Radiative double electron capture (RDEC) in ion-atom collisions

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Abstract. Radiative double electron capture (RDEC) observed in collisions of bare ions with atoms is a charge exchange process, during which two target electrons are captured into a bound state of the projectile and a single photon is emitted. This process could be related to the time inverse of double photoionization. For the past twenty years it has been studied, both experimentally and theoretically. However, significant discrepancies between theoretical predictions of the RDEC cross section and experimental results were noted. Here, an overview of the investigation of the RDEC process is given and various theoretical predictions are compared with experimental results.

1. Introduction

Electron capture processes are one of the cleanest tools for investigation of the structure and properties of atoms. Performed usually with bare or few electron ions, these experiments provide results not disturbed by the influence of the electrons not participating in the observed process, as is the case for photoionization experiments. Moreover, multi-electron capture experiments may provide insight into electron-electron interactions in the presence of the electromagnetic field generated by the nucleus motion.

The basic electron capture process is a single radiative electron capture (REC) during which transfer of a target electron to a bound state of the projectile is accompanied by emission of a single photon. In the nonrelativistic case, the energy of the emitted photon in the projectile frame of reference is given by:

\[ E_{REC} = \frac{m_e}{m_p} E + E_{B}^{proj} - E_{B}^{T} \pm \vec{p}\vec{v}, \tag{1} \]

where \( m_e \) and \( m_p \) are electron and proton rest masses, \( E \) is the projectile energy per nucleon, \( E_{B}^{proj} \) and \( E_{B}^{T} \) are the electron binding energies in the projectile and target, respectively, and \( \vec{p}\vec{v} \) is the projection of the electron momentum \( \vec{p} \) onto the projectile velocity \( \vec{v} \). The distribution of the momentum of the target electrons (Compton profile) defines the width of the REC peak.

The simplest atomic process that involves correlation between the captured electrons is radiative double electron capture (RDEC) during which two target electrons are captured into a bound state of the projectile and the energy difference between their final and initial states is
Figure 1. Radiative electron capture (REC) and radiative double electron capture (RDEC) processes. For symbol explanation see text.

emitted as a single photon. Energy of the RDEC photon is equal to about twice the energy of the REC photon and can be expressed as:

$$E_{REC} = \frac{m_e}{m_p} E + E_{B(1)}^{\text{proj}} + E_{B(2)}^{\text{proj}} - E_{B(1)}^T - E_{B(2)}^T \pm 2\bar{p}v,$$

(2)

where indices (1) and (2) denote the captured electrons. Both REC and RDEC processes are schematically shown in Fig. 1.

The RDEC process can be viewed as a part of time inversed double photoionization and as such is of fundamental interest. In particular, it may be a tool for probing of electron-electron correlations. Experimental approaches towards observation of RDEC were assisted by theoretical calculations [1–8] based on diverse approximations. However, significant discrepancies between calculated values of the RDEC cross sections were still noticed which complicated selection of favorable experimental conditions.

2. First experiments dedicated to RDEC

The first experiment dedicated to RDEC was performed with 11.4 MeV/u Ar$^{18+}$ ions impinging upon a carbon foil [9]. A typical spectrum obtained during that experiment is presented in Fig. 2. As shown in this figure, no significant line structure related to the RDEC process was observed. However, the number of counts collected in the expected RDEC energy window provided an upper limit for the RDEC cross section of about 5.2 mb.

This experiment was followed by a first theoretical treatment [2, 3], where nonrelativistic calculations of the RDEC process adapted to the kinematics and the energy range of the Ar$^{18+}$ + C experiment were presented. The calculations gave, for this particular collision system (Ar$^{18+}$ + C at 11.4 MeV/u), an RDEC cross section value close to the experimental upper limit. However, later on, another theoretical approach [7] showed that for various approximations, a broad range of calculated cross section values (ranging through four orders of magnitude) can be obtained. Recently, a cross section of 3.2 mb [4] was calculated for this particular system.

Moreover, calculations presented in [2, 3] predicted a strong enhancement of RDEC during...
Figure 2. Example x-ray spectrum associated with double electron capture obtained during the Ar$^{18+} + C$ experiment from [9].

