Electron- and Proton-Impact Excitation of Hydrogenlike Uranium in Relativistic Collisions

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The K shell excitation of H-like uranium (U^{91+}) in relativistic collisions with different gaseous targets has been studied at the experimental storage ring at GSI Darmstadt. By performing measurements with different targets as well as with different collision energies, we were able to observe for the first time the effect of electron-impact excitation (EIE) process in the heaviest hydrogenlike ion. The large fine-structure splitting in H-like uranium allowed us to unambiguously resolve excitation into different L shell levels. State-of-the-art calculations performed within the relativistic framework which include excitation mechanisms due to both protons (nucleus) and electrons are in good agreement with the experimental findings. Moreover, our experimental data clearly demonstrate the importance of including the generalized Breit interaction in the treatment of the EIE process.

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Electron-impact excitation (EIE) of bound electrons is one of the most fundamental processes and leads to the specific formation of spectral lines. In particular, it is responsible for the vast majority of x-ray radiation produced in various kinds of plasmas, in high energy density physics experiments, and at laboratory fusion devices. Relativistic and retardation effects are known to affect the EIE process through the generalized Breit interaction (GBI) [1,2]. The GBI can be derived as a lowest-order quantum electrodynamics (QED) correction from the Feynman diagram representing the interaction between two electrons [3]. Theorists have employed the GBI in order to provide more accurate calculations of atomic structure and of a variety of fundamental processes. Some recent examples include the experimental verification of the importance of the GBI in the process of dielectronic recombination for heavy H- and Li-like ions [4–6], and the successful reinterpretation of the polarization of Lyman-α x-ray emission in mid-Z hydrogenic ions [7].

Up to now, electron beam ion traps (EBITs) have been the preferred tool for studying the EIE. Because of the small electron-impact ionization and excitation cross sections for heavy highly charged ions, the focus of most of these EBIT studies has been confined to relatively low-Z systems. However, already for hydrogenlike ions such as argon, titanium, and iron, previous experiments using EBITs [8] have shown the need to include the generalized Breit interaction in the calculations [7,9]. Yet, the influence of the relativistic effects is largest for high-Z ions. As an example, for hydrogenlike uranium, the inclusion of the GBI has been shown to modify the cross section for electron-impact ionization by almost a factor of two [1,10]. It is also predicted to impact the linear polarization of the Lyman-α_1 spectral line [7].

Compared to ionization, excitation is mediated by the same interaction mechanism, but the bound electron is excited into a bound state of the ion and not into the continuum. Therefore, a much better experimental control can be expected by measuring the deexcitation photons which, in turn, allows for more thorough testing of corresponding theories. However, the predicted strong influence of the relativistic effects on the process of electron-impact excitation in the heaviest highly charged ions [3,9] has remained experimentally unexplored up to now.

Collisions between a highly charged ion and a light atom resulting in excitation of the ion are often characterized by momentum transfers that are much larger than the typical momenta of the atomic electrons. Under such conditions, the atoms can be regarded as a source of (quasi)free electrons and the nucleus, which act incoherently in the collision process [11,12]. This has been nicely demonstrated and utilized to study the ionization of light ions in collisions with neutral atoms in a series of experiments [13–16]. As a result, the cross sections for projectile excitation by the target nucleus and the electrons scale as Z_T^2 and Z_T, respectively (Z_T being the target atomic number). Therefore, for the H_2 target, the relative contribution of the electrons in the excitation process is largest. This argument applies also to molecular hydrogen (H_2). Due to the large distance between the two protons in a hydrogen molecule the molecular effects on projectile excitation are negligible, for the case of randomly oriented molecules (unpolarized target) [11,17].

The GSI heavy-ion accelerator and storage ring facility provides very favorable conditions for studying various elementary processes occurring in relativistic collisions involving high-Z ions [18]. Here, in particular for the case of projectile electron excitation by the electromagnetic field of the target nucleus (*Coulomb excitation*), several experiments have already been conducted for H- and He-like Bi and H-like Au ions [19–21]. In Ref. [22], the nuclear-field-induced excitation of H- and He-like uranium ions was studied. A markedly different behavior observed for the two systems could be explained by rigorous relativistic predictions emphasizing the importance of the magnetic-interaction and many-body effects in the strong-field domain. Here, we would like to note that in these experiments, targets of C and of heavier elements were used, which makes the excitation due to the target nucleus a dominant process.

