	Γ (eV)	S (kb eV)	Γ (eV)	S (kb eV)
$KL_{1/2}L_{1/2}$					
$[2s_{1/2}\,2p_{1/2}]_0$	63 923(3)	33.2	18.2	32.6	3.17
$[2p^2_{1/2}]_0$	63 936(4)	35.9	15.8	35.7	10.8
$[2s_{1/2}\,2p_{1/2}]_1$	63 963(3)	32.7	20.9	32.7	17.1
$[2s^2_{1/2}]_0$	64 105(4)	29.8	6.79	29.7	3.44
$KL_{1/2}L_{3/2}$					
$[2s_{1/2}\,2p_{3/2}]_2$	68 425(3)	21.9	5.44	21.9	2.26
$[2p_{1/2}\,2p_{3/2}]_1$	68 462(3)	54.1	2.18	54.1	0.17
$[2p_{1/2}\,2p_{3/2}]_2$	68 477(5)	54.3	19.4	54.3	23.1
$[2s_{1/2}\,2p_{3/2}]_1$	68 550(3)	22.0	8.44	22.0	8.65
$KL_{3/2}L_{3/2}$					
$[2p^2_{3/2}]_2$	72 988(3)	43.6	7.39	43.6	6.68
$[2p^2_{3/2}]_0$	73 084(4)	43.6	1.30	43.6	0.59

cross-check within our work (see Ref. [18] for details of all methods). With respect to resonance strengths, Table I and Fig. 2 reveal that the DR intensities change considerably when GBI is included in the calculations and in 8 out of 10 cases enhance the resonance peak area. This agrees with earlier theoretical findings for U^{91+} [7] and other very highly charged ions [8,9]. The largest enhancement due to the GBI is found for the $[2s_{1/2}2p_{1/2}]_0$ resonance and amounts to a factor of ~5.7. Similarly, the $[2s^2_{1/2}]_0$, $[2s_{1/2}2p_{3/2}]_2$, and $[2s^2_{3/2}]_0$ states feature a doubling of the resonance strength.

For the present experiment, U^{91+} ions were provided by the GSI chain of accelerators, injected into the ESR at an energy of ~400 MeV/u and subsequently stochastically cooled. On the "outer" orbit required for stochastic cooling the separation between the primary beam and the recombined ions in the first dipole magnet behind the cooler target is already too strong to guide the reaction products onto the particle detector. Therefore, for the measurement, stochastic cooling was switched off and the ion beam was rebunched and shifted to a more central orbit by deceleration to 0.8% lower momentum. A moderate RF bunching amplitude was retained in order to preserve a constant ion energy. Even

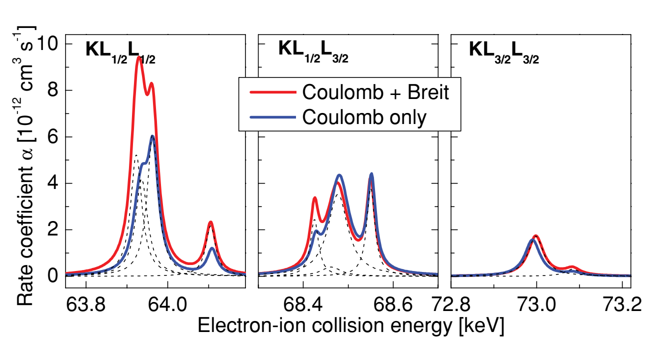

FIG. 2. (Color online) Theoretical DR rate coefficients including GBI (red line) and without (blue line). The line shape is natural line widths only. The individual resonances that form the respective group are indicated by black dashed lines (GBI included).

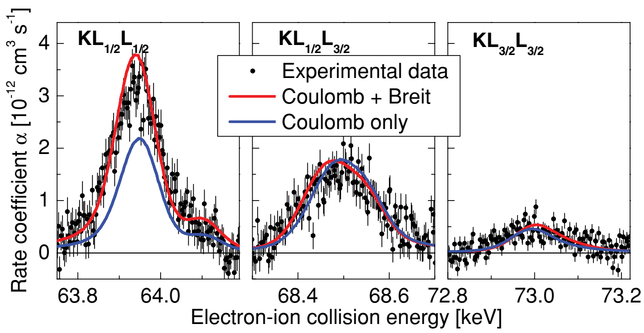

FIG. 3. (Color online) Measured (symbols) and simulated (lines) U^{91+} DR rate coefficient for the KLL resonance groups. The simulations comprise the theory values from Table I including GBI (red) and without (blue). The beam parameters are mimicked by an ion beam momentum spread of $\delta p/p = 7.5 \times 10^{-4}$, electron beam temperatures of $kT_\parallel = 0.2$ meV, and $kT_\perp = 120$ meV as well as the effects from drift tube, merging and demerging. It is noted that experiment and simulation are on independently absolute scales.

