Radiative Double Electron Capture in Collisions of O⁸⁺ Ions with Carbon

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Multielectron capture processes observed in low energy collisions of bare ions with target atoms open insight into electron correlations in electromagnetic fields. Radiative double electron capture (RDEC) provides the simplest tool for investigation of such processes. Here, the experimental observation of the RDEC process in collisions of O^{8+} ions with thin carbon films is presented and the RDEC cross section value obtained is compared with recent theoretical calculations.

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One of the best known atomic processes observed during ion-atom collisions is the radiative electron capture (REC) [1,2]. If the captured electron can be treated as quasifree [i.e., in systems where $Z_t \ll Z$, Z_t and Z being target and projectile atomic numbers, respectively], REC can be considered as a time reversed photoionization, the most important interaction process between radiation and matter [3]. Unlike single photoionization of multielectron systems, REC has been investigated in bare ion-atom interactions [4,5] and offers clean conditions for exploration of photoionization with only one electron, which allows for observation of pure photon-electron interaction.

During the last 30 years double photoionization has been of considerable interest ([6] and references therein). As a photon typically interacts only with one electron, this phenomenon is caused by the electron-electron interaction [7]. However, in high Z systems the subtle electron correlation effects are difficult to observe due to background contributions from the electrons not involved directly in the process. The time reversed process, radiative double electron capture (RDEC), which involves transfer of two target electrons into a bound state of the projectile with simultaneous emission of a single photon [8,9], does not have this background. Since bare ions are used in the experiment, RDEC can be considered as the simplest, clean tool for investigation of electron correlations [8]. Thus, investigation of the RDEC process can provide crucial information necessary for a proper description of electron correlations in atomic systems and, in addition, data required to define the wave function of two electrons in the projectile continuum. Both the REC and RDEC processes are shown in Fig. 1.

During the last 20 years the RDEC process was addressed not only experimentally [8,9], but also theoretically [10–12]. The calculations were found to be in disagreement with experimental data [9] and verification of the RDEC process was not possible. More recent calculations not only explain previous experimental results, but also suggest the choice of low energy mid-Z ($Z \le 35$) collision systems for observation of RDEC [13,14]. It is noted that for bare ions capture to an excited $1s^12s^1$ state might significantly contribute to the observed x-ray spectra [15].

In this Letter we present experimental evidence of radiative double electron capture. The RDEC cross section obtained for O^{8+} + C collisions at 38 MeV is compared to recent theoretical calculations [13–15].

The experiment was conducted at Western Michigan University using the tandem Van de Graaff accelerator. A schematic of the experimental setup is presented in Fig. 2. Incoming ions were collided with carbon foils of thickness of a few μ g/cm² mounted on an aluminum holder tilted at 45° to the beam direction. 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 μ m Be-window,



FIG. 1. Schematic diagram of radiative electron capture (REC) and radiative double electron capture (RDEC) processes. Capture of one (REC) or two (RDEC) electrons is accompanied by the simultaneous emission of a photon. T_K is the kinetic energy of the quasifree target electron, moving with the projectile velocity, in the projectile frame.



FIG. 2. Experimental setup.

provided a detection efficiency in the 2–4 keV energy range of more than 90%. The target chamber was designed in a way that minimizes the distance between detector window and target center in order to maximize the detection solid angle. The target-crystal distance was about 25 mm.

The magnetic spectrometer downstream from the target separated the final charge states of the ions and directed them towards surface barrier detectors corresponding to q - 2 and q - 1 charge states, where q is the charge state of the primary beam. The primary beam was registered by a Faraday cup.

The main goal of the experiment was observation of x rays generated by bare oxygen ions impinging on carbon at an energy of 38 MeV. A short run without the target foil was performed in order to establish the background due to scattered ions. This was necessary to eliminate the Al $K\alpha$ line from the x-ray spectra, which results from beam interactions with the aluminum target holder. An example of the obtained spectra for O⁸⁺ is presented in Fig. 3(a).

