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Preliminary Results for Observation of Radiative Double-Electron Capture by $F^{9+,8+}$ on Graphene

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Abstract: Radiative double-electron capture, which can be considered the inverse of double photoionization, has been investigated for 2.11 MeV/u F^{9+} and F^{8+} projectiles colliding with the two-dimensional target single-layer graphene. Preliminary results for the cross sections are obtained and presented and compared with our previous measurements for the one-dimensional gas targets N_2 and Ne, with the three-dimensional target thin-foil C, and with the most accurate theoretical results that currently exist. The graphene results reported here are reasonable when compared with the F^{9+}+N_2, Ne results given the thicknesses of the respective targets, being larger by about a factor of four.

Keywords: ion–atom collisions; electron capture; graphene

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1. Introduction

Electron capture to atoms and molecules is a fundamental process in the fields of physics and chemistry and has been studied for well over a century. Ions in particular, with one or more electron vacancies in the parent atom/molecule, are especially amenable to this process. Given an ionic projectile, one can broadly separate the classes of targets into plasmas, gases, and solids. The former two can be viewed as one-dimensional targets, while the latter is generally visualized as a three-dimensional target. An interesting extension of this concept is the two-dimensional target, consisting of a single-layer solid target. Graphene, an allotrope of carbon with a hexagonal structure, is such a material.

Upon capturing an electron, several processes may take place to satisfy conservation of energy. Of particular interest here is the capture of an electron simultaneous with the emission of a single photon. For single capture from a bound state in the target, this process is termed radiative electron capture (REC) [1–3]. This process is well-confirmed and has been studied both theoretically and experimentally for over half a century. Far less often studied is radiative double-electron capture (RDEC) [4] where two electrons are captured from bound states in the target (projectile continuum states) to bound states in the projectile with the emission of a single photon. Both REC and RDEC can be considered inverse processes of photoionization, with REC representing the inverse of single ionization and RDEC double ionization. Due to the energies involved and the difficulty of obtaining sufficiently bright ion and photon beams, double photoionization due to a single photon has not been studied experimentally in a system other than helium. Therefore, the study of its ion-atom analog gives insight into the process.

As mentioned, RDEC has not been extensively studied due to the difficulty in its measurement. Nevertheless, several intrepid groups have attempted to observe the process. The first such trial was reported from GSI Darmstadt for 11.4 MeV/u Ar^{18+} on C [5], with inconclusive results. Natural intuition then led to a far heavier, swifter projectile, when an investigation for 297 MeV/u U^{92+} on Ar [6] followed, again without definitive results.

It was shortly after this that Nefiodov et al. [7] theorized that mid-Z, low-energy projectiles should yield larger RDEC cross sections. Following this insight, experimental searches were initiated at Western Michigan University (WMU), yielding the first successful observation of RDEC for 2.38 MeV O^{8+} on C [8]. The GSI group in Darmstadt conducted one final investigation for 30 MeV/u Cr^{24+} on He and N_2, with no evidence of RDEC reported [9]. Finally, from WMU the most definitive observation of RDEC was reported for 2.11 MeV/u $F^{9+,8+}$ projectiles incident on N_2 and Ne gas targets [10], followed by 2.11 MeV/u $F^{9+,8+}$ and 2.19 MeV/u $O^{8+,7+}$ on thin-foil C [11]. The latter works are summarized and further explored in Tanis et al. [12]. The work presented here is an attempt to bridge the gap between one-dimensional targets (gaseous N_2 and Ne) and three-dimensional targets (thin-foil C) with a two-dimensional target (graphene). The first similar experiment was done at much lower projectile energies (60 and 135 keV Xe^{30+}) in which was found that the graphene target retained its integrity, to some surprise, as the slow ion passed through during the collisions [13].

