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Recombination measurements at low energies with Ar^{16+} and Ar^{18+} ions in a dense, cold electron target

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Recombination of multiply charged ions with electrons at very low relative energies has become a major topic of interest, due to the observation of rates which are enhanced beyond the expectations for radiative recombination. We present results for Ar^{16+} and Ar^{18+} ions from systematic measurements along the argon isonuclear sequence using a high density cold electron beam target ($n_e = 7 \times 10^9$ cm^{-3}) at the UNILAC of GSI. The transverse and longitudinal temperatures of the electron beam were determined from DR resonance features observed with metastable $Ar^{16+}(2\,^3S)$ ions. The rate at $E_{rel} = 0$ for radiative recombination of completely stripped Ar^{18+} calculated with electron beam temperatures $kT_\parallel = 0.002$ eV, $kT_\perp = 0.2$ eV amounts to $\alpha = 10^{-9}$ cm^3 s^{-1}. This is exceeded by nearly a factor of 10 by the rate measured in experiments with Ar^{18+} ions.

1. Introduction

The first quantitative direct observation of radiative recombination (RR) of ions with electrons was reported by Andersen et al. for experiments with C^{6+} ions [1]. The measured rates were in agreement with the available theory for completely stripped parent ions [2]. Unexpected high recombination rates at $E_{rel} = 0$ were first observed at the end of 1989 in an experiment with 6.3 MeV/u U^{28+} ions employing a cold dense electron target at the UNILAC accelerator of the GSI in Darmstadt [3,4]. Within an energy range between 0 and roughly 1 meV the observed recombination rate dropped from about 2×10^{-7} cm^3 s^{-1} to half this maximum value. The electron density was

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$n_e = 4 \cdot 10^8$ cm^{-3}. The electron beam temperatures were determined [5] to be $kT_\perp = 0.2$ eV and $kT_\| = 2$ meV. The cut-off by field ionization in the analyzing magnet downbeam from the electron target was at $n_F = 76$, ions up to $n_{cut} = 170$ probably decayed to states below n_F during the time of flight between the initial recombination and the arrival at the dipole field of the charge-state analyzing magnet. A realistic calculation (see ref. [6]) of the expected rate for RR of U^{28+} at a relative energy $E_{rel} = 0$ assuming an effective charge $Z_{eff} = 28$ yields $\alpha_{RR}(U^{28+}) = 0.9 \cdot 10^{-9}$ cm^{3}s^{-1}. This number is a factor of roughly 200 smaller than the experimental value.

For exclusion of experimental errors, the measurement was later repeated with 5.9 MeV/u U^{28+} ions (now $n_F = 77$, see above) and the investigated energy range was largely extended [7]. The electron density was $n_e = 5.9 \cdot 10^8$ cm^{-3} and thus slightly higher than that of the first run. The previous measurements were perfectly confirmed.

The huge discrepancies between theory and experiment found in this experiment were attributed to two possible reasons: (a) the fortuitous presence of dielectronic recombination (DR) resonances at energies below $E_{cm} = 1$ meV on top of the expected RR and (b) effects of the relatively high electron density in the target which, in that particular measurement, exceeded the electron densities in electron cooler targets of ion storage rings by roughly a factor of 10. The hypothesis about an involvement of DR was supported by the observation of a great many resonances in the experimental spectrum ranging up to about 600 eV. Some of the largest peaks in the measured spectrum range among the strongest resonance features ever observed in recombination experiments, but their maximum is yet a factor of 10 smaller than the rate observed at $E_{rel} < 2 \cdot 10^{-4}$ eV. Thus, it appeared unlikely that DR could be the sole reason for the recombination rate enhancement at low energies.

Meanwhile a number of experimental observations of rate enhancement effects have been reported for multiply charged ions and electrons with low relative energies [8–13]. And yet, the available data base is still too small to facilitate a satisfactory understanding of the new phenomenon. Therefore, the goal of the present work was to provide a set of systematic measurements with different charge states of ions which have a simple electron configuration. Since there are already data available for Ar^{13+} [14] and Ar^{15+} [10], the choice of charge states was on the sequence of helium-like, hydrogen-like, and completely stripped argon ions. The data are not finally analyzed yet. Therefore, in this progress report, we can only present preliminary results for Ar^{16+} and Ar^{18+} ions.

