

Excitation function of detachment with simultaneous target ionization in slow collisions of D^- with He and Ar

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We have measured momentum analyzed electrons and singly charged recoil ions ejected in slow collisions of D^- ions with He and Ar in triple coincidence with neutralized projectiles. From the data, we extracted relative triple differential cross sections for detachment and detachment with simultaneous target ionization as a function of projectile energy. The results were compared with our perturbative calculations. At large projectile energies, we observed signatures of a nonsequential process, in which the electrons are ejected from the projectile and the target by a single mutual interaction. We find that contributions from sequential processes are important as well and, at small energies, even dominant.

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I. INTRODUCTION

The reaction dynamics underlying collisions between slow ions and atoms play a key role in understanding transitions in nature on a fundamental level. Of particular interest is a problem known as the few-body problem (FBP), which is relevant in almost all areas of modern physics research. The essence of the FBP is that the Schrödinger equation is not analytically solvable for more than two mutually interacting particles, even when the underlying forces are accurately known (e.g. [1,2]). The FBP has been studied extensively for basic one-electron transitions, such as target ionization, occurring in atomic collisions ([3,4] and references therein). A specific aspect of the FBP arises in processes where one electron is ejected from each collision partner. Here, the transition of two electrons can occur simultaneously under the influence of a single interaction, which is the one between the two active electrons (e.g. [5–7]), i.e., in a first-order process. This is a manifestation of electron-electron correlation occurring between the two centers of the collision.

In the case of anion-atom collisions, the detachment from the projectile with simultaneous target ionization [Detachment from the projectile with simultaneous target ionisation (DI)] can be analyzed within the quasi-free-electron model (QEM) [8]. Because the detachment energy of anions is typically small (around 1 eV), in this model the binding of the electron to the projectile is initially neglected. The DI process is then viewed as target ionization by free-electron

impact, also known as an (e,2e) reaction (for the e,2e process, see, e.g., [9,10]). For an (e,2e) process, a threshold exists because the kinetic energy of the projectile electron must be larger than the target ionization potential. For the correlated first-order DI channel (C1 channel), this threshold energy is scaled up by the anion to electron mass ratio. Below the threshold, the C1 process can only occur due to the momentum that the projectile electron already possesses due to its initial binding to the anion. The convolution of the (e,2e) cross section with the momentum distribution in the initial bound state leads to a “washing out” of the threshold. A competing process to the C1 channel is a second-order uncorrelated (U2) channel. Here, each electron is ejected by an interaction with the core of the other respective collision partner. The threshold energy for this process is several orders of magnitude smaller than for the C1 channel and is irrelevant for the projectile energies usually encountered in accelerator experiments.

We recently reported measured two-dimensional momentum spectra of electrons ejected in DI for 300 keV $Si^- + Ar$ collisions [8]. We expected the C1 and U2 channels to exhibit different signatures in these momentum distributions. In the experiment, we could not directly distinguish between electrons ejected from the target or the projectile. Therefore, the U2 contributions to the electron spectra should exhibit a combination of the features which would be observed if each process (i.e., detachment and target ionization) was studied separately. Within the QEM, detachment alone is viewed as elastic scattering of the projectile electron from the target. It thus leads to a sharp magnitude of the electron momentum vector corresponding to the projectile speed v_p (i.e., a ring with radius v_p in the two-dimensional momentum spectrum) reflecting conservation of kinetic energy, which is characteristic of elastic scattering. A similar structure as seen for detachment should be present in the momentum spectrum for the U2 DI process.

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For the C1 process, qualitatively different momentum distributions are expected, depending on whether the projectile energy is below or above the threshold. Below the threshold, the most likely scenario is that the total velocity (sum of the electron velocity in the rest frame of the anion and the projectile velocity) would barely be large enough to overcome the target ionization potential because the Compton profile decreases rapidly with increasing momentum. We thus expected a pronounced maximum near an electron momentum of zero. Above the threshold, the maximum should gradually shift to larger momenta corresponding to the excess energy of the projectile electron in the target frame.