Figure 3. X-ray spectrum registered during the U$^{92+} + Ar$ experiment in coincidence with double electron capture [10]. The Gaussian line in the RDEC energy range shows the peak that would have been observed if the theoretical predictions [2, 3] were correct.

relativistic heavy ion-atom collisions. This particular prediction was tested during the second experiment dedicated to RDEC. Here, bare uranium ions at an energy of 297 MeV/u collided with an Ar target at the ESR storage ring of the GSI facility [10]. This experiment showed that for the collision system under consideration the RDEC cross section is at least three orders of magnitude smaller than the theoretical expectations [2, 3]. Fig. 3 shows a spectrum obtained during the experiment. Again, no significant line structure which could be assigned to the RDEC process was observed. The Gaussian line shown within the RDEC region of the spectrum in Fig. 3 represents the shape of the RDEC line which should be observed, if the theoretical calculations [2, 3] were reliable. This experiment also provided only an upper limit for the RDEC cross section value of about 10 mb.
3. Nonrelativistic calculations of the RDEC cross section

In order to explain the uranium experiment [10] a new theoretical approach for the correlated double electron capture into the K-shell of bare ions was proposed [4–6]. These calculations are in contradiction to the previous relativistic approach [3], which was not able to explain the existing experimental data [10]. As shown in [4], the enhancement of the wave function for the relativistic systems was calculated incorrectly in [2,3] and even the corrected value, which is 3 orders of magnitude smaller [4], was not confirmed by the experiment [10]. Therefore, the proposed enhancement of the RDEC cross section for relativistic systems [3] seems to be absent. This is similar to the behavior of the cross section for single REC events, where the cross section decreases with increasing projectile energy.

However, it has to be emphasized that the current estimate [4] of the RDEC cross section ($\sigma_{RDEC}$) gives values closer to the experimentally obtained upper limits for both the nonrelativistic case (Ar$^{18+} + C$, [9]) and the relativistic one (U$^{92+} + Ar$, [10]) (see Table 1), which suggests that this is so far the most reliable theoretical description of RDEC.

In addition, this theoretical approach anticipated the cross section for the correlated double electron capture into the excited 1$s^2$ projectile state to increase the chance for observation of the RDEC process [6]. It was shown that the ratio of $\sigma_{RDEC}$ to the 1$s^2$ state to $\sigma_{RDEC}$ to the 1$s^2$ ground state is strongly dependent on the Sommerfeld parameter. For example, in case of slow collisions the cross section for electron capture to the excited 1$s^2$s state can be significantly higher than that for capture to the 1$s^2$ ground state. Thus, slow low-Z ions collided with light targets were suggested as the best systems for observation of the RDEC process.

Based on these predictions two experiments with low-Z low energy beams were performed at Western Michigan University using the 6 MV tandem Van de Graaff accelerator. The two systems chosen for the experiments were O$^{8+} + C$ at 38 MeV and F$^{9+} + C$ at 42 MeV. The next sections describe in detail the setup and results of these experiments.

4. Experimental setup

A schematic of the experimental setup is shown in Fig. 4. For these experiments a target chamber for solid target foils was built, which not only allowed for mounting up to four films but also provided a simple mechanism for target rotation. This was necessary for optimization of the target position during the experiment. During data acquisition the target foil was positioned at 45° to the beam direction, facing the x-ray detector as shown in Fig. 4. The thicknesses of the target foils used during the experiments were of the order of 10 $\mu g/cm^2$ which corresponds
to an areal density on the order of $10^{17}$ particles/cm$^2$.

The target chamber was designed in a way that minimizes the distance between detector window and target center. The total crystal-target distance achieved was about 25 mm, which gives a detection solid angle of $\Delta \Omega = 0.044(1)$ sr.

Emitted x-rays were registered by an ORTEC single crystal Si(Li) detector placed perpendicular to the beam direction. The crystal of 6 mm diameter and 3 mm thickness, together with a 7.5 $\mu$m Be-window, gave a detection efficiency in the energy range 2-4 keV better than 90%. The detector was energy calibrated with a standard $^{55}$Fe calibration source. For the fluorine experiment an additional x-ray Si(Li) detector was mounted on the opposite side of the target chamber, perpendicular to the beam direction. This detector had a beryllium window six times as thick as the one in the first detector and thus was less sensitive to the low energy x-rays that were strongly attenuated when passing through the Be-window. This allowed for additional verification of the insignificance of the pile up effect that comes from events in the RDEC energy range of the spectrum resulting from simultaneous detection of two REC photons. It was confirmed that, as it was shown for the oxygen data [11], the pile up effect for this setup is negligible.

Along the beam direction, a set of two collimators was placed in front of the target chamber. The distance between collimators was about 2 m. Collimator apertures of 2 and 3 mm were set to ensure good beam collimation. An additional collimator between the target and analyzing magnet prevented scattered ions from entering the spectrometer and generating false coincidences (see Fig. 4).