2. Experiments and results

Recombination of highly charged ions with free electrons has been studied in several single pass merged beams experiments [3,7,10] at the UNILAC accelerator of GSI. The experimental technique and the set-up have been described in detail previously [10]. The outstanding feature of these experiments is the high density of the employed

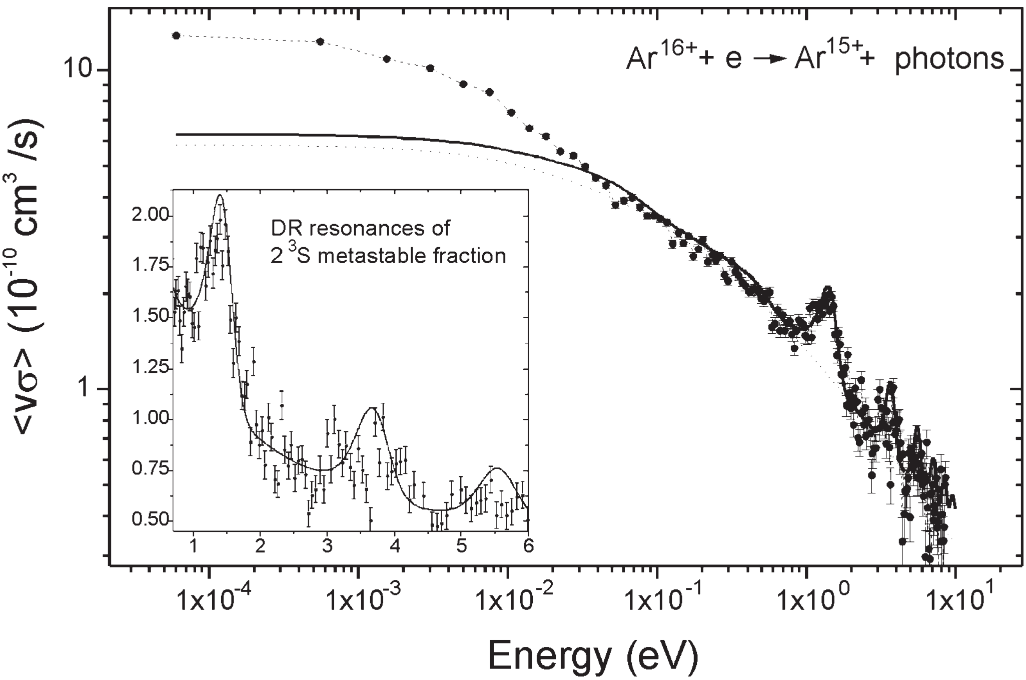

Fig. 1. Measured relative recombination rates of electrons with 7.1 MeV/u Ar^{16+} ions, vs. the relative energy in the electron–ion center-of-mass frame. An estimated fraction of 0.4% of the parent ion beam was in the $2\,^3$S metastable state. The solid line represents theoretical calculations for radiative recombination (RR) plus dielectronic recombination (DR). The dotted line is theoretical RR only. The experimental data were normalized to the calculated rate at $E_{\rm rel} = 0.1$ eV by multiplying the measured spectrum with a constant factor. The inset shows the measured and calculated rates in the range of the DR resonances arising from the metastable fraction of the parent ion beam. For more details see the text.

electron target, which can surpass the density of electron beams in storage rings by two to three orders of magnitude [15]. The electron beam temperatures at these high densities are still remarkably small. The lowest temperatures found in experiments at the UNILAC electron target facility correspond to thermal energies $kT_\perp = 0.2$ eV and $kT_\| = 2$ meV, where k denotes Boltzmann's constant.

In the present experiment a beam of 280 MeV Ar^{10+} ions from the UNILAC was passed through a stripper foil to produce the desired ion charge states $q = 16$, 17, and 18. The stripper was located about 30 m upstream from the electron target leaving a time-of-flight of nearly a microsecond for the ions between the foil and the electron target. This time is still sufficiently short to permit survival of some of the Ar^{16+} ions in the $2\,^3$S metastable state. Indeed, the experiments with helium-like argon clearly show the presence of metastable ions in the parent ion beam. This is made obvious by the resonance feature observed at an energy of about 1.5 eV in the measured recombination spectrum for Ar^{16+} which is shown in fig. 1. The electron density was about 7×10^9 cm^{-3}. The experimental data on Ar^{16+} are not yet calibrated to an absolute scale. In order to facilitate comparison of the shape of the spectrum with calculated recombination rates the available data were multiplied by a factor two. Thus, the measured and calculated rates at energies $E_{\rm rel} > 0.05$ eV are in reasonable agreement.