Regarding pure detachment, in [11] we indeed did observe a ring structure with radius v_p in the two-dimensional electron spectra for 300 keV $\text{Si}^- + \text{Ar}$ collisions, in accordance with the prediction of the QEM. However, in the corresponding spectra for DI, this ring structure was completely absent. Instead, only a sharp maximum was observed at zero momentum. This suggested that the U2 channel is insignificant compared to the C1 process. This would be quite surprising considering that the electron energy corresponding to v_p (5.8 eV) was well below the threshold of 15.5 eV (the sum of electron detachment energy of the anion and the ionization potential of the target). Clear signatures of the C1 process were observed previously for $\text{H}^- + \text{He}$ collisions [5]; however, these studies were performed for an anion energy of 200 keV, which was above the threshold energy (scaled up by the projectile mass) of 46 keV. This channel was also found in energetic collisions (far above the threshold) of cations with He (e.g. [6,7]) leading to electron ejection from both collision partners.

Here, we report triple differential momentum distributions of electrons ejected in collisions of $\text{D}^- + \text{He}$ by the DI process at projectile energies ranging from well below to above the threshold for the C1 process. The motivation was twofold: First, we wanted to investigate a collision system which was more amenable to theoretical calculations than $\text{Si}^- + \text{Ar}$ with its 33 electrons (13 of which are valence electrons). Second, we wanted to study the excitation function for DI, i.e., the projectile energy dependence of the cross sections. The excitation function should provide a clear signature of the C1 process as it should rapidly rise near the threshold (although the threshold will be “blurred” by the Compton profile). Indeed, at large projectile energies, a strong role played by the C1 process is confirmed by the present data and its comparison to our calculations. In contrast, at very small projectile energies, the U2 channel becomes dominant, at least for the He target. Apart from a sharp maximum at zero momentum, the theoretical momentum spectra also reveal surprising signatures of the C1 channel. Atomic units ($m_e = \hbar = e = 1$, where m_e is the electron mass, $\hbar$ is the reduced Planck constant, and e is the elementary charge) are used throughout, except where otherwise stated.

II. EXPERIMENTAL SETUP

The experiment was performed with the Reaction Microscope (ReMi) at the Cryogenic Storage Ring (CSR) [12] at the Max Planck Institute for Nuclear Physics in Heidelberg (CSR-ReMi). A schematic sketch of the setup is shown in

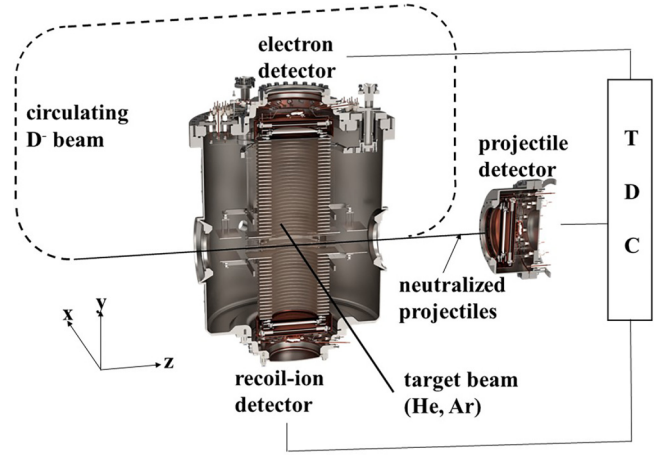


FIG. 1. Schematic sketch of the experimental setup.

