An energetic (e,2e) reaction away from the Bethe ridge: recoil versus binary

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Abstract
We analyse the recoil-to-binary (RB) peak intensity ratio in an energetic (e,2e) reaction performed on the valence ns sub-shell of noble gas atoms away from the Bethe ridge condition. A qualitative change in the RB ratio dependence on the ejected electron energy from He to Ar can be explained by the variation of reflectivity of the short-range Hartree–Fock potential. The reflectivity increases profoundly from lighter (He) to heavier (Ne and Ar) noble gas atoms because of modification of the scattering phases due to occupation of the target p orbitals (Levinson–Seaton theorem). This effect is further modified due to strong inter-shell correlations in Ar. These theoretical predictions are confirmed experimentally.

(Kinematical studies of electron impact atomic ionization, also known as the (e,2e) reaction, brought a wealth of information on the atomic structure and electron dynamics. Over the past 40 years, since the pioneering work of Ehrhardt \textit{et al} (1969), lighter atomic targets such as hydrogen and helium have been thoroughly studied. Indeed, (e,2e) on atomic hydrogen is now considered to be a solved problem (Rescigno \textit{et al} 1999), and significant progress has been made in describing the ionization of two-electron helium-like targets (Bray \textit{et al} 2002).

Now the focus of (e,2e) studies is shifting towards heavier atomic targets. The noble gas atoms are attractive candidates for this research because of a relatively simple closed-shell electronic structure. In the past decade, the (e,2e) reaction, in both the symmetric and the non-symmetric kinematics, was measured extensively on Ne, Ar and Xe valence shells at an incident energy typically 10 times larger than the corresponding ionization potential (Haynes and Lohmann 2000, 2001a,b, Biava \textit{et al} 2002, Haynes \textit{et al} 2003, Stevenson \textit{et al} 2005, 2009, Stevenson and Lohmann 2006, 2008). Various theoretical models were applied to describe these experimental data: the distorted wave Born approximation (DWBA) (McCarthy 1995), three-body distorted-wave Born approximation (3DWBA) (Jones and Madison 1994), DWBA corrected for the post-collision interaction (PCI) by a simple Gamow factor (DWBA-G) (Kheifets \textit{et al} 2008), DWBA-R-matrix calculation (Bartschat and Burke 1987) and convergent close-coupling (CCC) method (Bray and Fursa 1996). Agreement between theory and experiment was satisfactory at large ejected electron energies but gradually deteriorated as this energy was lowered to just few eV. The np shell ionization was more difficult to describe theoretically as compared to the ns shell. The non-perturbative CCC calculation could not be performed in the former case. The latest, presumably most sophisticated, calculation on the 3p shell of Ar by Otranto (2009) failed to reproduce the experimental data. Thus, the present experimental situation remains challenging with a ‘complete’ scattering theory, valid for all possible energies and any degree of target complexity, being some way off.

Another recent line of (e,2e) research explored the valence shell ionization of noble gas atoms at a larger incident energy with the ejected electron detector being tuned to 500 eV (Catoire \textit{et al} 2006, Naja \textit{et al} 2008, Kheifets \textit{et al} 2008, Staicu Casagrande \textit{et al} 2008). As compared to a lower
incident energy (e,2e) reaction, these moderate incident energy measurements are particularly useful for bridging between the high energy, keV domain where the success of theoretical models is well documented (e.g. Lahmam-Bennani et al. (1988)) and the low energy domain (about 100 eV or below), thus allowing us to quantitatively establish the range and the frontiers of validity of the various theoretical descriptions.

Besides, it can be quite revealing as far as the qualitative mechanisms of electron impact ionization are concerned. In particular, it was observed in a recent study by Kheifets et al. (2008) that the recoil-to-binary (RB) peak intensity ratio as a function of the ejected electron energy behaved very differently for He and Ne in identical kinematics. The chosen kinematics did not satisfy the Bethe ridge condition, i.e. the magnitude of the momentum transfer was not equal to that of the sum momentum of the emitted electrons. In this situation, the ejected electron, produced in the initial binary collision, should undergo a recoil scattering from the singly ionized target in order to form the recoil peak on the same side of the incident electron beam. However, the nucleus scattering alone is not sufficient to explain a different RB energy dependence in He and Ne at identical kinematics. It was suggested that this difference could be attributed to a different reflectivity of the short-range Hartree–Fock potential. The reflectivity increases profoundly from He to Ne because of modification of the scattering phases due to occupation of the target 2p orbital as prescribed by the Levinson–Seaton theorem (Seaton 1983).

