

Multiply differential study of detachment with simultaneous target excitation in anion-atom collisions

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We have measured momenta of electrons and recoil ions in triple coincidence with neutralized projectiles for slow collisions between various anions and target atoms. From the data we extracted triple differential momentum and angular distributions of electrons ejected in detachment, in detachment with simultaneous target ionization (DI), and in detachment with simultaneous target excitation (DE). The DI rates relative to the DE rates depend sensitively on the projectile speed and on the properties of the target.

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I. INTRODUCTION

Reaction microscopes (ReMi's) have proven their potential for advancing our understanding of the dynamics underlying atomic processes (for reviews see, e.g., Refs. [1–3]). By measuring momentum-analyzed fragments, produced in a collision or by photoabsorption, in coincidence, and taking advantage of the very large detection efficiencies of ReMi's, multiple differential cross sections have been measured with unprecedented detail for many processes leading to a charged recoiling target ion. Another important advantage of ReMi's is their large flexibility: they have been used to study ionization and capture processes induced by photon- [4], electron- [5,6], and ion impact [7,8] (for a recent review see Ref. [9]). Perhaps the biggest limitation of most ReMi's is that only charged particles can be detected. This means that processes leaving the target in a neutral state, e.g., target excitation, are very difficult, but not impossible to study, as we will demonstrate in this article.

A major recent advancement of the ReMi technology was the development of an in-ring apparatus for use at the cryogenic storage ring (CSR) in Heidelberg, Germany [10]. Due to the large beam intensities and excellent vacuum conditions, the CSR offers the possibility to measure very small cross sections with a very low background level. The combination of a ReMi with the CSR thus promises a new generation of experiments. We recently performed successful proof-of-principle experiments and presented multiple differential cross sections for electron detachment from various anions colliding with neutral target atoms and molecules [11]. More recently, we reported triple differential momentum distributions of electrons ejected in detachment with simultaneous ionization of the target (DI) in the same collision systems [12].

Some of the observations reported in both studies were qualitatively consistent with the quasifree-electron model. There, the initial bond of the electron to the anion was neglected and detachment was treated as elastic scattering of the electron from the target. Likewise, DI is considered as target ionization by electron impact, also known as an (e,2e) process. In Ref. [12] we referred to this process as the correlated first-order DI channel. In the case of detachment, we found momentum distributions exhibiting pronounced structures at electron momenta with a magnitude equal to the projectile speed v_p , which corresponds to kinetic energy conservation for elastic scattering. In the case of DI, a sharp maximum at zero momentum was observed. This is due to the energy loss the projectile electrons suffer due to the ionization potential of the target. In fact, the projectile speed was too small to give the electron sufficient energy to ionize the target. In other words, the projectile speed was below the threshold for truly free-electron impact ionization and the (e,2e) process was only possible due to the momentum distribution (Compton profile) of the electron in the initial bound state (neglected in the quasifree-electron model). Even if the (e,2e) process does occur due to the Compton profile, the most likely scenario is that essentially no energy is left for either of the 2 ejected electrons, leading to the maximum at zero momentum.

Another process which, with our experimental setup, cannot directly be distinguished from detachment is detachment with simultaneous excitation of the neutralized projectile or of the target (DE). DI can be distinguished from detachment because the recoil ions are detected; however, excitation does not lead to a recoil ion. The results for detachment are not significantly affected by the presence of the DE channel because the cross section is expected to be small compared to detachment. However, the DE cross section is not necessarily negligible compared to the DI cross section. Therefore, DE is an interesting research topic in its own right. While a rich literature exists on detachment (e.g., Refs. [11,13–28]), not much work has been reported on DE yet [29,30].

Here, we report such measured multiple differential momentum and angular distributions for excitation of the target.

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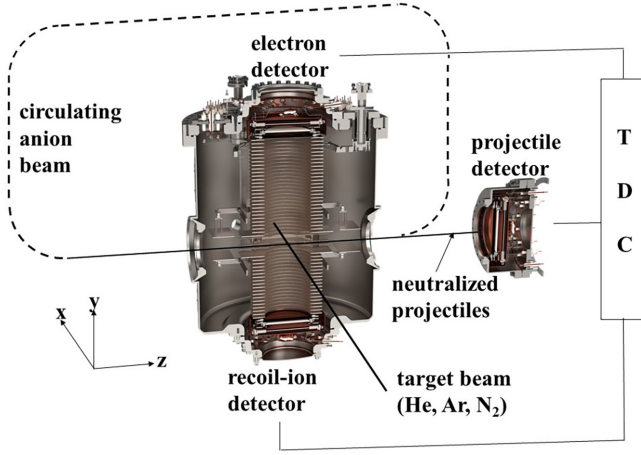


FIG. 1. Schematic sketch of the experimental setup.