Fig. 1. A D^- beam was generated with a Cs sputter source and the ions were accelerated to energies ranging from 20 to 120 keV using a high-voltage platform. The beam was then injected into the CSR and kept in the ring by electrostatic deflectors for 10–20 seconds (depending on beam energy) before a new particle bunch was injected. This relatively frequent periodic reloading of the ring was necessary because the storage ring ion optical lattice is more difficult to optimize for long storage times when using light ions at comparatively low kinetic energies. As we were changing beam energies frequently during the present experiment, we did not focus on achieving particularly long storage times. The projectiles were crossed with a very cold ($T < 1$ K) helium beam from a supersonic gas jet. The projectiles which were neutralized in the collision were separated from the stored ions by the electrostatic deflector located after the target region and detected by a two-dimensional position-sensitive microchannel-plate (MCP) detector equipped with a delay-line anode [13]. Due to the excellent vacuum conditions in the CSR, the background of the detected projectiles neutralized in the residual gas was negligible.

Using the CSR-ReMi apparatus, electrons and recoiling target ions produced in the collision were extracted in opposite directions by a weak electric field of about 1.2 V/cm directed perpendicular to the anion beam direction [14,15]. They were detected by MCP detectors essentially identical to the neutralized projectile detector. In order to optimize the uniformity of the extraction field, two grids with a transmission of 73% each were placed in front of each detector. Along with an open area ratio of the channel plates of 0.65, this amounts to a single-event detection efficiency of about 0.35 for the individual detectors. The electric extraction field was superimposed by a magnetic field of 5 G parallel to the electric field. It forced the electrons into cyclotron motion with a radius proportional to the transverse (relative to the magnetic field) electron momentum. For momenta smaller than about 1.5 a.u., the radii were smaller than the detector size (120 mm in diameter), i.e., these electrons were detected with essentially 100% solid-angle acceptance. Due to their much larger mass, the recoil ions were only slightly deflected by the

magnetic field, but did not complete a significant fraction of a cyclotron cycle.

All three detectors were set up to allow triple coincidence detection of neutral, recoil ion, and electron hits. The time of flight of the electrons and the recoil ions relative to the neutralized projectiles, which are contained in the respective coincidence time, provide the corresponding momentum components in the direction of the extraction field (y direction). The two momentum components in the plane perpendicular to the extraction field are contained in the position information of the respective detector. The x direction is defined by the target beam and the z direction by the projectile beam direction.

III. THEORETICAL CONSIDERATION

Both D^- ions and helium atoms consist of a nucleus and two electrons and collisions of D^- projectiles with helium targets thus represent a dynamical six-body problem, a rigorous solution of which is at present well beyond reach.

To simplify the problem, we first note that the electron affinity (≈ 0.75 eV) in D^- is more than an order of magnitude smaller than the first excitation energy (≈ 10 eV) in D^0 . Furthermore, the first ionization potential of He (≈ 24.6 eV) is significantly smaller than the first excitation energy (≈ 40 eV) in He^+ . Therefore, to a good approximation in DI, only one electron on each of the colliding particles can be regarded as “active,” whereas the other one plays a “passive” role. As a result, the collision problem can effectively be reduced to a four-body problem involving two active electrons and two heavy cores consisting of a nucleus and a passive electron.

In fast collisions, where the impact velocity v_p is much larger than the typical orbiting velocities of the projectile and target electrons, the DI process proceeds mainly via a single interaction between two electrons belonging to the different colliding centers (the so-called two-center dielectronic interaction [16]). This channel appears already in the first order of perturbation theory in the projectile-target interaction.

When the collision velocity decreases, another channel of DI becomes important. Here, DI proceeds via two simultaneous interactions: the target electron is emitted by an interaction with the projectile core, whereas the projectile electron is knocked out by the interaction with the target core. This reaction channel appears in a theoretical description starting with the second-order perturbation expansion in the projectile-target interaction.

Our theoretical treatment of DI in $D^- + He$ collisions is based on the semiclassical approach in which the heavy particles—the D^- ion core and the helium atomic core—are treated classically, while the active electrons are described quantum mechanically. It is convenient to choose a reference frame in which the target nucleus is at rest and taken as the origin. In this frame, the projectile ion is assumed to move along a straight-line classical trajectory given by $\mathbf{R}(t) = \mathbf{b} + \mathbf{v}_p t$, where $\mathbf{b} = (b_x, b_y, 0)$ denotes the impact parameter, $\mathbf{v}_p = (0, 0, v_p)$ is the collision velocity, and t is time.