In this Letter, we present an experimental and theoretical study of the electron-impact excitation effects in hydrogen-like uranium. Recent developments, such as the anticoincidence mode [21] and new microdroplet target development [23], have rendered such studies feasible. In order to gain access to the EIE effects, we performed measurements with different targets, namely H_2 and N_2, as well as with different collision energies, 212.9 and 393.9 MeV/u. These energies were chosen to be near and well above the EIE threshold. By performing measurements with the different targets and different energies, we were able to change the relative contribution of the two processes, the proton or nucleus impact excitation (PIE) and the EIE, which allows for a consistent test of the corresponding theories. In addition, the large fine-structure splitting in H-like uranium allows us to unambiguously resolve excitation into different L shell levels. This experimental information enables a stringent and detailed test of the state-of-the-art relativistic calculations which include excitation mechanisms due to both protons (nucleus) and electrons.

The experiment was performed at the experimental storage ring (ESR) in GSI Darmstadt. H-like uranium ions were delivered by the heavy ion synchrotron (SIS) and stored in the ESR. An efficient electron cooling provided beams with very low emittance (beam size of less than 5 mm) and a longitudinal momentum spread of $\Delta p/p \sim 10^{-4}$–10^{-5} which enabled storage of the beam with long lifetimes as well as a decrease of the uncertainties due to the relativistic Doppler effect. After injection into the ring and subsequent electron cooling, the ion beam interacted with a droplet target beam [23] of H_2 or N_2 molecules. For the experiment, the atomic physics photon detection chamber at the internal target of the ESR was utilized. Here, projectile x rays produced in collisions of the stored ion beams with the jet target were detected by four Ge(i) semiconductor detectors, mounted at observation angles of 35°, 90°, 120°, and 150° with respect to the beam axis. The photon detectors were energy and efficiency calibrated using a set of appropriate radioactive sources. In addition, those projectile ions that captured an electron were detected after the next dipole magnet of the ESR with a multiwire proportional counter. A detailed description of the detection setup at the ESR jet target and of the utilized anticoincidence technique can be found in Refs. [21,24] and in references therein.

As an example, we depict in Fig. 1 x-ray spectra recorded for $U^{91+} \rightarrow H_2$ collisions at the observation angle of 35° with respect to the ion beam direction. In the total or raw spectrum, the characteristic transitions arising from both electron capture ($K\alpha$ transitions in He-like uranium) and from excitation (Lyα transitions in H-like uranium) are clearly visible. Here, $K\alpha_1$ and $K\alpha_2$ denote 2^1P_1, $2^3P_2 \rightarrow 1^1S_0$ and 2^3P_1, $2^3S_1 \rightarrow 1^1S_0$ transitions, respectively, whereas Lyα_1 and Lyα_2 correspond to $2p_{3/2} \rightarrow 1s_{1/2}$ and $2p_{1/2}$, $2s_{1/2} \rightarrow 1s_{1/2}$. Applying a coincidence condition with respect to the down-charged projectile (U^{90+}), we obtain the characteristic x-ray spectra corresponding exclusively to the events of capture of one electron from the target into initially H-like uranium (U^{91+}). Furthermore, by subtraction of the spectrum corresponding to the capture channel from the total one, we obtain the spectrum in anticoincidence with the projectile charge exchange which comprises only the events corresponding to $K \rightarrow L$ excitation and following decay (Lyα_1 and Lyα_2) of the projectile electron. Indeed, no $K\alpha$ transitions were

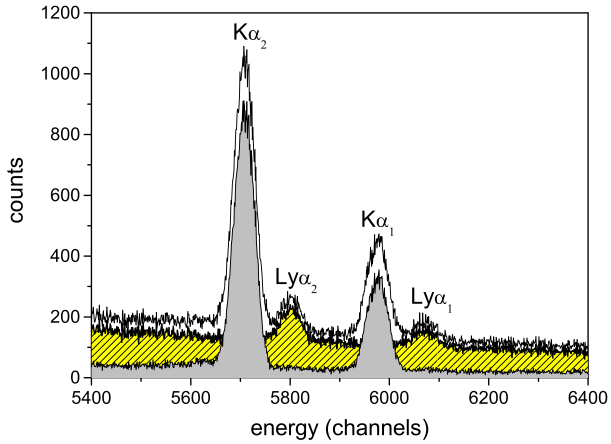

FIG. 1 (color online). X-ray spectra recorded for 212.9 MeV/u $U^{91+} \rightarrow H_2$ collisions with a Ge(i) detector at the observation angle of 35° with respect to the ion beam: (solid line) total emission spectrum without coincidence requirement; (gray-filled area) photons in coincidence with electron capture, $L \rightarrow K$ transitions in He-like uranium; (yellow-filled area with stripes) photons in anti-coincidence with electron capture, $L \rightarrow K$ transitions in H-like uranium.

observed in the anticoincidence spectrum. Here, we also would like to note that multiple electron capture contributions are negligible for our collision systems and energies and average target areal densities of $\sim 10^{13}$ cm^{-2} [25].