The data were obtained by simultaneously analyzing double coincidences between the neutralized projectiles and the ejected electrons and triple coincidences between the neutralized projectiles, the ejected electrons, and the recoil ions. As expected, we find that the ratio between the DI and DE cross sections increases with increasing projectile speed and with decreasing binding energy of the target.

II. EXPERIMENTAL SETUP

The experiment was performed at the cryogenic storage ring (CSR) at the Max-Planck-Institute for Nuclear Physics in Heidelberg. The details were presented previously [11,12] and a schematic sketch of the experimental setup is presented in Fig. 1. In brief, various species of anions (Si^- , C^- , and C_2^-) with energies of 30 and 300 keV were injected into the CSR and stored for 30 s. The vacuum in the ring was better than 10^{-12} mbar so that data could be taken essentially free of background from the residual gas. The projectile beam was crossed at an angle of 90° with a very cold target beam from a supersonic jet. The projectiles neutralized in the intersection region were detected by a two-dimensional position-sensitive microchannel plate (MCP) detector with an open area ratio of 0.65 and an active diameter of 120 mm.

The electrons and recoil ions produced in the collisions were extracted by a weak electric field of about 2 V/cm directed perpendicularly to both the projectile and target beams. They were then detected by two MCP detectors essentially identical to the projectile detector. A uniform magnetic field of about 5 G forced the electrons into cyclotron motion. For a momentum component perpendicular to the electric extraction field smaller than 1.5 a.u. (atomic units), this magnetic field was strong enough to confine the electrons to the detector size. In order to optimize the extraction field conditions, two grids were placed in front of the electron and recoil-ion detectors, each with a transmission of 75%. Along with the open area ratio this yields an overall detection efficiency for each ReMi detector of 35%, while the projectile detector had a larger efficiency of 0.65 because of the missing grids. All three detectors were set in coincidence with each other.

The electron and recoil-ion momentum components in the y direction (defined by the extraction field direction) and the

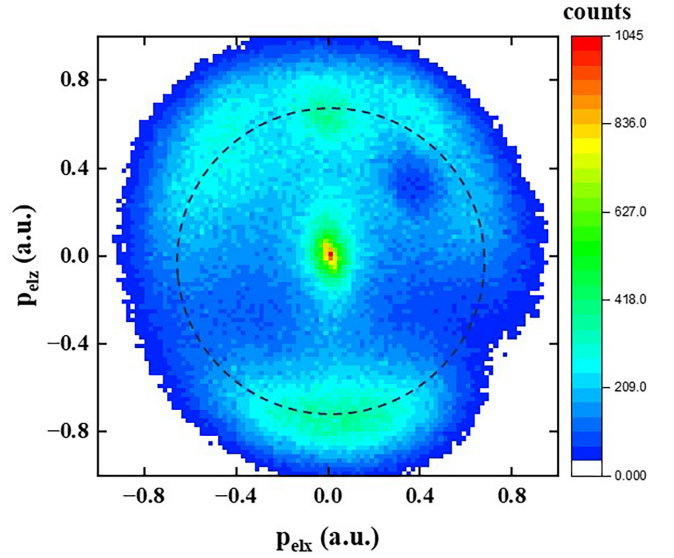


FIG. 2. Triple differential momentum distribution of electrons ejected in 300-keV $\text{Si}^- + \text{Ar}$ collisions and measured in coincidence with the neutralized projectiles as a function of the x - and z components. The z component is in the direction of the projectile beam. The y component was restricted to $|p_{ely}| < 0.05$ a.u.

z direction (defined by the projectile beam direction) were obtained from the position information of the respective detectors. In the x direction (defined by the target beam direction), the momentum components were determined by the coincidence time between the respective particle and the neutralized projectile.

III. RESULTS AND DISCUSSION

Before we present the results for the triple coincidences, we first review the data extracted from the neutralized projectile-electron double coincidences which we reported previously [11,12]. In Fig. 2 we present a two-dimensional momentum distribution of electrons ejected in 300-keV $\text{Si}^- + \text{Ar}$ collisions. The horizontal axis represents the x component (determined by the target beam direction) and the vertical axis the z component (determined by the projectile beam direction) of the momentum. The spectrum was generated under the condition that the y component of the momentum has a magnitude smaller than 0.05 a.u. Therefore, the momentum distribution of Fig. 2 is proportional to the triple differential cross section $d^3\sigma/d\mathbf{p}^3$.