The energetically lowest DR resonances of ground state $Ar^{16+}(1s^2)$ ions are related to the intermediate configuration $(1s2s^2)$ occurring at a resonance energy not lower than about 935 eV. Thus the resonance feature at 1.5 eV can only be due to excited parent ions. Considering lifetimes of excited states ($\tau = 2.48$ ns for the $1s2s\,^1S$ and $\tau = 210$ ns for the $1s2s\,^3S$ metastable states [16]) and the time-of-flight (≈ 800 ns) of the ions between the stripper and the electron target, only ions in the $2\,^3S$ state can be involved in the collision process. DR via

$$e + Ar^{16+}(1s2s\,^3S) \rightarrow Ar^{15+}(1s2pn\ell) \rightarrow Ar^{15+}(1s^22s) + \text{photons} \tag{1}$$

produces resonances with $n = 15$ at relatively low energies. A theoretical calculation of the DR cross sections convoluted with the present experimental energy distribution (with temperatures corresponding to $kT_\parallel = 5.2$ meV, $kT_\perp = 0.22$ eV, which are slightly higher than “normal”) yields the correct shape of that feature. Quantitative agreement is obtained when a metastable fraction of only 0.4% in the parent ion beam is assumed. This fraction can be expected when about 20% of the parent Ar^{16+} beam are in the ^3S state right after passage of the stripper foil. With a lifetime of the $2\,^3S$ state of 210 ns [16] and an estimated time-of-flight of 800 ns the metastable fraction of 0.4% inferred from the comparison between theory and experiment can be easily rationalized. Apart from the DR resonance, fig. 1 shows a comparison of the shape of the recombination rate with calculations for RR. At relative energies $E_{\rm rel} < 0.02$ eV a peak is observed in the experimental spectrum rising to a size which is well above the expectation on the basis of the energy dependence of simple radiative recombination. The calculations are based on the Bethe and Salpeter [17] approach to RR of completely stripped ions into hydrogenic states. The formula is corrected with factors depending on the principal quantum number following the procedure used previously by Andersen et al. [2] to resemble the theoretical result of Stobbe's RR theory [18]. The contributions of the different shells to the total recombination cross section are summed taking into account the population of the lowest electron shells (for helium-like Ar^{16+} in the ground state the K-shell is filled and thus cannot contribute to RR). The summation is carried to the highest Rydberg quantum numbers of states which survive the field ionization in the analyzing magnet. Instead of the nuclear charge Z an effective charge $Z_{\rm eff} = q$ is assumed where q is the parent ion charge state. The cross section is then convoluted with the electron velocity distribution in order to obtain a rate coefficient.

The enhancement of the RR rate for Ar^{16+} ions visible at $E_{\rm rel} = 0$ is smaller than that in previous measurements for Ar^{15+} ions [9,10]. Very close to $E_{\rm rel} = 0$, the metastable Ar^{16+} ions even produce DR resonance contributions which are included in the calculation presented in fig. 1. These contributions already produce a slight rate enhancement beyond the pure RR rate. What comes to mind as a possible explanation for the smaller enhancement factor for Ar^{16+} in comparison with Ar^{15+} is the presence of the metastable ions in the parent beam: whenever an electron is captured via RR into a state with $n > 1$ the resulting recombined ion $Ar^{15+}(1s2sn\ell)$ can autoionize and hence only $n = 1$ can contribute to the total recombination rate. However, not more than a fraction of 0.4% of the parent ion beam is expected to be in a metastable state so that the difference cannot be explained on that basis. A more probable explanation is based on the alignment of the electron and ion beams. In the experiment, this alignment is controlled by two apertures of 3 mm diameter each, placed before and behind the interaction region (which is 42 cm long). The expected residual angle between the electron and ion beams is estimated to be usually much less than 1 mrad, given the ion beam emittance and the control of ion currents to the slits in case of a misalignment. In the worst case, assuming an ion beam with zero diameter which traverses the slits at a maximum angle without intensity loss, there can be a misalignment with an angle of $\theta = 3/420$. The angle θ generally results in a minimum obtainable relative energy which is greater than 0 if $\theta > 0$. By a simple classical calculation one gets