2. Experimental Setup and Methods

It is appropriate to first consider the energy of the process one wishes to observe. The energy schematic for RDEC can be seen in Figure 1. Shown there is double capture from the projectile continuum states to bound states in the projectile (KK capture shown as an example) with the simultaneous emission of a single photon. The projectile may of course capture the target electrons to the same or different bound states while emitting a photon with a distinct energy. The energy for this process is given as:

$$E_{RDEC} = 2K_t + B_p^1 + B_p^2 - B_t^1 - B_t^2 + \tilde{v_p} \cdot \tilde{p_{it}}^1 + \tilde{v_p} \cdot \tilde{p_{it}}^2$$

where the superscripts 1 and 2 denote each captured electron, K_t is the kinetic energy of the captured electron in the rest frame of the projectile, B_p is the positive binding energy in the projectile, B_t is the positive binding energy in the target, v_p is the velocity of the projectile ion in the lab frame, and p_{it} is the intrinsic momentum of the captured target electron due to its orbital motion. This velocity and momentum vector overlap is termed the Compton profile, and is generally quite large (∼100 eV per electron captured) in this instance.

For the REC process (not shown), only a single electron is captured. With the appropriate choice of target and projectile, the energy of an RDEC photon is approximately double that of an REC photon, giving a way to distinguish the two processes in the laboratory. The differential cross section for REC has been shown to have a $\sin^2\theta$ dependence [14,15], and that relationship is assumed for RDEC here as well.

When attempting to observe the RDEC process, one must consider the possibility of observing two REC photons within the time bandwidth and the same emittance angle of the detector. Should this happen, the two REC photon energies will be additive within the physical X-ray detector and will falsely appear as a single photon with an apparent RDEC energy. However, double REC scales as $(\sigma_{REC}/a_0)^2$, where σ_{REC} is the REC cross section and a_0 is the Bohr radius, with $\sigma_{REC} \ll a_0$ [16]. It follows that the probability of observing double REC is about two orders of magnitude smaller than for RDEC and is, therefore, disregarded in this case.

The experiment was conducted using the 6-MV tandem Van de Graaff accelerator facility at WMU. Following acceleration, appropriate magnets and a thin C stripper foil selected the desired energy and charge state for the fluorine projectiles (40 MeV F^{9+} and F^{8+}). The ion beam was then directed towards the experimental chamber, which is depicted in Figure 2. The graphene was placed in a target ladder situated at 45° to the beamline. Additionally, the ladder contained a thin-foil carbon target as well as no target for calibration and background runs, respectively. X-rays characteristic of the target holder are outside of the region of interest. The cleanliness of the graphene target was ensured via proton-induced X-ray emission runs separate from the RDEC investigation. The commercially sourced single-layer graphene target [17] is approximately ∼0.35 nm thick and is mounted on a 200 nm thick silicon nitride transmission electron microscopy grid. The grid has

approximately 6400 holes of 2 μm diameter on a 3.0 mm hexagonal, 200 μm thick silicon nitrate substrate with a 0.5 × 0.5 mm aperture. A similar target was used by the Vienna group [13] in their work with low-energy projectiles. A Si(Li) X-ray detector with an effective area of ∼60 mm^2 was positioned 2.8 ± 0.1 cm away from the target at 90° to the beamline, yielding a solid detection angle of 0.0765 steradians. The detector has a 0.4 μm polymer window whose transmission efficiency near the F K X-ray energy (∼1 keV) is nearly 85%. The detection efficiency of the X-ray detector in the calculated RDEC energy range is greater than 98%. Following the interaction region, the ion beam was charge-state selected using a dipole magnet, with the doubly- (q-2), singly- (q-1), and no-charge (q) changed components directed onto separate silicon surface-barrier detectors. Data acquisition was performed using event-mode collection with the coincidences between X-rays and charge-changed particles being recorded. This allows the X-rays in the RDEC energy range (see Table 1) to be assigned to their respective charge-changed particles, or vice versa. Additional calibration was provided by a ^{55}Fe source to more accurately determine the RDEC energies.