Two features are quite prominent in Fig. 2: (a) a ringlike structure with a radius corresponding to the projectile speed v_p (indicated by the circle in Fig. 2) and (b) a sharp peak at zero momentum. The first feature, which we discussed in some detail in Ref. [11], can be understood within the quasifree-electron model. As mentioned in the Introduction, detachment can then be viewed as elastic scattering of the electron off the target so that its speed is constant at v_p . The maximum at zero momentum cannot be explained within the quasifree-electron model, unless detachment is accompanied by a transition of a second electron.

Three processes can contribute to the peak structure at zero momentum. The first contributor is a higher-order detachment process, which is perhaps best viewed in the rest frame of the projectile. It involves at least two interactions of the target core (moving at v_p) with the active projectile electron. In the first interaction, the target transfers sufficient energy to the electron to be released from the projectile (which is at rest). In the second interaction the target core and the electron attract each other so that the electron gets accelerated towards the target velocity in the projectile frame. This leads to an enhancement of the electron flux at the target velocity in the projectile frame or at zero in the laboratory frame. For target ionization by projectile impact the same process (in reversed kinematics) can be important and is known as postcollision interaction (PCI), e.g., Refs. [31–33]. Here, we adopt the same notation for detachment from the projectile by target impact.

The second contributor is DI, which is actually suppressed in the data of Fig. 2 because we applied a condition that no valid recoil-ion was detected. However, because of the limited detection efficiency ($\sim 35\%$), this anticondition is no guarantee that no recoil-ion was produced in the collision. Earlier, we demonstrated that the momentum spectrum of electrons ejected in DI is dominated by a sharp peak structure at zero momentum [12], which is the expected behavior within the quasifree-electron model.

Finally, detachment with simultaneous excitation (DE) of either the residual projectile or the target can contribute. DE leads to the same signatures in the experiment as detachment, namely exactly one ejected electron, a neutralized projectile, and an absent recoil ion. Furthermore, detachment with simultaneous projectile excitation is not expected to significantly modify the momentum distribution of the detached electron because both reaction steps are independent of each other. In contrast, detachment with simultaneous target excitation is likely to be dominated by the same channel as DI, i.e., the correlated first-order process involving an interaction between the 2 electrons which get ejected [12]. Like for DI, in the quasifree-electron model there is a threshold for this DE channel and, since the projectile speed is below this threshold, this is expected to lead to a sharp maximum at zero momentum.

Information about the cross sections for the various contributions to the zero-momentum peak can be obtained by simultaneously analyzing the rates for projectile-electron double coincidences N_{DC} and for triple coincidences N_{TC} . The double-coincidence rate is given by

$$N_{DC} = (\sigma_{\text{det}} + \sigma_{\text{DE}})\varepsilon_e\varepsilon_p + \sigma_{\text{DI}}2\varepsilon_e(1 - \varepsilon_e)(1 - \varepsilon_r)\varepsilon_p. \quad (1)$$

Likewise, the triple-coincidence count rate is obtained from

$$N_{TC} = \sigma_{\text{DI}}2\varepsilon_e(1 - \varepsilon_e)\varepsilon_r\varepsilon_p. \quad (2)$$

Here, σ_{det} , σ_{DE} , and σ_{DI} are the triple differential cross sections for detachment, DE, and DI, respectively. The detection efficiencies for the recoil ions and projectiles are given by ε_r and ε_p , and $2\varepsilon_e(1 - \varepsilon_e)$ is the probability for detecting exactly 1 out of the 2 ejected electrons in DI. Combining Eqs. (1) and (2) yields

$$N_{DC} - (1 - \varepsilon_r)/\varepsilon_r N_{TC} = (\sigma_{\text{det}} + \sigma_{\text{DE}})\varepsilon_e\varepsilon_p. \quad (3)$$