$$E_{\mathrm{rel}} = \frac{\mu}{2} v_{\mathrm{rel}}^2 = \frac{\mu}{2} \left[\vec{v}_{\mathrm{e}} - \vec{v}_{\mathrm{i}}\right]^2, \tag{2}$$

with the reduced mass $\mu = m_{\mathrm{e}} m_{\mathrm{i}}/(m_{\mathrm{e}} + m_{\mathrm{i}})$ of the electron–ion collision system and v_{rel} the relative velocity between the electrons and the ions. The electron mass m_{e} is much smaller than the ion mass m_{i} and hence $\mu \approx m_{\mathrm{e}}$. The velocities $\vec{v}_{\mathrm{e}}$ and $\vec{v}_{\mathrm{i}}$ of the electron and the ion beam, respectively, are determined by the acceleration voltages. An angle θ between $\vec{v}_{\mathrm{e}}$ and $\vec{v}_{\mathrm{i}}$ leads to a minimum relative velocity $v_{\mathrm{rel}} \approx v_{\mathrm{i}}\theta$ and to a minimum relative energy $E_{\mathrm{rel}} \approx \theta^2 E_{\mathrm{cool}}$, where E_{cool} is the electron energy corresponding to the cooling condition $v_{\mathrm{e}} = v_{\mathrm{i}}$.

For the present experiment the minimum relative energy that can be reached with a misalignment by $\theta = 7$ mrad is 0.19 eV. Lower energies would not be accessible at that angle. With all the previous experience, the energy range where a recombination rate enhancement can be expected is well below this limit, so that no deviation from the normal RR theory would be visible in that case. Since the experiment does show a rate enhancement at energies below 0.02 eV the misalignment angle must be much smaller than 7 mrad. The example demonstrates, however, that even a misalignment of 0.7 mrad would make it difficult to see the enhancement. In general, slight deviations in the alignment of the electron and ion beams will reduce the observed enhancement factor.

Another effect occurs when there is a finite angle between the electron and the ion beam. Since the electron beam is parallel with the magnetic guiding field, the ions are subject to a $\vec{v}_{\mathrm{i}} \times \vec{B}$ motional electric field. In the present experiment the magnetic field was 0.76 T. An angle $\theta = 0.7$ mrad would result in a motional electric field of more than 190 V/cm seen by the ions. Such a field would lead to field mixing of ℓ-states and would also strip off very high Rydberg states. This may be important for the rate enhancement effect studied in this paper since there is experimental [19] and theoretical [20] evidence for the importance of very slightly bound electrons in the enhanced-recombination phenomenon. When these electrons are immediately

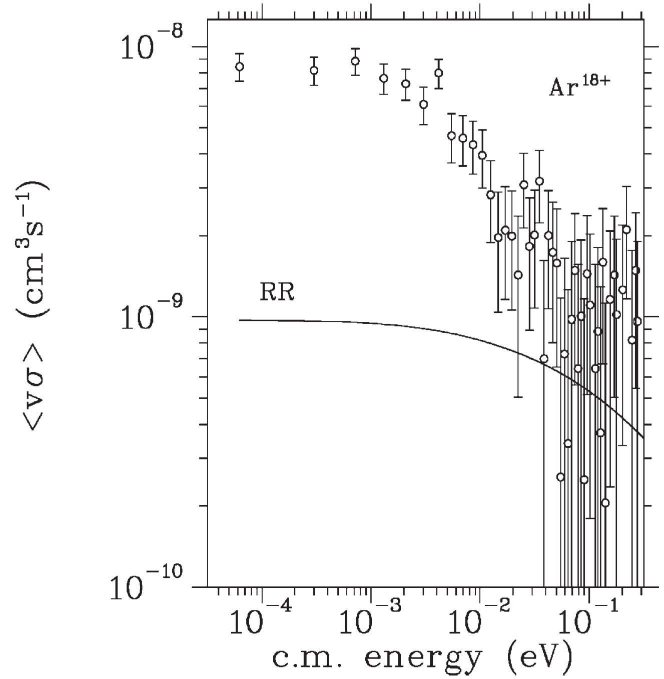

Fig. 2. Measured absolute recombination rates of electrons with 7.1 MeV/u Ar^{18+} ions, vs. the relative energy in the electron–ion center-of-mass frame. The solid line represents theoretical calculations for radiative recombination (RR) using electron beam temperatures which correspond to $kT_{\parallel} = 0.002$ eV and $kT_{\perp} = 0.2$ eV.

stripped off after they are attached to the ion, there may not be sufficient time for the development of an enhanced recombination into more tightly bound states. Thus, it appears to be easy to miss the observation of an extra peak in the recombination rate at $E_{\rm rel} = 0$ just because of a slight misalignment of the electron and ion beams.