The undistorted bound-state wave function $\psi_{He}(\mathbf{r})$ and the continuum wave function $\psi_{k_c}(\mathbf{r})$ of the target electron are obtained as numerical solutions of the Schrödinger equation for a single active electron moving in the effective potential

$V(\mathbf{r}) = -\frac{1}{r} - (\frac{1}{r} + a)e^{-ar}$ of the He^+ core, where $\alpha = 3.36$ and $a = 1.665$ (see [17]).

In order to obtain the initial and final undistorted states of the active electron in D^- , following [18–20], we shall use a nonlocal separable potential of Yamaguchi [21] to model a short-range effective interaction of the active electron with the core of D^- ,

$$V(r, r') = -\lambda g(r)g(r'), \quad (1)$$

where

$$g(r) = \sqrt{\frac{\beta}{2\pi}} \frac{\exp(-\beta r)}{r}, \quad (2)$$

with $l = 0.659$ and $b = 0.913$.

The potential (1) and (2) leads to the following expressions for the initial (bound) and final (continuum) states of the active electron in D^- :

$$\varphi_D(\mathbf{s}) = N \frac{e^{-\kappa s} - e^{-\beta s}}{s} \quad (3)$$

and

$$\varphi_p(\mathbf{s}) = \frac{1}{(2\pi)^{3/2}} \left(e^{i\mathbf{p} \cdot \mathbf{s}} + A \frac{e^{-i\mathbf{p} \cdot \mathbf{s}} - e^{-i\beta s}}{s} \right). \quad (4)$$

Here, $\mathbf{s}$ is the coordinate of the active electron with respect to the deuterium nucleus ($s = |\mathbf{s}|$), $\kappa = \sqrt{2\lambda} - \beta \approx 0.235$, $\varepsilon_b = -\kappa^2/2 \approx 0.75$ eV is the electron affinity, the normalization factor $N = \sqrt{\kappa\beta(\kappa + \beta)/2\pi(\beta - \kappa)^2}$, $\mathbf{p}$ is the momentum of the electron emitted from D^- , and $A = \frac{4\beta\lambda}{(p^2 + \beta^2)^2 + 2\lambda(p + i\beta)^2}$.

A. Correlated mechanism

The first-order transition amplitude, which describes the contribution to the process for the simultaneous neutralization of the projectile and ionization of the helium target driven by the two-center dielectronic interaction W^{ee} , is given by

$$a_{fi}^{ee}(b) = -i \int_{-\infty}^{+\infty} dt \langle \Psi_f(t) | W^{ee}(t) | \Psi_i(t) \rangle. \quad (5)$$

Here, $\Psi_i(\Psi_f)$ are the initial (final) undistorted states of the combined projectile-target system.

The initial state Ψ_i is constructed as a product of the projectile electron bound state and the helium ground state and is given by

$$\Psi_i(t) = \varphi_D(\mathbf{s}) e^{i\mathbf{v}_p \cdot \boldsymbol{\rho}} e^{-i\left(\varepsilon_i + \frac{v_p^2}{2}\right)t} \psi_{He}(\mathbf{r}) e^{-i\varepsilon_i t}, \quad (6)$$

where $\mathbf{s}$ is the coordinate of the projectile electron relative to the projectile ion core, and $\boldsymbol{\rho} = \mathbf{s} + \mathbf{R}(t)$ is its coordinate relative to the target nucleus. Here, ε_i and φ_D are the binding energy and the ground-state wave function, respectively, of the weakly bound electron of the negative deuterium ion. Furthermore, $\mathbf{r}$ denotes the coordinate of the active target electron, while ε_i and ψ_{He} are its binding energy and its single-electron ground-state wave function in the field of the He^+ core. The translational motion of the projectile electron is accounted for by the phase factors $e^{i\mathbf{v}_p \cdot \boldsymbol{\rho}}$ and $e^{-i\mathbf{v}_p^2 t/2}$, which ensure the correct Galilean invariance of the wave function. Similarly, the electronic final state Ψ_f , describing the projectile electron