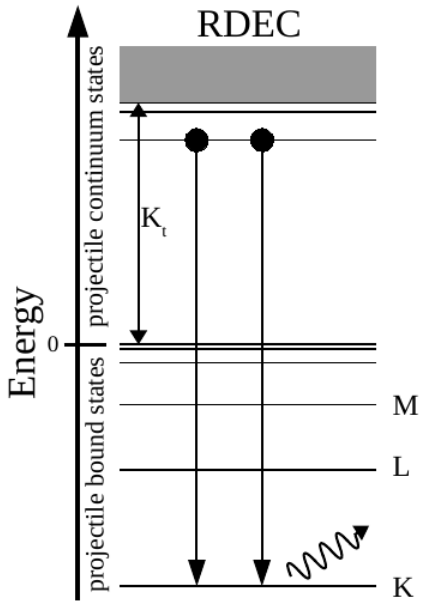

Figure 1. Energy schematic of the RDEC process. Two electrons are captured from bound states in the target (projectile continuum) to bound states in the projectile simultaneous with the emission of a single photon. Here, capture to the projectile KK states is shown. In general, the electrons may be captured from any target bound states to any available projectile bound states.

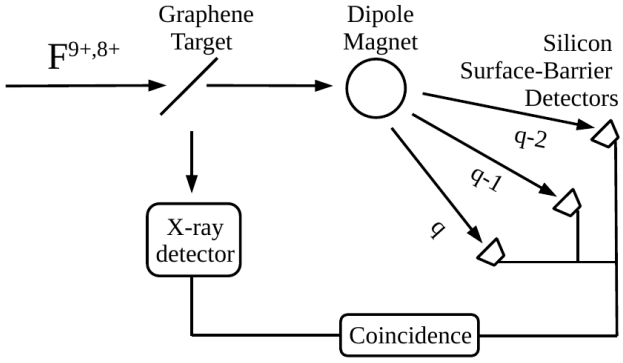

Figure 2. Schematic of the experimental setup following the accelerator facility. The charge-changed particles following projectile collisions with the target are denoted by q, q-1, and q-2.

Table 1. Calculated energies (eV) of RDEC for at least one electron going to the projectile K shell for 40 MeV (2.11 MeV/u) $F^{9+,8+}$ ions incident on graphene. Transitions with both electrons going to the K shell are not possible for the H-like projectiles. V refers to valence (quasi-free) electrons.

RDEC Transition	F^{9+}	F^{8+}
VV→KK	4333	-
VK→KK	4056	-
KK→KK	3779	-
VV→KL	3615	3414
VK→KL	3338	3137
KK→KL	3061	2859

3. Results

Examples of raw X-ray and particle/X-ray spectra (particles in coincidence with X-rays) for 2.11 MeV/u F^{9+} on graphene are shown in Figure 3. The X-ray spectrum Figure 3a represent the total X-rays emitted and include all the events associated with the charge states q-2, q-1, and q. The particle event spectra show that the q-1 counts (Figure 3c) are the most probable, with the q counts (Figure 3d) being slightly less and the q-2 (Figure 3b) counts having the smallest number with about one-quarter that of the q-1. These results are roughly consistent with the thin-foil carbon results [11]. Unfortunately, sparse beamtime allowed for only totals of about 0.02×10^{12} particles for F^{9+} and F^{8+}. In contrast, about 1.0×10^{12} particles (50 times as many) were collected for each projectile with the thin-foil C target [11]. Separately, the spectra of Figure 3 do not reveal RDEC.

