Dielectronic and Radiative Recombination of Lithiumlike Gold

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We report the first measurements on radiative and dielectronic recombination (RR and DR) of very highly charged ions at energies $E_{\rm c.m.}$ as low as 0 to 50 eV. Novel techniques were employed at the heavy-ion storage ring ESR to obtain absolute recombination rates of $Au^{76+}(1s^22s)$. The increase of the RR rate for $E_{\rm c.m.}\rightarrow 0$ could be recorded and single DR resonances in $Au^{75+}(1s^22p_{3/2}6l_j)$ resolved. The experimental data allow sensitive judgement of calculations based on the Bethe-Salpeter approach to RR combined with results for DR from perturbative-relativistic, semirelativistic, and fully relativistic theories. Significant discrepancies are found only in the energy region of the $2p_{3/2}6l_{3/2}$ resonances.

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Since the very first direct observations of electron-ion recombination in colliding-beam experiments nine years ago the field has seen tremendous experimental progress [1]. Now, after demonstrating the capability of the GSI experimental storage ring (ESR) [2] to store and cool even the heaviest highly charged ions, a new window has been pushed open towards electron-ion collision phenomena in a regime of charge states and energies that has not been accessible before. In this Letter we report the first successful experiment carried out at the ESR. For Li-like Au^{76+} ions we present experimental and theoretical data on radiative recombination (RR),

$$Au^{76+}(1s^22s)+e\rightarrow Au^{75+}(1s^22s\,nl)+\text{photon}\,, \quad (1)$$

and dielectronic recombination (DR) with $\Delta n=0$ core excitations,

$$Au^{76+}(1s^22s)+e\rightarrow Au^{75+}(1s^22p_{1/2,3/2}nl)^{**} \rightarrow Au^{75+}+\text{photons}\,. \quad (2)$$

While RR is a direct process, inverse to photoionization, DR proceeds through the formation of an intermediate doubly excited state in a radiationless transition, inverse to autoionization, with subsequent stabilization of the new ion charge state by photoemission.

Measurements based on the observation of resonant transfer and excitation (RTE) of He-like U^{90+} in molecular hydrogen [3] have provided a benchmark test of relativistic calculations for DR [4,5]. The sensitivity of this test, however, is limited by the center-of-mass energy spread (in the target-electron, projectile-ion system) which is of the order of a keV in these RTE measurements due to the Compton profiles of bound target electrons. Previous merged-beam experiments with electrons and ions at heavy-ion storage rings (see, e.g., [1,6]) or with single-pass electron-target arrangements at accelerators (see, e.g., [1,7]) have provided very good energy resolution but were restricted to ion charge states of at most $q=28$. For ions in higher charge states up to Ne-like gold, electron-ion collisions have been studied using electron-beam ion traps [1,8]. However, cross sections obtained there for DR are not absolute and usually involve contributions from several parent ion charge states with an unknown fractional distribution. The lowest energies are in the range of keV and the collision energy spread is typically 50 to 100 eV. Recently a trap experiment was reported for Ne-like Xe^{44+} ions [9] using improved techniques. Relative cross sections were measured for $\Delta n=1$ core transitions with a collision energy spread of 16 eV at energies $E_{\rm c.m.}\geq 1$ keV. In contrast to that, our storage ring experiment provided absolute data and allowed us to access $\Delta n=0$ core transitions at energies $E_{\rm c.m.}\geq 0$ eV with longitudinal and transverse beam temperatures $T_{\parallel}=0.004$ eV/k and $T_{\perp}=0.4$ eV/k, where k is the Boltzmann constant.

The ESR is fed from the heavy-ion synchrotron (SIS) which has provided so far heavy ion beams with energies up to 1 GeV/u. Ions like Bi^{82+} (H like) were successfully stored and cooled in the ESR at energies of about 200 MeV/u with half lives of the order of 1000 s depending on the cooler electron current. For the present experiment we chose to study Li-like ions because they are the species with the fewest core electrons providing resonances at low energies. The direct production of the desired Li-like Au^{76+} ions in a stripper would have been favorable at ion energies between 50 and 100 MeV. However, better beam emittance and easier beam handling at higher ion energies together with the duty-factor advantage of the "breeding technique" described below led us to the decision to start with an intense beam of He-like Au^{77+} ions at 230 MeV/u. In order to provide the desired Li-like Au^{76+} ions for the experiment we first stacked SIS pulses of Au^{77+} in the ring, accumulating typically a total current of about 40 μA during 30 min. The orbit of the Au^{77+} ion beam was positioned in such a way that the ring acceptance allowed for a second, simultaneously stored beam of Au^{76+}. For our measurements the ESR cooler [10] served three purposes at a time. It was used to breed the desired Au^{76+} ions from Au^{77+} by electron-ion recombination; it kept the ion beam cold and stable in energy; and it served as an electron target providing defined energies $E_{\text{c.m.}}$ in the electron-ion center-of-mass frame.