without permanent cooling the ion beam momentum spread could be kept as low as $\delta p/p \sim 7.5 \times 10^{-4}$. The voltage in the electron cooler was set to a center value U_c, and sequences of swiftly switched accelerating and decelerating voltages U_d (53 ms dwell time per point) were applied to drift tubes that enclose the beam overlap region (see Ref. [19] for details). Thereby, the electron energy was varied around the center value. The electron velocity $\beta_e = v_e/c$ (in units of the speed of light) is determined from the sum $U_c + U_d$ corrected for space charge [4,19]. For example, for the used ion velocity $\beta_i = v_i/c = 0.7122$ with $U_c = 38.87$ kV and $-2.9\,\text{kV} \leqslant U_d \leqslant 3.1$ kV, the energy range 70.5 keV $\geqslant E_{\text{CM}} \geqslant$ 62.5 keV was accessed. The sequence of beam injection, stochastic cooling, orbit change, and ramping of the drift tube voltage was repeated many times in order to reduce the statistical uncertainties of the measurement. Different energy ranges were scanned by setting U_c to a new center value. Recombined ions were recorded as a function of U_d using a gas counter (∼100% efficiency). For normalization of the count rate R, the number of ions N_i and the electron density n_e were measured for every single measurement step as well. The absolute rate coefficient $\alpha(E_{\text{CM}})$ is then given by

$$\alpha(E_{\text{CM}}) = \langle \sigma v \rangle = \frac{RU}{(1-\beta_e\beta_i)LN_i n_e}$$

where $L = 250$ cm is the overlap length of electron and ion beam and $U = 108.36$ m the ring circumference. The measured $\alpha(E_{\text{CM}})$ (Fig. 3 for the KLL group) is a convolution of the cross section σ with the relative electron-ion velocity distribution $f(v)$. Besides the temperatures of the electron beam $T_\perp$ and $T_\parallel$ and the momentum spread of the ion beam $\delta p/p$ this experimental response function $f(v)$ contains contributions from the finite length of the drift tube ($L_d = 194$ cm), fringing field effects at both its ends, and the change of interaction angle in the merging and demerging section of the cooler. The latter three contributions depend on the individual combination of U_c and U_d and are well understood [4,19]. Resonances of individual groups overlap even within their natural line widths (Fig. 2). Therefore, the resonance positions E_{res} were determined as the centroid of the group and the strength S from direct integration of the measured $\alpha(E_{\text{CM}})$. The experimental response function was taken into account and corrected for by comparison of E_{res} and S with results of a Monte Carlo convolution of resonances of unit resonance strength [4]. This procedure is chosen in preference over deconvolution since the latter is prone to introducing artifacts, shifts in energy, and changes in normalization. However, for display in the overview spectrum in Fig. 1, in order to provide the same ordinate scale for the different data sets, drift tube and merging effects were unfolded from the data.

The uncertainty of the experimental energy scale stems mainly from uncertainties of the voltages U_d and U_c and from the uncertainty of the ion velocity. An inherent improvement of the energy determination was achieved using different combinations of U_d and U_c that lead to identical energies in the c.m. frame. By employing this procedure the systematic error for the determination of resonance positions is reduced to less than 0.05%. The error budget of the absolute rate coefficient comprises ±5% uncertainty from ion current determination, ±10% for the electron density, and −5% to 0 for the detection efficiency. For the resonance strengths S an additional ±5% arise from the inclusion of the experimental response function $f(v)$ resulting in a total systematic error for S of 13%.

Experimental data were taken for 63 keV $\leqslant E_{\text{CM}} \leqslant$ 90 keV thus covering all U^{91+} KLL and KLM resonance groups (Fig. 1). For the KLL manifolds the unaltered experimental DR rate coefficient is shown in Fig. 3 together with a Monte Carlo simulation based on the present theoretical result (Table I and Fig. 2). Experimental resonance positions and strengths are listed in Table II as well as the corresponding theoretical results. The effect of the GBI is immediately evident for the $KL_{1/2}L_{1/2}$ manifold and mainly caused by the $[2s_{1/2}2p_{1/2}]_0$ configuration, but it is also clearly visible in the $[2s_{1/2}^2]_0$ resonance in the shoulder of the $KL_{1/2}L_{1/2}$ group. Most experimental and theoretical resonance strengths

TABLE II. Measured and calculated (C + B = Coulomb + Breit, C = Coulomb only) U^{91+} KLL, and KLM-DR resonance energies E_{res} and strengths S. The experimental uncertainties (in parentheses) comprise statistical errors only. Systematic errors are 0.05% for E_{res} and 13% for S. The MCDF theoretical resonance energies in this table are given as weighted averages for a given group. Experimental values for the weak $KL_jM_{5/2}$ groups could not reliably be obtained.