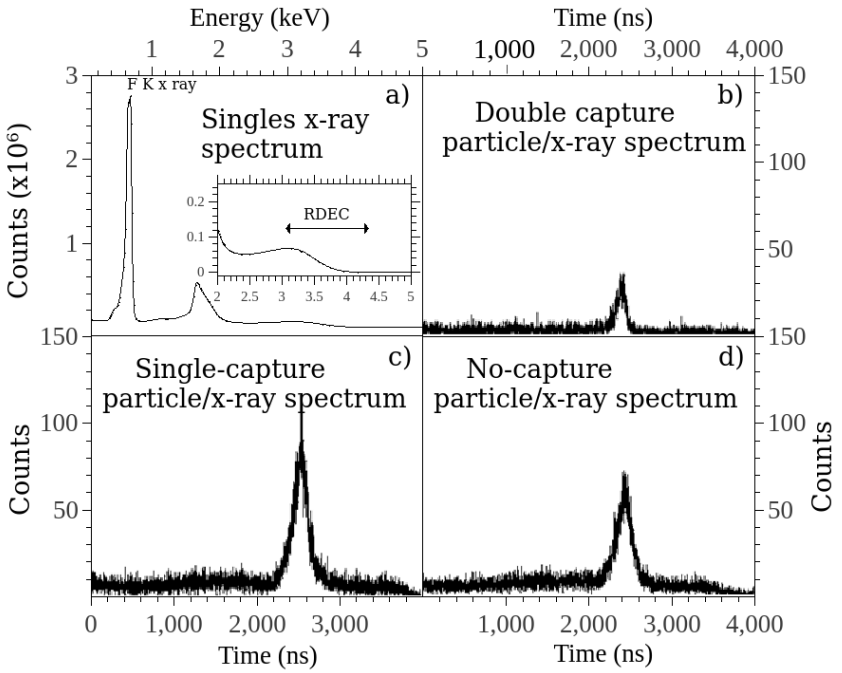

Figure 3. Representative raw spectra for 2.11 MeV/u F^{9+} on graphene. Shown are the sums of (**a**) X-ray events, (**b**) no-charge change particle events, (**c**) singly-charge changed particle events, and (**d**) doubly-charge changed particle events.

To observe RDEC, in this instance one must assign the charge-changed particles to X rays within the appropriate energy range (see Table 1, expanded by ±100 eV to allow for the Compton profile of the transition [18]). This process eliminates the particles coincident with characteristic X-rays from the projectile and target, therefore revealing a peak at an earlier time (higher energy) in Figure 4a,c than those seen in Figure 3b–d. Due to the low number of counts obtained in the limited beamtime, the X-ray spectra associated with charge-changed particle events do not reveal significant counts after background

subtraction. Double capture events associated with RDEC are shown in Figure 4. Figure 4a is the spectrum for the F^{9+} projectile and shows ∼7–8 counts near time channel 1950 ns after background subtraction. Figure 4b shows no counts for the null run (no graphene on the target), as expected. Figure 4c shows the F^{8+} spectrum, with possibly one count, but no definitive results in the region of interest.

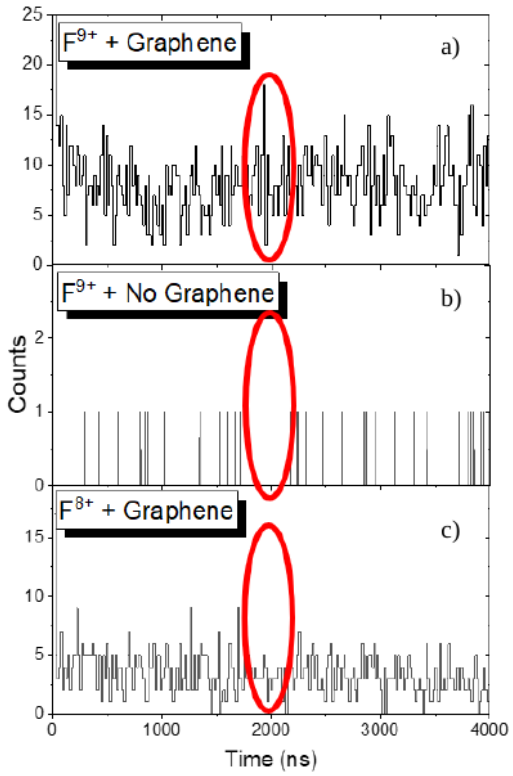

Figure 4. Sorted doubly charge-changed particle spectra associated with RDEC energy photons: (**a**) 2.11 MeV/u F^{9+} on graphene; (**b**) 2.11 MeV/u F^{9+} with no target (null run); and (**c**) 2.11 MeV/u F^{8+} on graphene.