While the first two tasks can be served best when the ion and electron velocities are matched, i.e., $E_{\text{c.m.}}=0$ eV, the latter requires that the energy of the electrons of the cooler be offset from the cooling energy. Such detuning, however, inevitably invokes strong traction forces on the stored highly charged ions. These forces counteract any change in the relative velocity between the electrons and the ions. Within a short time, depending on the electron density and the degree of energy detuning, the velocities of electrons and ions are equilibrated again with the result that $E_{\text{c.m.}}$ is back to zero. Two independent methods were employed to defy the traction forces.

The potential along the ESR cooler axis is determined by two separate drift tubes. Each of the drift tubes can be set to potentials between 0 and -5 kV. Thus it is possible to leave one section of the cooler on cooling potential ($U_1=0$ V), while in the other section the electron energy is varied ($U_2=0$ to -5 kV). In addition, the voltage U_2 was switched off every 20 ms for a 20-ms cooling period. In all, U_2 was ramped through 2300 discrete settings within about 90 s covering a range $E_{\text{c.m.}}=0$–50 eV in the electron-ion center-of-mass frame and then the whole cycle was repeated. The frequency spectrum which represents the momentum distribution of the circulating ions was monitored by Fourier analysis of the Schottky noise of the ion beam. In spite of the permanent ion beam cooling in the longer first drift tube and additional intermittent cooling in the second drift tube we observed shifts of the average revolution frequency of the ion beam with changing U_2. However, these shifts were so small that the ion beam momentum spread $\Delta p/p$ was kept below 4×10^{-5} including excursions from the average frequency of the cooled ion beam.

The Au^{75+} recombination products were collected by a position-sensitive multiwire proportional counter behind the first dipole magnet downbeam from the cooler. The detector rate dropped approximately by a factor of 200 when the cooler was switched off, indicating negligible background from processes other than electron-ion collisions. The experimental counting rate R_{75} of recombined Au^{75+} ions was accumulated and recorded every 2 ms and also at every change of the voltage U_2 along with information on the instantaneous ring current, energy of the electrons, and corresponding time interval. For the evaluation of recombination rates, time intervals of the order of 4 ms, during which the electron energy was badly defined as a consequence of the finite slew rate of the high-voltage power supply, were excluded from the final data analysis. Normalized recombination rates α_{expt} were determined from

$$\alpha_{\text{expt}}=C\gamma^2R_{75}/n_eN_{76}L\,, \tag{3}$$

where $C=108.4$ m is the circumference of the ring, $\gamma=(1-\beta^2)^{-1/2}$ the Lorentz factor, n_e the electron density in the cooler, N_{76} the number of parent ions stored in the ring, and $L=2.50$ m the interaction length. In the present experiment γ is 1.25. The counting rate R_{75} of recombined Au^{75+} ions is totally determined by RR in the longer drift tube and by U_2-dependent RR and DR in the shorter tube.

With different voltages U_1 and U_2 on the drift tubes of the cooler and the resulting probability distribution $dl(E',E_{\text{c.m.}})/dE'$ along the cooler axis, where U_2 corresponds to $E_{\text{c.m.}}$, the observed recombination rate is a convolution product:

$$\alpha_{\text{expt}}(E_{\text{c.m.}})=\frac{1}{L}\int_0^{E_{\text{c.m.}}}\frac{dl(E_{\text{c.m.}},E')}{dE'}\alpha(E')dE'\,, \tag{4}$$