	Experiment		MCDF C + B		MCDF C	
Resonance group	E_{res} (eV)	S (kb eV)	E_{res} (eV)	S (kb eV)	E_{res} (eV)	S (kb eV)
$KL_{1/2}L_{1/2}$	63 954(1)	58.7(11)	63 961	61.7	63 966	34.5
$KL_{1/2}L_{3/2}$	68 513(16)	33.1(11)	68 488	35.5	68 495	34.2
$KL_{3/2}L_{3/2}$	73 019(29)	9.1(14)	73 005	8.69	72 998	7.27
$KL_{1/2}M_{1/2}$	83 284(8)	18.9(9)	83 284	21.4	83 288	11.9
$KL_{1/2}M_{3/2}$	84 628(21)	8.0(4)	84 596	8.46	84 591	7.91
$KL_{1/2}M_{5/2}$			84 944	1.17	84 947	0.90
$KL_{3/2}M_{1/2}$	87 792(24)	5.6(16)	87 782	5.41	87 783	5.29
$KL_{3/2}M_{3/2}$	89 162(23)	4.0(6)	89 146	4.62	89 144	3.95
$KL_{3/2}M_{5/2}$			89 482	0.77	89 481	0.66

agree well within the combined statistical and systematic experimental uncertainty. For the strongest resonance group, i.e., for the $KL_{1/2}L_{1/2}$ resonances the agreement between experiment and theory is within 5%, but only if GBI is taken into account. Without GBI the theoretical $KL_{1/2}L_{1/2}$ resonance strength is significantly lower (∼60%) than the experimental value (Table II). A similar situation is found for the $KL_{1/2}M_{1/2}$ resonance group. These findings directly reveal the decisive influence of non-Coulombic contributions to the electron-electron interaction of very heavy atomic ions. According to our calculations, the GBI contribution to the $KL_{1/2}X_{1/2}$ ($X = L,M$) DR resonance strengths amounts to 44% (Table II). Other resonance groups are much less affected by GBI since they involve electronic orbitals with higher angular momenta.

In summary, resonant recombination of H-like U^{91+} ions has been experimentally observed at a heavy ion storage ring. Resonance strengths were measured on an absolute scale, thus providing a benchmark of the Auger rates on a high significance level. The comparison of experiment and state-of-the-art relativistic calculations shows directly and unambiguously that the Breit interaction, depending on the configuration, makes a major contribution to the Auger rates. The measurement was rendered possible by our technique with a stochastically cooled ion beam. The energy range of electron-ion collisions processes that can be covered using this approach is greatly extended and spans $0 \leqslant E_{\rm CM} \lesssim 200$ keV. In addition, since intermittent electron cooling is omitted, the beam storage is essentially loss-free and leads to storage times of the order of several hours to days. We will further exploit and improve the potential of the method in upcoming experiments, e.g., by introducing continuous stochastic cooling of the ion beam. This will allow collision experiments at a very low energy spread (<0.1 eV at $E_{\rm CM} = 1$ eV and <10 eV at $E_{\rm CM} = 100$ keV) with a collection efficiency and a duty cycle close to one. In the long run this opens new opportunities for the investigation of processes that were previously rarely studied or even inaccessible: (i) The energy resolution is sufficient to test line shapes and line widths of the KLL resonances. One can thereby probe the radiative lifetime of the levels, QED of overlapping DR resonances, and DR-RR interference effects [6]. (ii) Electron impact ionization of arbitrary ions and charge states up to U^{91+} and energies up to 200 keV can be measured. More exotic processes such as nuclear excitation by electron capture come into reach (for a possible scenario see Ref. [20]). (iii) By employing the DR technique in combination with the prolonged beam storage one gets unique access to the long lifetimes of hyperfine transitions of highly charged ions, of atomic metastable states and their hyperfine quenching [21], as well as of nuclear metastable states [22].

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