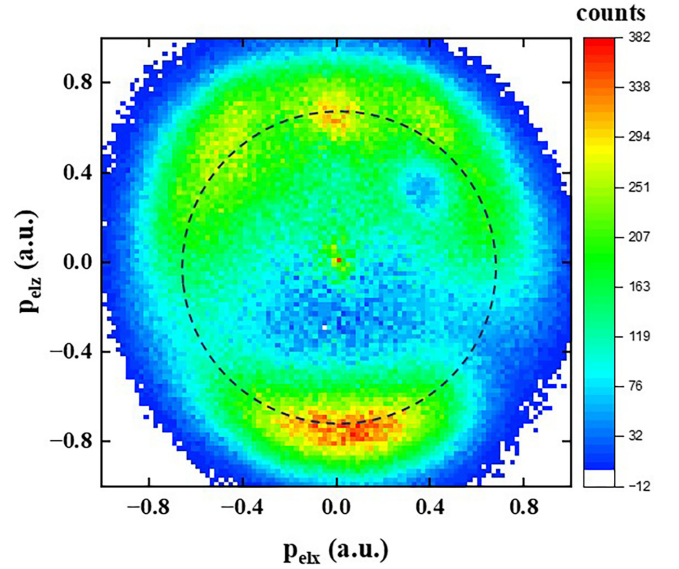


FIG. 3. Same as Fig. 2, but the rates are obtained from Eq. (3). For small p_{el} (approx. less than < 0.25 a.u.), the spectrum is thus mainly due to DI and DE (see text). For larger p_{el} , detachment is dominant with some contributions from detachment with simultaneous projectile excitation.

In Fig. 3 we present the same spectrum as shown in Fig. 2, except for the events represented by the difference of Eq. (3), where we used $\varepsilon_r = 0.35$. The strong peak at zero momentum is significantly weakened in this difference spectrum, showing that DI is a major contributor to this structure. However, the peak is not completely removed, demonstrating that either DE or detachment proceeding through PCI (in the following referred to as D_{PCI}), or both, are not negligible either.

An important question to answer is what are the relative contributions of D_{PCI} and DE to the residual zero-momentum peak in Fig. 3. Initially, in the case of target ionization, it was assumed that the occurrence of a sharp structure at $\mathbf{p}_{\text{el}} = \mathbf{v}_p$ (referred to as cusp peak) through PCI would require an attractive long-range interaction between the projectile and the ejected electron, i.e., that it would not occur for neutral or negatively charged projectiles, e.g., Ref. [34]. However, a surprisingly strong and narrow cusp structure at $\mathbf{p}_{\text{el}} = \mathbf{v}_p$ was experimentally found for neutral projectile impact [35,36]. Later, it was shown that indeed even for the short-range potential of a neutral projectile a cusp peak can be formed, but only if the projectile enters the collision in an excited metastable state or a second projectile electron gets excited during the collision. This interpretation was subsequently confirmed by two experimental studies [37,38].

Correspondingly, one would expect a cusp peak at zero momentum generated by D_{PCI} if the target is produced in a metastable state or a second target electron gets excited. However, in the supersonic jet used to produce the target beam in our setup there is no mechanism that would produce metastable target atoms in any significant number. The cross section for ionization plus excitation of the target is negligible because of the very large negative- Q value of this process. Therefore, D_{PCI} is unlikely to cause the zero-momentum peak in the data of Fig. 3. This is further supported by the triple