In the following, we focus on two observables obtained from the experimental data, namely, intensity ratios of the Lyα_1 and Lyα_2 transitions and angular distributions of the Lyα_1. This has been proven to be a highly sensitive probe for modern theories describing collision dynamics and the structure of high-Z ions [24,26]. In our analysis, we exploit the fact that the Lyα_2 transition arising from the decay of the $2s_{1/2}$ and $2p_{1/2}$ levels is known to be isotropic in the projectile frame [26]. Therefore, this line provides a tool to measure a possible anisotropy of the neighboring Lyα_1 transition. By using the Lyα_2 transition for normalization purposes, various systematic effects, associated, for example, with solid angle corrections, possible error in detector efficiency calibration, etc., cancel out or are substantially reduced. This technique has already been successfully applied in several previous studies [22,27,28]. The number of counts recorded in the Lyα_2 and Lyα_1 spectral lines were determined by fitting the corresponding peaks in the spectra with Gaussian functions and taking the bremsstrahlung background into account. Afterwards, the ratios were corrected for the relative detector efficiency. For this correction, an error of 3% is included. The Lyα_1/Lyα_2 ratio as a function of the observation angle is described by the following formula [26,27]:

$$W(\theta_{\rm lab}) = A_0\left[1 + \frac{\mathcal{A}_2}{2} f(E1, M2) \times \left(1 - \frac{3}{2}\frac{\sin^2\theta_{\rm lab}}{\gamma^2(1-\beta\cos\theta_{\rm lab})^2}\right)\right], \quad (1)$$

where $\theta_{\rm lab}$ is the angle between the direction of the deexcitation photon and the beam direction. β and γ are the relativistic factors corresponding to the particular projectile energy. The alignment parameter $\mathcal{A}_2$ quantifies the amount of nonstatistical population of different magnetic substates and it is defined as

$$\mathcal{A}_2 = \frac{\sigma(\frac{3}{2}, \pm\frac{3}{2}) - \sigma(\frac{3}{2}, \pm\frac{1}{2})}{\sigma(\frac{3}{2}, \pm\frac{3}{2}) + \sigma(\frac{3}{2}, \pm\frac{1}{2})}. \quad (2)$$

Here, $\sigma(j\mu)$ are the partial cross sections for populating the magnetic substates $j\mu$. The so-called structure function $f(E1, M2)$ describes the interference between the leading $E1$ and the much weaker $M2$ decay channels and contributes by as much as $f(E1, M2) = 1.28$ to the angular distribution of the characteristic photon emission from H-like uranium ions [26]. A_0 gives the Lyα_1/Lyα_2 ratio at the "magic angle," i.e., the angle for which the angle-dependent part of Eq. (1) is zero. Then, by fitting the Lyα_1/Lyα_2 ratios with the angular distribution formula (1), experimental values for the ratio A_0 and the alignment parameter $\mathcal{A}_2$ are obtained. In Fig. 2, we show results of this procedure for 393.9 MeV/u collision energy. From the figure, one can see that the angular pattern for the two targets is very similar, whereas the ratios differ. This change of ratios can be seen as a direct indication of the EIE process which is expected to play a more pronounced role for the H_2 target.

In Fig. 3, we present results obtained for the Lyα_1/Lyα_2 ratios (A_0) for both collision energies and targets. The experimental uncertainties comprise contributions due to statistics and the detector efficiencies. The theoretical predictions include both excitation channels, the PIE and the EIE assuming (quasi)free electrons. This approximation should be well justified for the current case of large

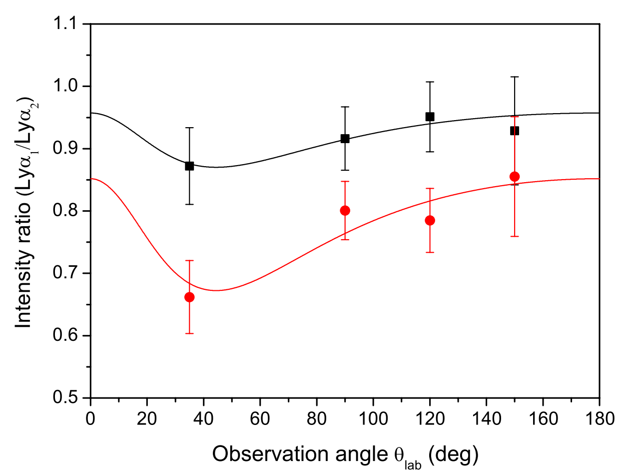

FIG. 2 (color online). The intensity of Lyα_1-transition normalized to the Lyα_2 line intensity as a function of the observation angle for 393.9 MeV/u U^{91+} collisions with N_2 (black squares) and H_2 targets (red circles). The solid lines refer to corresponding fits of Eq. (1) to the data (see also text).