In the present work, to prove this hypothesis conclusively, we perform a systematic experimental and theoretical investigation of the RB ratio across a wide range of ejected electron energies through a broader range of targets including the He 1s, Ne 2s and Ar 3s shells. As in the previous work (Kheifets et al. 2008), the scattered electron energy and angle were fixed at respectively 500 eV and $\theta_b = -6^\circ$ measured as a function of the ejected electron angle $\theta_e$. The experimental data are plotted with error bars. Vertical arrows indicate the frontiers of validity of the various theoretical descriptions.

The results of the helium measurements are shown in figure 1 for several fixed ejected electron energies. The experimental data for $E_b = 37$, 74 and 205 eV were reported earlier (Staicu Casagrande et al. 2008). These data are normalized to the respective CCC calculations presented in the same work. The measurements at the lowest ejected energy of 12 eV (not shown in the figure) and 17 eV are new. These data are normalized to the presently performed DWBA calculations which were run systematically across the whole range of the ejected electron energies. Another set of calculations was performed using a modified DWBA-G model (Kheifets et al. 2008). The results of these calculations are also shown in figure 1. As the Gamow factor violates the normalization, the DWBA-G results are normalized to the CCC calculation (where available) or DWBA. Figure 1 shows that there is a good agreement between the experiment and the two sets of calculations. A small shift can be seen in the position of the experimental binary peak relative to the theory predictions at $E_b = 17$ and 205 eV. Also, the DWBA-G model tends to overestimate the recoil peak intensity as the ejected electron energy grows.

An analogous set of TDCS results is presented in figure 2 for the Ne 2s shell. The experimental data for Ne are normalized to the DWBA calculation and the CCC results (for 74 eV) reported by Naja et al. (2008). The DWBA-G calculation shown in figure 2 is in fair agreement with experiment except for the largest ejected electron energy of 205 eV where the recoil peak in experiment is displaced further away from the direction of the momentum transfer $-\vec{q}$ (shown as an arrow in the figure). At this kinematics, the ejected and scattered electrons acquire comparable energies and their PCI becomes strong. It may not be accounted fully by a simplified Gamow factor.

The manifest difference, which becomes obvious when comparing figures 1 and 2, is increasing relative recoil peak intensity with energy in Ne which, conversely, decreases in He. This fact was pointed out in our previous work (Kheifets et al. 2008) where the Ne 2s TDCCs were analysed at $E_b = 37$ and 74 eV. Here, we expand our measurement range by adding additional data points at $E_b = 17$ and 205 eV to see this tendency more clearly. In the whole ejected electron energy range studied here, the RB ratio decreases with energy in He and increases in Ne.

![Figure 1. TDCS of (e,2e) on He 1s at the scattered electron energy $E_b = 500$ eV and angle $\theta_b = -6^\circ$ measured as a function of the ejected electron angle $\theta_e$. The experimental data are plotted with error bars. Vertical arrows indicate the frontiers of validity of the various theoretical descriptions.](image-url)
energies of 12 and 17 eV. The RB ratio seems to be flattening at small ejected electron energies, with considerable growth towards larger ejected electron energies. Also, at the energies between 17 and 37 eV is of statistical nature (we recall that 17 eV). We therefore believe that the small bump occurring at 17 eV. We therefore believe that the small bump occurring at 17 eV where the theory predicts a relatively large recoil peak in a blunt contradiction to the experiment. Also, at the energies $E_b = 37$ and 74 eV, the theory predicts a prominent central peak positioned in between the binary and recoil peaks which is not confirmed experimentally.

These findings are summarized in figure 4 where the RB ratios are plotted versus the ejected electron energy for all three targets. The experimental RB ratio for He decreases with increasing ejected electron energy, despite the reasonably small scattering of the two data points at 17 and 37 eV below and above a monotonous decrease, respectively. The CCC results, where available, show a gradual decline of the RB ratio above 37 eV as does the present experiment. A monotonous behavior was also reported in earlier measurement by Jung et al. (1985) in the ejected energy range 2.5–10 eV, and is confirmed by our present DWBA calculation (not presented here). This is in qualitative agreement with the present experimental observation of a decrease in the RB ratio between 12 and 17 eV. We therefore believe that the small bump occurring between 17 and 37 eV is of statistical nature (we recall that the quoted error bars only represent one standard deviation).