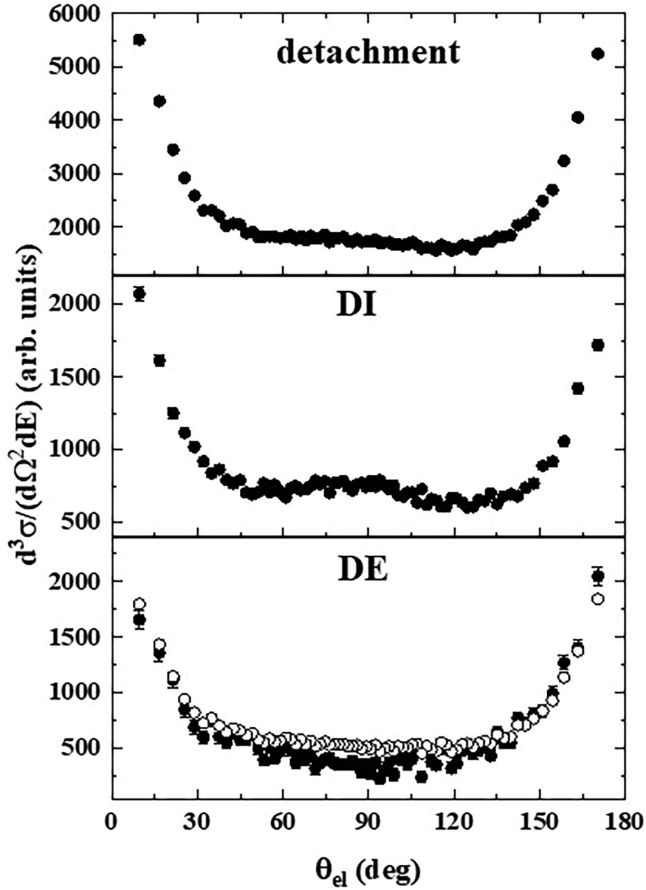


FIG. 4. Triple differential angular distribution of electrons ejected with an energy of less than 1 eV in detachment (top panel), DI (center panel), and DE (bottom panel) in 300-keV $\text{Si}^- + \text{Ar}$ collisions. For the closed symbols, the recoil-ion detector efficiency is assumed to be 0.35 and for the open symbols to be 0.5.

differential cross section $d^3\sigma/d\Omega^2 dE_{el}$, which is given by $p d^3\sigma/d\mathbf{p}^3$ and which we reported in Ref. [11]. For a long-range potential a divergence even in $d^3\sigma/d\Omega^2 dE_{el}$ leads to a cusp peak in spite of the multiplication with p . For a short-range potential due to a neutral atom, the divergence would even be stronger in the presence of a metastable excited state [36]. In contrast, in the $d^3\sigma/d\Omega^2 dE_{el}$ presented in Ref. [11] we observed a pronounced minimum at $\mathbf{p} = 0$. We therefore conclude that the zero-momentum peak in Fig. 3 is predominantly due to DE involving target excitation.

In Fig. 4 we present the angular distribution of electrons with an energy between 0 and 1 eV ejected with the condition that no recoil ion was detected (top panel), ejected in DI (center panel), and in DE (bottom panel) for 300-keV $\text{Si}^- + \text{Ar}$ collisions, where the latter was obtained from Eq. (3). In order to give a feeling about the uncertainty introduced by any inaccuracy in the detector efficiencies on the shape of the angular distribution for DE, the open circles in the bottom panel represent the result of Eq. (3) with $\varepsilon_r = 0.5$ instead of 0.35. The magnitude was matched to the one of the data for $\varepsilon_r = 0.35$. Only marginal differences can be seen between the two datasets for the different ε_r . However, the magnitude of the DE rates is obviously significantly affected by ε_r .

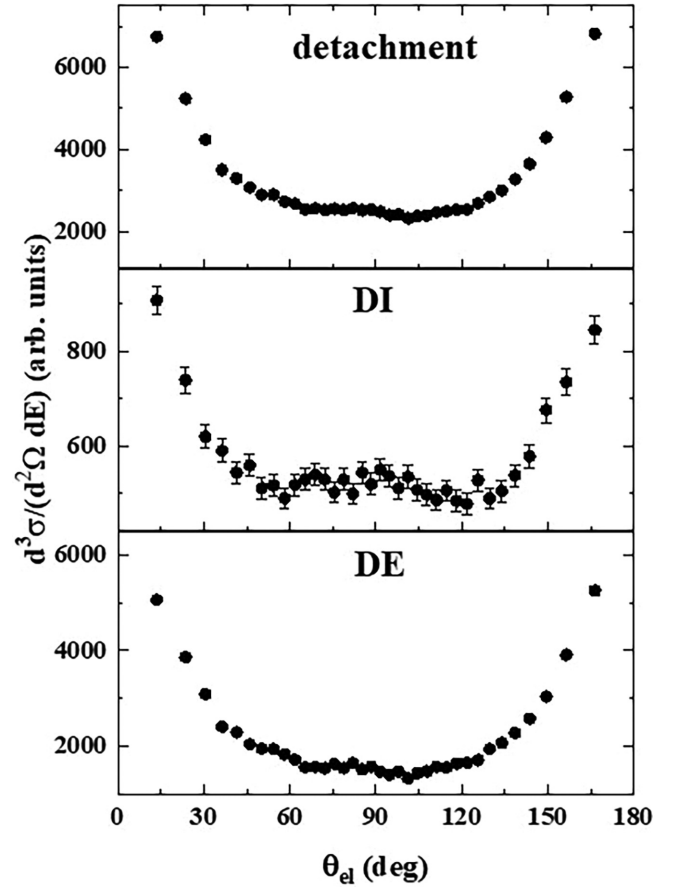
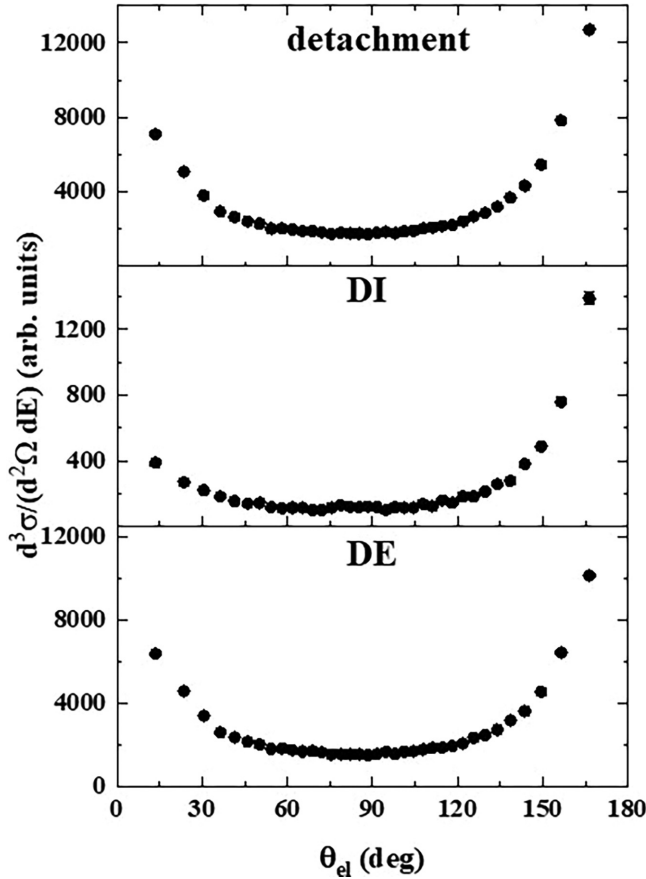