While the measurements with Ar^{16+} yielded a maximum rate which is slightly lower than that observed previously with Ar^{15+}, the measurement with Ar^{18+} resulted in a recombination rate maximum that exceeds the expectation by nearly a factor 10. The experimental result obtained for Ar^{18+} in the low energy range from about 10^{-4} eV up to ≈ 0.1 eV is displayed in fig. 2. The data were obtained by the procedure described in detail in ref. [10] which is appropriate to yield absolute data. In the present set of experiments, however, technical reasons made it especially difficult to control the overlap of the electron and ion beams. This does not influence the relative uncertainties between individual data points but if the ion beam is not fully immersed in the electron beam (which is 3 mm in diameter) the inferred rates would be too low. In the case of the Ar^{18+} data the rates displayed in the figure were determined assuming complete beam overlap, i.e. a lower limit for the rates is presented here. But without any correction factors the rates measured at energies between 0.5 eV and 2.5 eV are in satisfactory agreement with RR theory providing confidence in the validity of the assumption about beam overlap. (Due to space limitations these data are not displayed here.) Nevertheless, at the present stage of the data analysis we make a conservative estimate of the total uncertainty of 50% for the data set displayed in fig. 2.

The solid curve in fig. 2 is the theoretical calculation, based on the Bethe–Salpeter formula with appropriate corrections. The electron beam temperatures can only be inferred from other experiments where e.g. DR resonances are analyzed. With completely stripped parent ions DR is not possible. Therefore, and in order to obtain an upper limit for the RR rate, the lowest temperatures ($kT_{\|} = 0.002$ eV, $kT_{\perp} = 0.2$ eV) seen so far at the electron target facility have been used to calculate the RR rate.

The measured maximum rate always has to be taken as a lower limit for the real recombination rate at $E_{\rm rel} = 0$, since – besides the beam overlap question mentioned above – the experiment may always miss this energy due to a slight misalignment. This is true for every merged beams experiment. Even so, the present measurement with Ar^{18+} ions yields the highest recombination rate enhancement factor ever observed with a completely stripped species. At the high-charge-state end of a sequence of detailed merged beams experiments with C^{6+} (enhancement factor ≈ 1.5) [12], N^{7+} (enhancement factor ≈ 2) [21], Ne^{10+} (enhancement factor ≈ 3 to 4) [11], and Si^{14+} (enhancement factor ≈ 2) [21], the present measurement for Ar^{18+} drops out of the systematic trend. The excessively large enhancement (which – following the above arguments – is a lower limit) may be due to the very high electron density in this very experiment in comparison with all the other measurements with fully stripped ions which were carried out at storage ring coolers with electron densities roughly a factor of 200 lower. A detailed understanding of this behaviour is not yet available.

3. Summary and outlook

Radiative recombination of multiply charged ions is enhanced over the expectations of generally accepted theoretical calculations. Deviations for completely stripped ions by up to a factor of 10 are found in merged-beams experiments at relative energies $E_{\rm rel} \ll kT_{\perp}$ between the electrons and the ions, where k is Boltzmann's constant and $T_{\perp}$ the transverse temperature in the electron beam. This is documented by the present experiment with completely stripped argon ions. By a slight misalignment between the two beams the enhancement effects can easily be missed. With this in mind, the measured maximum recombination rates always constitute lower limits of the real rates at $E_{\rm rel} = 0$ (provided the experiment was carried out correctly and all parameters apart from the interaction angle of the two beams are well controlled). Small but non-zero residual angles in some of the experiments carried out so far may be the explanation for not seeing the enhancement effect which is well established by now through many experiments at different facilities. That experimental uncertainty makes the interpretation of a truly puzzling result even more difficult. More experiments will be necessary to gain a satisfactory understanding of the recombination rate enhancement effects at very low energies. Systematic measurements along isonuclear sequences of ions in different charge states and along isoelectronic sequences will hopefully shed light onto a new unexpected phenomenon which has a significant influence on electron cooling of ions in storage rings.

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