In order to eliminate the Al $K\alpha$ line from the $O^{8+} + C$ data, a spectrum obtained for bare ions without the carbon target was normalized and subtracted from the data taken with the foil. This procedure resolved the REC structure in the single spectrum, as shown in Fig. 3(b). In case of the REC process the captured electron could come either from the target *K*-shell or the valence band. Thus, a double peak structure in the REC region was expected with the peak position difference in the spectra equal to the differences in the binding energy of the *K*-shell and valence electrons in the carbon foil. Expected positions of the corresponding REC peaks are given in Table I.

An additional run with 38 MeV O^{7+} ions was also performed. This was to check if the structure of the x-ray spectra in the RDEC region changes when one electron in the *K* shell is present, which means that RDEC to the $1s^2$ state is blocked. As shown in Fig. 3(c), no significant structure in the RDEC region was observed.

As an additional test of experimental conditions, PIXE analysis of the target foil was performed with 2.375 MeV protons, i.e., at the same collision velocity as for O^{8+} ions. In this process the impinging proton beam excites atomic states of the atoms within the material, which then deexcite emitting characteristic x rays. Intensities of the lines allow for estimation of the percentage of the impurities in the material. Here, PIXE analysis was performed to check if



FIG. 3. Single x-ray spectra obtained during the experiment. In all spectra: solid line—38 MeV O^{8+} . (a) dotted line— O^{8+} data taken without the carbon foil, (b) O^{8+} data after subtraction of normalized O^{8+} spectrum obtained without target foil, (c) dashed line—38 MeV O^{7+} , (d) dot-dashed line—2.375 MeV protons. See text for detailed explanation of REC and RDEC ranges.

TABLE I. Calculated positions of the RDEC and REC peaks in the x-ray spectrum corresponding to different combinations of the initial and final states of the captured electrons. All values are given in keV.

	Captured target electrons One K shell		
	Valence	and one valence	K shell
REC	2.16	-	1.88
RDEC $1s^2$	4.18	3.91	3.64
RDEC $1s^12s^1$	3.58	3.31	3.04

any impurities, that might produce x rays in the RDEC range, are present in the foil. As can be seen in Fig. 3(d), no structure in the RDEC region was observed, nor was there evidence for REC.

In the investigated $O^{8+} + C$ collision system the energy of an REC photon is around 1.9 keV, thus the RDEC line in the x-ray spectrum is expected to appear at about 3.8 keV. As the energy of the RDEC photon is almost twice as large as the energy of the REC photon, it might be impossible to distinguish a real RDEC event from a situation when two REC photons are simultaneously detected by the x-ray detector. It can be shown that the probability of double collisions in the target resulting in emission of two REC photons in this collision system is negligible. If the cross section for RDEC is of the order of 0.1 b [16] and the given geometry of the experiment is taken into account, one obtains the probability of observing an RDEC photon during one ion-atom collision equal to 1.3×10^{-8} . The REC cross section of 512 b can be obtained from the wellknown Stobbe formula [17]. The probability that one ion interacts on its path with two atoms and emits two single REC photons, which are then registered as one photon with doubled energy, is about 5×10^{-14} . This value is negligible if compared to the probability of observation of an RDEC event.

This estimation was supported by calculations of the $N_{\rm RDEC}/N_{\rm REC}$ ratio, where $N_{\rm RDEC}$ and $N_{\rm REC}$ are the numbers of counts in RDEC and REC region, respectively. For true events the numbers of counts in both the REC and RDEC regions are proportional to the beam intensity. Thus, the ratio $N_{\rm RDEC}/N_{\rm REC}$ does not change with the beam intensity. If double REC counts are registered in the RDEC energy range, $N_{\rm RDEC}$ is proportional to the square of beam intensity and the $N_{\rm RDEC}$ to $N_{\rm REC}$ ratio should increase, when the beam intensity increases. Experimental values of the $N_{\rm RDEC}/N_{\rm REC}$ ratio for low, medium, and high beam intensities do not depend on the beam intensity within the limits of uncertainties.

Coincidence spectra obtained for bare O^{8+} ions associated finally with double or single charge exchange are presented in Fig. 4. In both spectra, a peak structure in the RDEC region is evident. The REC counts visible in coincidence with ions which captured two electrons come

week ending 26 MARCH 2010



FIG. 4. X rays registered for $O^{8+} + C$ in coincidence with ions which captured (a) two electrons and (b) one electron. Solid line—the sum of REC Compton profile (REC) and Gaussian distribution (O $K\alpha$) fitted to the spectrum.

from noncorrelated capture events in the target with at least one radiative capture. This could not be avoided due to the very high cross section for nonradiative electron capture which is of the order of 0.5 Mb, as estimated according to the commonly used scaling formula [18].