FIG. 5. Same as Fig. 4 for 30-keV $\text{C}^- + \text{Ar}$ collisions, except the electron energy was less than 0.5 eV and no data are shown for a detector efficiency of 0.5.

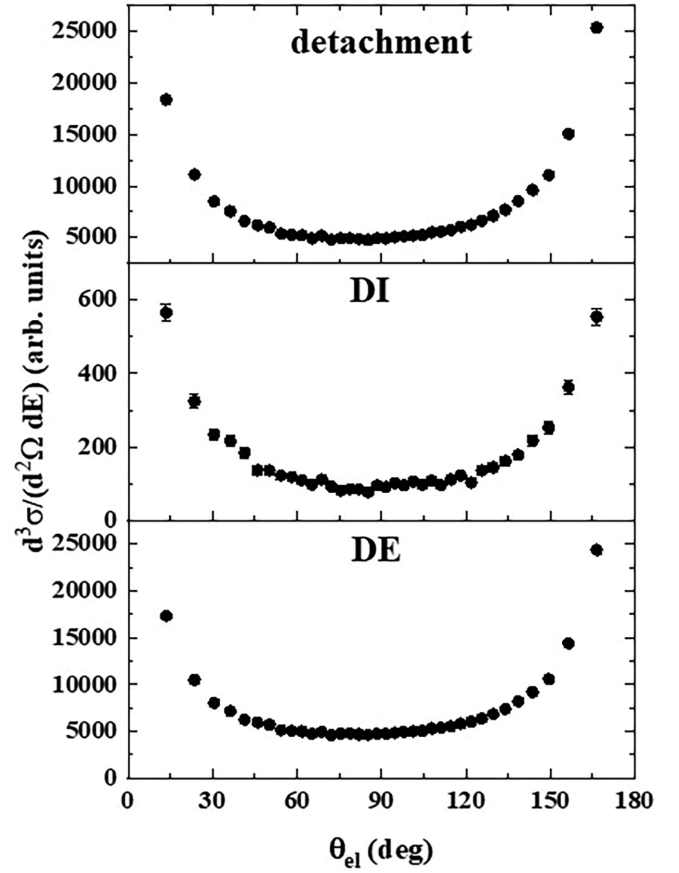
As expected, the angular distributions for DI and DE are quite similar. Both are dominated by pronounced maxima in the forward and backward directions. However, a closer inspection also reveals some differences: first, for DI a small side maximum is observed at 90°, while for DE a broad minimum is found in this region. Second, both angular distributions exhibit a forward-backward asymmetry, where the forward direction is favored by DI, but the backward direction by DE. A similar trend is observed for 30-keV $\text{C}^- + \text{Ar}$ collisions, shown in Fig. 5, except that the forward-backward asymmetry is weaker than for the Si^- projectiles. Here, and in Figs. 5 and 6, the electron energy was restricted to less than 0.5 eV because of the smaller projectile speed compared to 300-keV Si^- . Larger differences can be seen in the data for 30-keV $\text{C}_2^- + \text{Ar}$ collisions (see Fig. 6): the maximum at 90° in the DI data is nearly absent. Furthermore, both DI and DE now favor the backward- over the forward direction, where this asymmetry is quite large in the case of DI.