4. Discussion and Conclusions

Given the counts displayed in Figure 4 and the total number of incident particles, tentative cross sections can be calculated for both projectiles on graphene. The most reliable certainly are those for the F^{9+} projectile, with a differential value at 90° of 1.3 b/sr, corresponding to a total cross section of 11 b, assuming isotropy. For the F^{8+} projectile, a fractional count in the region of interest was calculated after the appropriate background subtraction over the region of interest. A cross section can be estimated by assuming a single count in the spectrum for F^{8+} (panel Figure 4c). Such an assumption gives a value of $<$0.16 b/sr, corresponding to a total cross section of $<$1.4 b. Given the dearth of statistics, we assign a total error of 75% as a safe upper limit to our cross sections. These values are displayed in Figure 5 and are compared with the experimental values for the gas and thin-foil targets, along with the most accurate theoretical predictions to date [19].

In summary, RDEC has been investigated for 2.11 MeV/u F^{9+} and F^{8+} projectiles colliding with the two-dimensional target graphene, hopefully serving to bridge the gap between one-dimensional (gaseous) and three-dimensional targets (thin-foil C). The value for F^{9+} incident on graphene reported here are more in line with earlier values found for the carbon target, and are larger by a factor of about four than for the gas targets. Given the thickness of the target, these values appear reasonable.

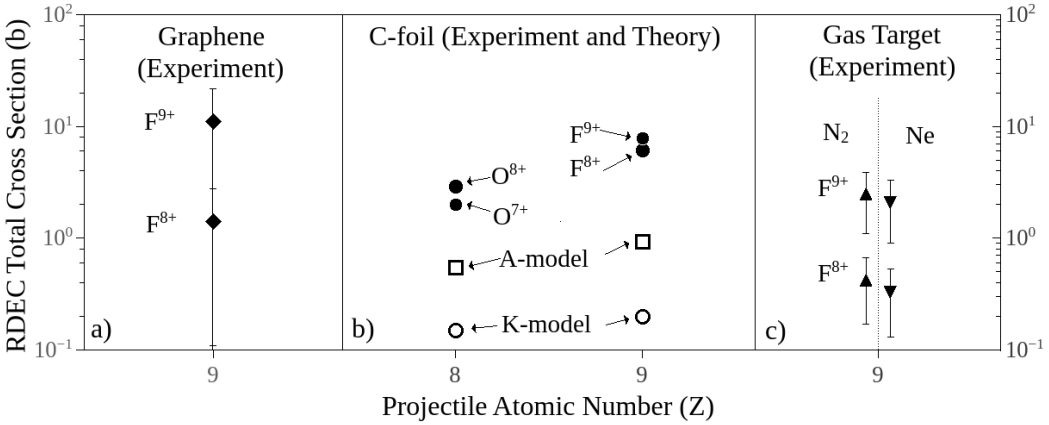

Figure 5. Present tentative cross sections for 2.11 MeV/u F^{9+} and F^{8+} on graphene (**a**), compared with our earlier experimental results for $O^{8+,7+}$ (Z = 8) and $F^{9+,8+}$ (Z = 9) and thin-foil C target [11] and the most accurate theoretical model to date (this includes two, labeled A, which includes all target electrons, and K, which includes only the innermost target electrons) (**b**), and $F^{9+,8+}$ and gas N_2 and Ne targets (**c**) [10].

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