It can be seen in Fig. 4 that application of the coincidence conditions eliminated almost all the background counts in the region of interest. The only remaining background structure is the REC line. REC results in a very broad structure with a long tail extending towards high photon energies [19], which is clearly visible in Fig. 4(b). In order to eliminate REC counts from the RDEC region, a Compton profile based on Ref. [20] was fitted to the coincidence spectrum shown in Fig. 4(b). The remaining structure consisted of photons which could be assigned to the RDEC process. The inset in Fig. 4(b) shows clearly the existence of a peak structure above the Compton profile fit. Furthermore, a structure of at least two peaks can be noticed in the RDEC energy range. This structure is not only a result of capture to both ground and excited states of the projectile, but can also be associated with capture of either K-shell or valence target electrons. Possible combinations of the initial and final electrons states and the resulting RDEC peak positions are given in Table I.

Here, a question about the presence of the RDEC counts in the q - 1 coincidence spectrum arises. One has to keep

TABLE II. Ionization cross sections for oxygen at 38 MeV estimated from [21,22].

	Process	Cross section [Mb]
<i>L</i> -shell ionization	$\begin{array}{c} O^{5+} \rightarrow O^{6+} \\ O^{6+} \rightarrow O^{7+} \end{array}$	19.0 3.6
K Shell JohnZution	$0^{7+} \rightarrow 0^{8+}$	0.4
	$O_{0^+} \rightarrow O_{0^+}$	0.1

in mind that the ionization probability can be high for the weakly bound ionic systems that are formed. Ionization cross sections obtained by interpolation of relevant data [21,22] are given in Table II. These cross sections are in fair agreement with the data of Shima et al. [23], where the bare and H-like ions are indicated as the most populated charge states observed when the 38 MeV oxygen beam traverses through a carbon foil. Together, they account for more than 80% of the final charge states, with 50% in the H-like (7+) state. Moreover, the ionization probability strongly varies with the final states of the two captured electrons. In the system investigated, the cross section for L-shell ionization is about 1 order of magnitude greater than that for the K shell (see Table II). Thus, in case of double capture to the $1s^{1}2s^{1}$ state, one of the electrons (in the 2s state) is promptly removed, while in case of capture to $1s^2$ the final charge state of the ion is more likely to remain unchanged. Therefore, one would expect most of the photons originating from RDEC to the projectile excited state to show up in the single charge exchange channel, while the capture to the ground state will be less affected by ionization of its K-shell electrons. This can be observed in Fig. 4 where the $1s^{1}2s^{1}$ peak is clearly visible in the q-1 coincidence spectrum, while it is almost absent in q - 2, compared to $1s^2$ which is still visible in the q-2 channel.

The total ratio of the numbers of RDEC to REC counts obtained in the experiment $N_{\text{RDEC}}/N_{\text{REC}} = 0.0092(6)$ allows for estimation of the differential RDEC cross section of 0.71(5) b/sr at 90° observation angle. This value gives the total RDEC cross section of about a factor of 5 greater than the total cross section obtained from the theory $(\sigma_{\text{RDEC}} = 0.15 \text{ b} [16])$. However, the theoretical value accounts only for the capture of target K-shell electrons to the projectile ground state. The obtained experimental data did not allow for separation of the different capture channels. Thus, the experimental value of the cross section is the sum of the cross sections both for capture to the ground and to the excited states of the projectile. Furthermore, according to Nefiodov [16], the system chosen for the experiment does not fully meet the theoretical conditions, since for $Z \sim Z_T$ the captured electrons cannot be treated as quasifree and theoretical calculations might be underestimated.

In summary, an experimental observation of the RDEC process is reported. The obtained cross section value for $O^{8+} + C$ system at a collision energy of 38 MeV is compared to recent theoretical calculations. The experimental value of the total cross section is about 5 times greater than the theoretical prediction for this system. This discrepancy might be due to the fact that the theory did not include capture to the excited states and that the captured electrons cannot be treated as quasifree in the experimental system under consideration.

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