The comparison of Figs. 4–6 shows that although there is some variation of the shape of the angular distribution with projectile speed, this dependence is not very strong. In Figs. 7 and 8 we present the angular electron distributions for 300-keV Si^- colliding with He (Fig. 7) and N_2 (Fig. 8). These spectra demonstrate that the shape of the angular distributions is not very sensitive to the target either.

FIG. 6. Same as Fig. 5 for 30-keV $C_2^- + Ar$ collisions.

A very different picture emerges when the relative magnitude between the angular distributions for DI and DE is considered. In Fig. 9 the ratios R between the integrated counts of the angular distributions for DI and DE are shown as a function of the projectile center-of-mass energy per nucleon E_p^{CM}/amu (atomic mass unit) for Ar (closed circles), N_2 (open triangle), and He (open circle). For Ar, R increases (nearly linearly) with increasing E_p^{CM}/amu . This is the expected trend if DI and DE are predominantly caused by the correlated first-order processes. For DE the threshold is smaller than for DI so that a larger fraction of the Compton profile can lead to the transition. Therefore, for projectile energies below the DI threshold the DE cross section should be larger. Above this threshold, the cross section for both processes should become more similar and the dependence on E_p^{CM}/amu should become much flatter. Above about $E_p^{CM}/amu = 5$ keV, which is still below the DI threshold, the measured R is larger than 1. However, it should be noted that this is at least partly due to the fact that in DI 2 electrons are ejected, but in DE only 1, i.e., the probability for detecting 1 electron is twice as large for DI.

For 300-keV $Si^- + Ar$ collisions we demonstrated that DI is dominated by the correlated first-order [or (e,2e)] process [12]. It should be noted that in the present data on DE we actually select this channel by setting a condition on a small ejected electron energy. The uncorrelated second-order process leads to a similar momentum distribution as for electrons ejected in detachment. Therefore, with our experimental setup

FIG. 7. Same as Fig. 5 for 300-keV $Si^- + He$ collisions.

the uncorrelated DE mechanism cannot be distinguished from detachment and we have no information about the relative importance between the correlated and uncorrelated DE channels. However, it seems likely that the correlated channel, since it is dominant in DI, is much more important in DE as well.

For He, R is much smaller than for Ar even at the same E_p^{CM}/amu . Here, DI seems to be completely insignificant. This is not particularly surprising either. It can be explained by the much larger ionization potential of He (24.6 eV) compared to Ar (15.76 eV). While for electron-impact excitation the cross sections scale inversely with the excitation energy [39] for ionization near threshold, they scale inversely with the square of the ionization potential [40]. On the other hand, the He data were obtained for C^- projectiles, which have a broader Compton profile than Si^- . Apparently, this effect is not sufficient to compensate the larger ionization potential. A more surprising observation is that even for N_2 R is significantly smaller than for Ar, although the ionization potentials are almost the same. This shows that it is not only the target ionization potential that matters, but other aspects of the target structure need to be considered as well. Theoretically, the only other target property, apart from the energy levels, entering in the excitation and ionization cross sections is the number of valence electrons [39,40], which does not differ much between Ar and N_2 . Furthermore, in the DI-to-DE cross-section ratio the number of valence electrons cancels. One possible explanation is that the difference between an atomic and a