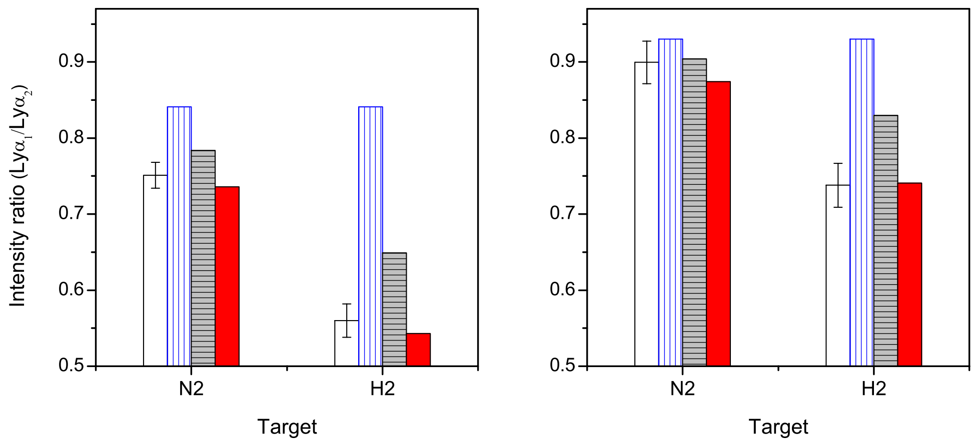

FIG. 3 (color online). Experimental results (white columns) in comparison with theoretical predictions for $Ly\alpha_1/Ly\alpha_2$ ratios (A_0) for the K shell excitation of U^{91+} in collisions with N_2 and H_2 targets at 212.9 (left) and 393.9 MeV/u (right). Blue columns with vertical stripes show PIE results. Solid red columns depict combined (PIE + EIE) calculations. In addition, the combined calculations are presented without inclusion of the GBI, by gray columns with horizontal stripes.

momentum transfer collisions of relativistic heavy ions on neutral gas atoms [12]. The effect of the Compton profile of the bound electrons has been estimated to be negligible on the current level of the experimental precision. The EIE cross section calculations are based on a relativistic distorted-wave approach [9,12] and include the effects of the GBI. In addition, the calculations include excitation to higher levels ($n \geq 3$) and subsequent cascade contributions to the observed Lyman radiation.

From our experimental results (see Fig. 3), it is obvious that the $Ly\alpha_1/Ly\alpha_2$ intensity ratios (A_0) are significantly smaller for the H_2 target as compared to the N_2 case. As can be clearly seen from Fig. 3, this effect can not be described by the PIE theory, because, here, the ratios are independent of the target atomic number. The deviation is especially pronounced for the H_2 target, where the EIE contributes strongly. However, it is also important to emphasize that even for the case of the N_2 target, where the nuclear contribution can be expected to be dominant, our experimental results for the beam energy of 212.9 MeV/u (closer to the EIE threshold) deviate from the PIE predictions by more than 4σ. In contrast, the full calculations which include the effect of EIE together with the PIE, are in very good agreement with the experimental data within the error bars. The reduction of the intensity ratios (A_0) when going from the N_2 to the H_2 target is mainly due to the significantly larger EIE cross sections for the $2s_{1/2}$ state as compared to the corresponding PIE values. For the collision energy of 212.9 MeV/u, the EIE cross section (including the GBI) exceeds the corresponding PIE cross section by more than three times [29].

Considering the fact that the PIE calculations have already been tested in few previous studies [19,21], the current results can be regarded as a clear identification and test of the EIE excitation effects in H-like uranium in the relativistic collisions. Moreover, our experimental results can not be reproduced by the full calculations which do not take into account the effect of GBI in the EIE process (see Fig. 3). This clearly demonstrates the great importance of inclusion of the GBI in the EIE calculations for H-like uranium.

In the case of alignment (A_2), our experimental uncertainties are too large to distinguish between only PIE and the combined (PIE + EIE) calculations.

In summary, we have measured K shell excitation of H-like uranium (U^{91+}) in collisions with N_2 and H_2 targets at 212.9 and 393.9 MeV/u energies. By looking at the intensity ratios of the subsequent decay photons ($Ly\alpha_1/Ly\alpha_2$), we were able to clearly identify and study the effect of the electron-impact excitation in H-like uranium. Combined calculations which treat both processes, PIE and EIE, provide a good agreement with the experimental data. Moreover, the experimentally determined intensity ratios clearly demonstrate the importance of including the effect of the GBI in the EIE calculations. As a next step, it would be desirable to increase the experimental precision for the alignment of the $2p_{3/2}$ states and/or to measure polarization of the $Ly\alpha$ radiation [28]. This would allow us to test the theory even further by investigating the magnetic sublevel population mechanisms by the EIE process.

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