Quite conversely, the experimental RB ratio in Ne steadily grows across the whole studied ejected electron energy range. As has already been pointed out earlier, the corresponding RB ratio in Ar is non-monotonous. There is a hint of a minimum in between the experimental points at $E_b = 37$ and 74 eV and a considerable growth towards larger ejected electron energies. The RB ratio seems to be flattening at small ejected electron energies of 12 and 17 eV.

In our earlier paper (Kheifets et al. 2008), in order to answer the question why the recoil peak intensity decreased with the ejected electron energy in He and increased in Ne, we turned to the partial wave analysis following Vriens (1969). Most straightforwardly, this analysis can be carried out in the plane wave Born approximation (PWBA) in which the energetic projectile is represented by a plane wave. In this approximation, the vector of the momentum transfer $q$ becomes a quantization axis. The ionization amplitude can be expanded over the Legendre polynomials with respect to the azimuthal angle of the ejected electron $\theta = \theta_b - \theta_q$ counted relative to $q$:

$$\langle k_b | \exp(iqr) | n s \rangle = \sum_{l=0}^{l_{\text{max}}} a_l P_l(\cos \theta)$$

$$= \left( a_0 - \frac{1}{2} a_2 \right) + \left( a_1 - \frac{3}{2} a_2 \right) \cos \theta$$

$$+ \frac{3}{2} a_2 \cos^2 \theta + \frac{5}{2} a_3 \cos^3 \theta.$$  (1)

For the ns-shell ionization, $a_l \propto e^{ib} (2l+1) \int \, d\mathbf{r} \, P_{E_l j_l} (qr) P_n s$. We truncate this expansion at $l_{\text{max}} \approx k_b R_{ns} \approx 3$ which is appropriate in the studied ejected electron energy range for the valence shell radius in noble gases $R_{ns} \approx 1$. The binary and recoil peaks correspond to $\theta = 0$ and $\pi$, respectively. A near zero minimum around $\theta = \pi/2$, most obvious on the He plots of figure 1, can occur only when $a_0 - \frac{1}{2} a_2 \approx 0$. Under this condition, the ionization amplitude (1) reduces to

$$\langle k_b | \exp(iqr) | n s \rangle = \left[ \left( a_1 - \frac{3}{2} a_3 \right) + \frac{3}{2} a_2 \cos \theta + \frac{5}{2} a_3 \cos^2 \theta \right] \cos \theta.$$  (2)

Comparable intensities of the binary and recoil peaks ($\cos \theta = \pm 1$), as is the case for the Ne 2s^2 ionization, require that

The corresponding TDCS plots for the Ar 3s shell are shown in figure 3. Here, the situation is more complex. The experimental RB ratio is minimal at $E_b = 74$ eV, increasing towards larger and smaller ejected electron energies. This tendency is confirmed by the DWBA-G model except for the lowest $E_b = 17$ eV where the theory predicts a relatively large recoil peak in a blunt contradiction to the experiment. Also, at the energies $E_b = 37$ and 74 eV, the theory predicts a prominent central peak positioned in between the binary and recoil peaks which is not confirmed experimentally.

Quite conversely, the experimental RB ratio in Ne steadily grows across the whole studied ejected electron energy range. As has already been pointed out earlier, the corresponding RB ratio in Ar is non-monotonous. There is a hint of a minimum in between the experimental points at $E_b = 37$ and 74 eV and a considerable growth towards larger ejected electron energies. The RB ratio seems to be flattening at small ejected electron energies of 12 and 17 eV.
\[ |a_1 + \frac{1}{2}a_2 + a_3| \approx |a_1 - \frac{1}{2}a_2 + a_3| \text{ and either } a_2 \text{ is very small (which implies that } a_0 \text{ is very small) or is shifted in phase with respect to } a_1 + a_3 \text{ by a factor } \pi/2. \]