with

$$\int_0^{E_{\text{c.m.}}}\frac{dl(E_{\text{c.m.}},E')}{dE'}dE'=L\,. \tag{5}$$

The electron density in the laboratory frame n_e was about 8×10^6 cm^{-3}. This number results directly from the measured electron current (480 mA), the electron velocity ($\beta=0.6$), and the diameter of the electron beam (5 cm) which is determined by the size of the cathode and the fact that the electron beam is guided by a strong magnetic field. Uniform current density in the electron beam was tested by measurements of the cooling force as the ion beam (few mm diameter) was displaced within the electron beam parallel to the beam axis. No irregularities were found in the cooling force which would have indicated nonuniform electron density. Hence, we conclude that n_e is determined correctly within $\pm5\%$. The total ion current $I_{\text{tot}}=I_{77}+I_{76}$ circulating in the ring was measured by a transformer pickup. The counting rate of R_{75} ions averaged over one complete cycle of ramping U_2 is proportional to the instantaneous ion current I_{76} of Au^{76+} parent ions. Since electron-ion recombination has been shown to be the dominant loss mechanism for ions in a given charge state, the observed time dependences I_{tot} and R_{75} can be described as in a simple mother-daughter radioactive decay of nuclei. By fitting solutions of the corresponding differential equations to the experimental functions $I_{\text{tot}}(t)$ and $R_{75}(t)$, average recombination rates for Au^{77+} and Au^{76+} could be deduced and the instantaneous current and number N_{76} of Au^{76+} ions in the ring calculated with an uncertainty of $\pm15\%$. The current I_{76} increased to about 10 μA within 15 min after the stacking procedure was stopped, and, as a consequence of the breeding technique, stayed within 3-10 μA for up to 3 h off a single stack in the ring. Normalized experimental recombination rates $\alpha_{\rm expt}$ obtained from Eq. (3) are displayed in Fig. 1. Statistical uncertainties are typically 3%; the total uncertainty amounts to about 20%. The experimental energy calibration is estimated to be correct within 0.5 eV.

Energy-averaged DR cross sections were calculated in the isolated-resonance approximation [11]. For DR of Li-like Au^{76+}, we consider the reaction pathways indicated by Eq. (2) with $n \geq 20$ for the $2s_{1/2} \rightarrow 2p_{1/2}$ core excitation and $n \geq 6$ for the $2s_{1/2} \rightarrow 2p_{3/2}$ core excitation. Since the ESR measurements range from 0 to 50 eV, it is important to obtain the highest possible accuracy on the core energy splittings. The core excitation energies $\Delta_1 = E(2p_{1/2}) - E(2s_{1/2}) = 217$ eV and $\Delta_2 = E(2p_{3/2}) - E(2s_{1/2}) = 2244$ eV were calculated for the target ion Au^{76+} using the multiconfiguration Dirac-Fock atomic structure code of Grant *et al.* [12], including nuclear size effects, Breit interaction, and QED corrections [13].

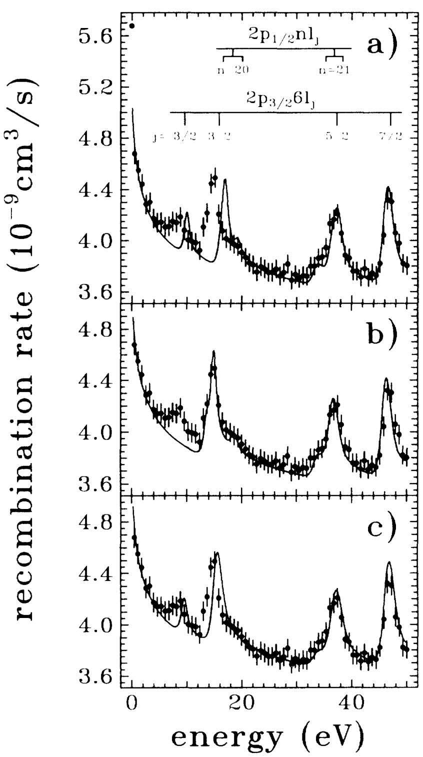

FIG. 1. Comparison of convoluted theoretical recombination rates (see text) and experimental data for recombination of Au^{76+} ions. Energetic positions expected on the basis of the fully relativistic theory are indicated in (a) for intermediate resonances $2p_{3/2}6l_j$ with $j = \frac{3}{2}$, $\frac{5}{2}$, and $\frac{7}{2}$ as well as for $2p_{1/2}nl_j$ resonance groups with $n = 20$ and 21. In all cases the contribution from RR is obtained from a modified Bethe-Salpeter formula [20]. Theoretical DR rates are displayed for $2p_{1/2}6l_j$ resonances only. They were obtained from three different theoretical approaches: (a) perturbative-relativistic, (b) semirelativistic, and (c) fully relativistic. Note that the modeling of theory to experiment according to Eq. (4) does not conserve resonance peak areas.

We carried out three different types of intermediate-coupling atomic structure calculations for the resonance energies, autoionization decay rates, and radiative decay rates needed for the calculation of cross sections. For highly charged atomic ions, like Au^{76+}, the most important relativistic effects for these nl_j resonance parameters are from the one-body kinetic-energy correction, spin-orbit, and Darwin terms. Breit interaction and QED corrections can be safely ignored, except for excitations from the K shell, which are not present in this case. The first method [14,15], which we call perturbative-relativistic, solves standard radial Schrödinger equations for all bound and continuum orbitals and then diagonalizes a Hamiltonian which includes the kinetic energy, electrostatic potential energy, and one-body relativistic terms to obtain resonance energies and wave functions. First-order many-body perturbation theory is then applied to obtain autoionization and radiative rates using the previously calculated energies and wave functions. The second method [5], which we call semirelativistic, uses the radial Schrödinger equations including the relativistic kinetic energy correction and Darwin terms [16] and a Hamiltonian with the kinetic energy, electrostatic potential energy, and spin-orbit terms. Finally, the third method [17-19], which we call fully relativistic, solves the radial Dirac-Fock equations for all bound and continuum orbitals, including in a nonperturbative manner all the one-body fine-structure effects. This method required an extensive modification of the atomic structure codes of Grant *et al.*