in the short-range field of the neutral deuterium atom in its ground state and the ejected target electron in the field of the residual helium ion, reads

$$\Psi_f(t) = \varphi_p(\mathbf{s}) e^{i\mathbf{v}_p \cdot \mathbf{p}} e^{-i(\varepsilon_p + \frac{v^2}{2})t} \psi_{\mathbf{k}_e}(\mathbf{r}) e^{-i\varepsilon_k t}, \quad (7)$$

where $\varphi_p(\mathbf{s})$ describes the continuum projectile electron in the field of the neutral D atom, with energy ε_p and asymptotic momentum $\mathbf{p}$ in the projectile frame. $\psi_{\mathbf{k}_e}(\mathbf{r})$ is the continuum wave function of the ejected target electron with energy ε_k and asymptotic momentum $\mathbf{k}_e$ in the target frame.

The corresponding amplitude in momentum space is obtained according to

$$S_{fi}^{ee}(\mathbf{Q}) = \frac{1}{2\pi} \int db \exp(i\mathbf{Q} \cdot \mathbf{b}) a_{fi}^{ee}(\mathbf{b}), \quad (8)$$

where the integration is carried out over the two-dimensional impact parameter plane and $\mathbf{Q}$ represents the transverse momentum transferred to the target nucleus in the collision. After a simple calculation, Eq. (8) can be expressed as

$$S_{fi}^{ee}(\mathbf{Q}) = -2 \frac{i}{v_p q^2} \langle \psi_{\mathbf{k}_e} | e^{i\mathbf{q} \cdot \mathbf{r}} | \psi_{\text{He}} \rangle \langle \varphi_p | e^{-i\mathbf{q} \cdot \mathbf{s}} | \varphi_D \rangle, \quad (9)$$

where $\mathbf{q} = (\mathbf{Q}, \frac{\varepsilon_p + \varepsilon_k - \varepsilon_i - \varepsilon_i}{v_p})$ is the momentum transferred to the target in the collision.

B. Uncorrelated mechanism

To describe the higher-order process, we assume that the two electrons are removed independently. The corresponding transition amplitude is therefore given by a product of two independent single-electron amplitudes, corresponding to the interaction of each active electron with the core of the respective other collision partner. In the momentum-space amplitude of Eq. (8), we replace the correlated amplitude $a_{fi}^{ee}(\mathbf{b})$ with the product $a_{fi}^{\text{det}}(\mathbf{b}) a_{fi}^{\text{ion}}(\mathbf{b})$, where the first factor describes the detachment of the projectile electron induced by the target core and the second factor describes the ionization of the target electron induced by the projectile core. Thus, after some straightforward algebra, the sequential transition amplitude reads

$$S_{fi}^{ne}(\mathbf{Q}) = \frac{1}{2\pi} \int d\eta S_{fi}^{\text{ion}}(\eta) S_{fi}^{\text{det}}(\mathbf{Q} - \eta), \quad (10)$$

where S_{fi}^{det} describes the transition amplitude for the detachment of the projectile electron due to its interaction with the target and is given by

$$S_{fi}^{\text{det}}(\mathbf{Q} - \eta) = 2 \frac{i}{v_p} \langle \varphi_p | \frac{e^{i\mathbf{q}_1 \cdot \mathbf{s}}}{q_1^2} | \varphi_D \rangle,$$

with $\mathbf{q}_1 = [\eta - \mathbf{Q}, (\varepsilon_i - \varepsilon_p)/v_p]$. Furthermore, S_{fi}^{ion} describes the transition amplitude for the ionization of the target electron due to its interaction with the screened short-range field of the neutral deuterium and reads

$$S_{fi}^{\text{ion}}(\eta) = \frac{2i}{v} \sum_{i=1}^3 \frac{A_i}{q_2^2 + \alpha_i^2} \langle \psi_{\mathbf{k}_e} | e^{i\mathbf{q}_2 \cdot \mathbf{r}} | \psi_{\text{He}} \rangle,$$

where $\mathbf{q}_2 = [\eta, (\varepsilon_k - \varepsilon_i)/v_p]$ is the momentum transfer to the target, and A_i and α_i are the screening parameters of the Salvat potential [22]. Note that the integration in Eq. (10) is carried