In figure 5, we plot the ejected electron phases \( \delta_l \), \( l = 0, \ldots, 3 \), calculated in the field of the corresponding singly charged ion. These phases determine the phases of the complex coefficients \( a_l \) since the radial integrals in their definition are real. We note a manifest difference between the scattering phases for He\(^+\) (left panel), Ne\(^+\) (central panel) and Ar\(^+\) (right panel). In the case of He\(^+\), the phases \( \delta_l \), \( l = 1, \ldots, 3 \), are close together whereas the s-phase \( \delta_0 \) is far apart. This can be explained by the presence of the occupied 1s state in the ionic core. The phase shift due to the short range potential, i.e. the difference of the total phase and the Coulomb phase, is related to the quantum defect according to the Levinson–Seaton theorem \( \delta_l(k \to 0) - \sigma_l(k \to 0) = \mu_l(\infty)\pi \) (Seaton 1983). The quantum defect \( \mu_l(\infty) \) is determined by fitting the sequence of energy levels with a given orbital momentum \( E_{nl} = 0.5Z_{\text{eff}}^l [n - \mu_l(n)] \) with the effective charge of the screened nucleus \( Z_{\text{eff}} \approx 1 \). The Coulomb phase is given by the expression \( \sigma_l(k) = \arg \Gamma(1 + l - iz_{\text{eff}}/k) \). The presence of the bound target state with a given \( l \) perturbs the energy level sequence and results in a large quantum defect. This is the case of the s-phase in He\(^+\). The other \( l \)-phases are quite close and tend to converge at larger energies as \( \sigma_{a2}(k) = \sigma_{a3}(k) - \text{arctan} \frac{Z_{\text{eff}}}{k(l + 1)} \). Because of this phase behaviour, the coefficients \( a_1, a_2 \) and \( a_3 \) are more or less collinear and the recoil intensity is fairly small relative to the binary ratio.

The situation in Ne\(^+\) is very different because of the bound 2p state which results in a large quantum defect and the p-phase deviating strongly from the d- and f-phases. We see on the central panel of figure 5 that the phase difference between the p- and d-phases reaches \( \pi/2 \) at an energy of about 100 eV which is a prerequisite of the orthogonality of \( a_1 + a_3 \) and \( a_2 \) (\( a_3 \) is relatively small). Thus, we have a large RB ratio which increases with energy up to about 100 eV. This is exactly the behaviour which is predicted by the PWBA model on the central panel of figure 4. The DWBA calculation changes this behaviour insignificantly. A stronger change is caused by the PCI effects which favour the recoil peak at the expense of the binary since in the recoil direction, the scattered and ejected electrons are much further apart. That is why the DWBA-G calculation predicts the constantly growing RB ratio as is indeed the case experimentally.

Further disturbance of the ejected electron phases is seen in Ar due to multiple occupancy of the s and p target orbitals. The phase difference between the p and d waves reaches \( \pi \) at \( \sim 30 \) eV ejected electron energy and then decreases to \( \pi/2 \) at around 200 eV. Correspondingly, the RB ratio, as prescribed by the PWBA model, reaches a deep minimum at \( \sim 30 \) eV and then recovers to a broad maximum at around 200 eV. The distortion and PCI effects change this behaviour and the RB ratio continues to grow at larger ejected electron energies which is in accordance with the experiment.

However, a strong peak of the RB ratio predicted theoretically at small energies contradicts experimental observations. To explain this deviation, we have to take...
into account a strong inter-shell correlation between the 3s and 3p electrons which is accounted for within the random phase approximation (RPA). This type of correlation is known to significantly modify the photoionization cross-section of the ns subvalence shells of heavier noble gas atoms (Ar and Xe) due to strong polarizability of the outermost np sub-shell (Amusia 1990). The 3p shell in Ar is strongly polarizable due to a large overlap with an unoccupied 3d shell. Such an overlap is significantly smaller for the Ne 2p shell because of a difference in principle quantum numbers. When the RPA correlation is taken into account using the method described by Kheifets et al. (2008), the RB ratio becomes much closer to the experimental data points as is seen on the right panel of figure 4. Unfortunately, this type of calculation can only be carried out for small ejected electron energies as the growing number of necessary partial waves renders this procedure very computationally intensive. Fortunately, this type of correlation is known to be prevalent at this particular energy range.

In conclusion, we analyse the recoil-to-binary (RB) peak intensity ratio in the energetic (e,2e) reaction on the He 1s, Ne 2s and Ar 3s outer valence sub-shells. The RB ratio, as a function of the ejected electron energy, is manifestly different in He, Ne and Ar where it respectively decreases, increases and has a minimum due to a changing occupancy of the target s and p orbitals. In Ar, the electronic structure of the target leads to a strong inter-shell correlation due to a significant polarizability of the outermost 3p valence sub-shell which overlaps readily with the vacant 3d shell. Qualitatively, formation of the recoil peak can be viewed as reflection of the ejected electron from the ionized target potential. The smooth Coulomb potential of the singly charged ion is a poor reflector. It is the Hartree–Fock potential of the target core orbitals that can be quite an effective reflector due to specific scattering phase behavior imposed by the Levinson–Seaton theorem.

**References**

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Queries

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