The semirelativistic method was used to calculate the DR cross section for Au^{76+} from 0 to $\Delta_1 = 217$ eV for the $2p_{1/2}nl_j$ resonances and from 0 to $\Delta_2 = 2244$ eV for the $2p_{3/2}nl_j$ resonances. In the energy range of the experiment, from 0 to 50 eV, only resonances from the $2p_{1/2}20l_j$, $2p_{1/2}21l_j$, and $2p_{3/2}6l_j$ subconfigurations are found. The $2p_{1/2}20l_j$ resonances are located at 17.0 eV, and the $2p_{1/2}21l_j$ resonances are at 35.5 eV. When the DR cross section in the experimental energy range is folded with a 1-eV FWHM Gaussian, the $2p_{1/2}20l_j$ peak cross section is around 6.0 Mb, the $2p_{1/2}21l_j$ peak cross section is around 2.5 Mb, while several $2p_{3/2}6l_j$ peaks are in the 25.0-Mb range.

The perturbative-relativistic, semirelativistic, and fully relativistic methods were used to calculate the DR cross section for Au^{76+} from 0 to 50 eV for the $2p_{3/2}6l_j$ resonances. For comparison with the experiment the theoretical DR cross sections were combined with calculated cross sections for RR, then folded with a flattened Maxwellian velocity distribution for the electron and ion beams arrangement and convoluted with the energy distribution according to Eq. (4). The necessary information on the potential distribution along the cooler axis was obtained by solving the Poisson equation for the given geometry and the applied voltages under inclusion of the electron space charge. RR and DR were treated as independent processes. Cross sections for RR were calculated from the formula given by Bethe and Salpeter [20] corrected at low quantum numbers to match the Stobbe theory [21]. Contributions from $n=2$ (first open shell in Au^{76+}) up to $n=110$ [cutoff limit for field ionization of $Au^{75+}(nl)$ in the magnetic dipole fields] were included with an effective atomic number $Z_{\rm eff}=76$. The longitudinal and transverse energy spreads fitting the experiment are characterized by $kT_{\parallel}=0.004$ eV and $kT_{\perp}=0.4$ eV. A nonzero angle between electron and ion beams corresponding to a minimum energy of $E_{\rm c.m.}=0.37$ eV instead of 0 eV was assumed in order to match the RR theory curve with the measurements.

The convoluted theoretical rates for the three different calculations of the DR contributions added to the RR contributions are compared to the experimental recombination rates in Fig. 1. The "background" of RR is perfectly modeled by using the procedures discussed above. Thus it becomes immediately visible that except for deficiencies in describing the $j=\frac{3}{2}$ structures between about 5 and 20 eV there is good agreement between all three DR theories and experiment. We found the energy position of the $2p_{3/2}6p_{3/2}$ structures to be extremely sensitive to the strength of the $6p$ spin-orbit term in the perturbative-relativistic and semirelativistic calculations. The Dirac-Fock-Breit calculation shows the correct energy splitting of the $j=\frac{3}{2}$ levels. However, it also does not completely agree with the absolute experimental energy positions. In principle, the fully relativistic theory contains the most physics and thus it is comforting to see that on the whole it is in good agreement with the experimental results. It is instructive, however, to see how well the computationally simpler perturbative-relativistic and semirelativistic theories do for such highly charged ions.

In summary, we have carried out the first recombination measurements with very highly charged ions at low $E_{\rm c.m.}$. By exploiting the capabilities of the ESR storage ring together with some novel experimental techniques, such as cooling and experimenting simultaneously and "breeding" of the desired parent ion charge state, we were able to access $\Delta n=0$ core transitions in Li-like Au^{76+} with very good energy resolution. Although different theoretical calculations show good general agreement with the experimental results, there are still some deficiencies in describing the lowest angular momentum states. The quality of the present measurements is promising with respect to an experimental assessment of influences from relativistic and QED effects such as Lamb-shift contributions to binding energies. Particularly for this end we plan to study H-like parent ions. Further measurements will also be carried out to obtain a better understanding of RR rates which depend so strongly on the electron beam temperatures and the beam alignment.

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