The target chamber was followed by a magnetic spectrometer. The magnetic field of the dipole magnet separated the final charge states of the ions and directed them towards surface barrier detectors. Surface barrier detectors counted ions with charge states equal to $q - 1$ and $q - 2$, where $q$ is the charge state of the primary beam. Both the magnetic field of the spectrometer and the position of the surface barrier detectors were adjustable and created a versatile system, which could be used for various beam charge states and energies. The primary beam was registered by a Faraday cup during the oxygen experiment and with a surface barrier detector during fluorine runs. The data acquisition system registered coincidences between x-rays and each of the particles detectors, which allowed for separation of various electron capture processes.

Figure 5. Example of the x-ray spectra obtained in coincidence with single ($q - 1$) and double ($q - 2$) charge change for the oxygen experiment. Insets in each of the panels show the expanded RDEC range of the x-ray spectra.
5. Results
The first experiment was performed with a bare oxygen ion beam at a total energy of 38 MeV collided with a 20 μg/cm² carbon foil. The coincidence spectra obtained for this experiment are presented in Fig. 5. A structure that could be associated with RDEC to various final electrons states, as discussed in [11], can be seen in 3.3-4.2 keV range of the spectra. Due to multiple collisions occurring in the target, the projectile was likely to lose one of the previously captured electrons. Thus, a significant number of RDEC photons was observed in the q – 1 coincidence channel and not as expected in the q – 2 channel. Detailed analysis of the experiment is given in [11, 12]. The calculations of the experimental RDEC cross section were based on the ratio of RDEC to REC events, as the REC cross section can be easily obtained from the well known Stobbe formula [13]. The total number of counts observed in the RDEC range of the x-ray spectra was 357, which corresponds to a cross section value of 3.2(1.9) b.

The second experiment incorporated a bare fluorine beam at 42 MeV and a carbon target with areal density of about 11 μg/cm². For better x-ray detection efficiency another Si(Li) detector was mounted at 90° to the beam direction opposite to the previously used one. For this experiment, the Faraday cup for the beam current measurement was replaced by an additional surface barrier detector. This allowed for a more accurate determination of the beam intensity so that the RDEC cross section could be calculated from the experimental geometry. Sample spectra obtained in coincidence with single and double charge change during the fluorine experiment are presented Fig. 6. The RDEC range of the x-ray spectra of 3.9-4.7 keV is shown in the insets. Despite the low statistics a structure in the RDEC range of the spectra seems to resemble the one obtained for oxygen ions. During this experiment a total of about 60 events that are likely to be RDEC photons was observed, which gives the total RDEC cross section of about 0.5(0.3) b.

It can be noticed that despite the fact that the collision systems during both experiments were very similar, the cross sections differ by a factor of six. However, the fluorine experiment was performed with higher background and in addition lower statistics than for the experiment with oxygen ions. The background could be significantly reduced by application of the coincidence condition, but the statistical uncertainty of the obtained results is high and the results should be considered as less reliable. A detailed analysis of the F + C data is still in progress.
Table 1. Summary of the experimentally verified RDEC cross sections for various systems compared with theoretical predictions.

| Z  | E [MeV/u] | Zt | \( \sigma_{RDEC} \) [mb] |
|----|-----------|----|-------------------------|
|    |           |    | Ref. [3] | Ref. [4] | experiment |
| 18 | 11.4      | 6  | 1.85       | 3.2      | \( \leq 5.2 \) [9] |
| 92 | 297       | 18 | 5.0\times10^3 | 2.5\times10^{-2} | \( \leq 10 \) [10] |
| 8  | 2.37      | 6  | 1.4\times10^2 (a) | 1.5\times10^2 (b) | 3.2(1.9)\times10^3 [14] |
| 9  | 2.21      | 6  | 1.0\times10^2 (a) | 1.1\times10^2 (c) | 0.5(0.3)\times10^3 |

(a) estimated from the \( \sigma_{RDEC}/\sigma_{REC} \) ratio given in [3] as shown in [11]  
(b) provided by Nefiodov [15]  
(c) calculated based on formulae given in [4, 5]

6. Summary
An overview of the experimental and theoretical approach towards RDEC was presented. All cross section data, both measured and calculated, obtained up to now are summarized in Table 1. It can be noticed that significant discrepancies between experiment and theory exist. It should be pointed out that this is consistent with [7], where the authors show that the theoretical predictions of the RDEC cross section may differ by four orders of magnitude, depending on the considered approximation of the two electron wave function. However, the latest theoretical treatment of RDEC [4–6] seems to be the most reliable and the closest to all the experimental values. But it is still necessary to experimentally verify the RDEC cross section for various collision systems, in order to narrow down the range of theoretically predicted values.

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