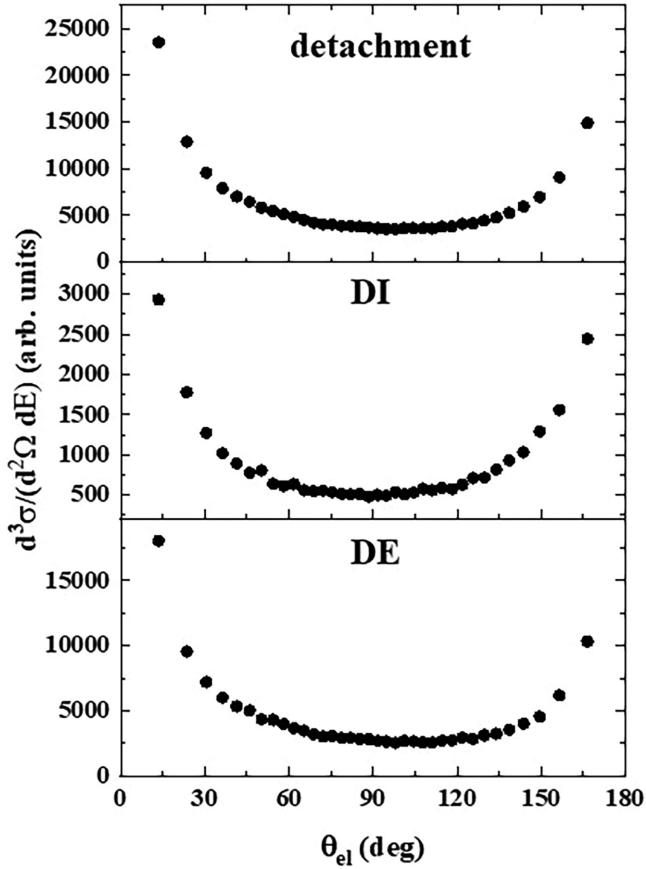


FIG. 8. Same as Fig. 5 for 300-keV $\text{Si}^- + \text{N}_2$ collisions except the electron energy was less than 1 eV.

molecular target, featuring more excited states in the relevant energy range, might play an important role. However, the system is too complex to provide a meaningful quantitative

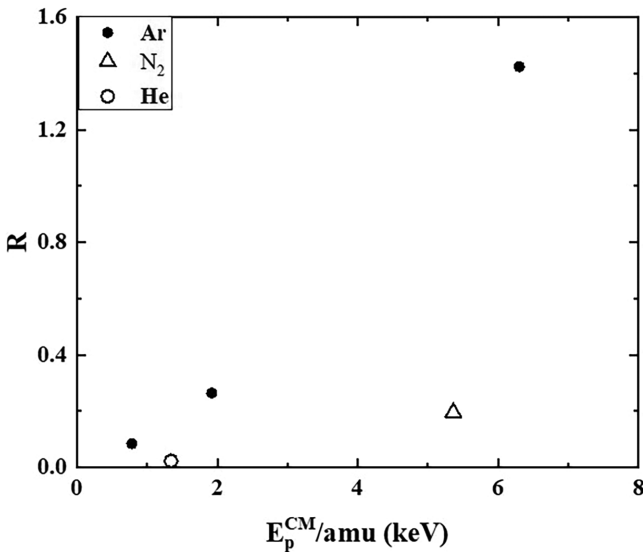


FIG. 9. Ratio between the integrated counts of Figs. 3–7 for DI and DE as a function of the center-of-mass projectile energy per nucleon.

estimate. Therefore, at present, we cannot offer a quantitative explanation for the large differences between N_2 and Ar and hope that our work will stimulate theoretical follow-up work.

IV. CONCLUSIONS

We have measured momentum-analyzed electrons and recoil ions ejected in collisions of slow anions with various targets in triple coincidence with neutralized projectiles. Double coincidences between the electrons and neutralized projectiles, with a condition that no recoil ion was detected, were also analyzed. From the data we extracted triple differential momentum distributions of electrons ejected in detachment accompanied by target excitation (DE). The data were compared to previously published results on detachment and detachment accompanied by target ionization [11,12].

At electron momenta close to the projectile speed the double-coincident momentum spectra are dominated by detachment, while at very small momenta DI and DE predominantly contribute. Only DI events contribute to the triple-coincident momentum distributions. Therefore, the momentum distributions for DE were obtained by subtracting the triple-coincident from the double-coincident data and setting a condition on small electron momenta. For all three processes we also analyzed triple differential angular distributions of the ejected electrons. While we showed earlier that the momentum- and angular distributions for detachment sensitively vary with projectile speed and target [11], this dependence is much weaker for DI and DE.

In contrast, the magnitudes of the cross sections for DI relative to DE strongly depend on the projectile speed and the target. The relative importance of DI increases with increasing projectile speed, which can easily be explained by the higher-energy threshold for DI. For He DI is much less important than for Ar and is, in fact, negligible compared to DE. This is not surprising either because the binding energy of He is nearly a factor of 2 larger than the one of Ar. In contrast, an observation which was not necessarily expected is that for a N_2 target DI is also much less important compared to DE than for Ar, although the ionization potential is nearly the same for both targets. This strongly suggests that other aspects of the target structure play an important role as well.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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