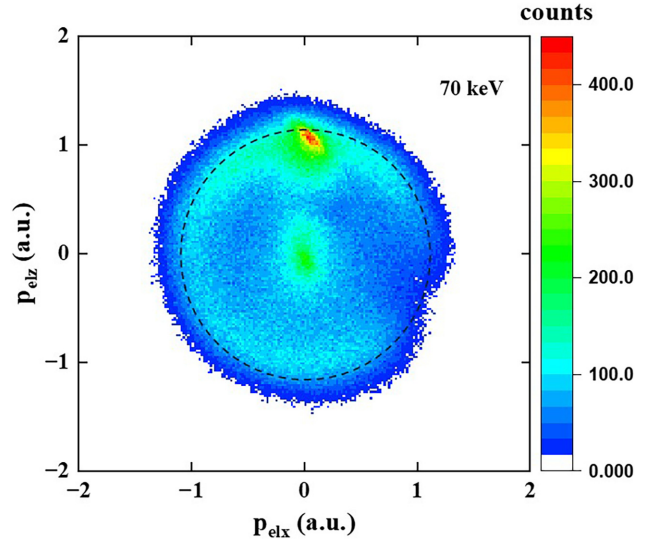


FIG. 2. Triple differential momentum distribution of electrons ejected in 70 keV $D^- + \text{He}$ collisions and measured in coincidence with the neutralized projectiles as a function of the x and z components. The z component is in the direction of the projectile beam. The y component was restricted to $|\mathbf{p}_{\text{ely}}| < 0.05$ a.u.

out over the two-dimensional momentum η . The differential cross section for the emission of the two electrons is given by the integral over the transverse momentum transfer $\mathbf{Q}$ of the squared modulus of the sum of the correlated and sequential amplitudes and reads

$$\frac{d^6\sigma_{fi}}{d^3\mathbf{p} d^3\mathbf{k}_e} = \int d\mathbf{Q} |S_{fi}^{ee}(\mathbf{Q}) + S_{fi}^{ne}(\mathbf{Q})|^2, \quad (11)$$

where $\mathbf{p}$ is the momentum of the ejected projectile electron in the projectile frame, $\mathbf{k}_e$ is the momentum of the ionized target electron given in the target frame, and the coherent sum of the two amplitudes accounts for the interference between the correlated and uncorrelated channels. The differential cross section $\frac{d^3\sigma_{fi}}{d^3\mathbf{p}_{\text{el}}}$ for detecting any electron with a given momentum p_{el} (in the laboratory frame) is obtained by standard symmetrization of the two-electron amplitude and integration over the momentum of the other electron.

IV. RESULTS AND DISCUSSION

A. $D^- + \text{He}$

In Fig. 2, we present a two-dimensional momentum distribution of electrons ejected in 70 keV $D^- + \text{He}$ collisions and measured in coincidence with the neutralized projectiles. Furthermore, we set an anticondition on the recoil ions, i.e., we demanded that no valid recoil ion was detected. The vertical axis represents the z component (projectile beam direction) and the horizontal axis the x component (one of the transverse components). The y component, i.e., the second transverse component, was kept fixed at 0 ± 0.05 a.u. The momentum distribution is thus proportional to $d^3\sigma/dp_{\text{el}}^3$. The coincidence condition with the projectiles combined with the anticondition on the recoil ions favors pure detachment without ionizing the target.

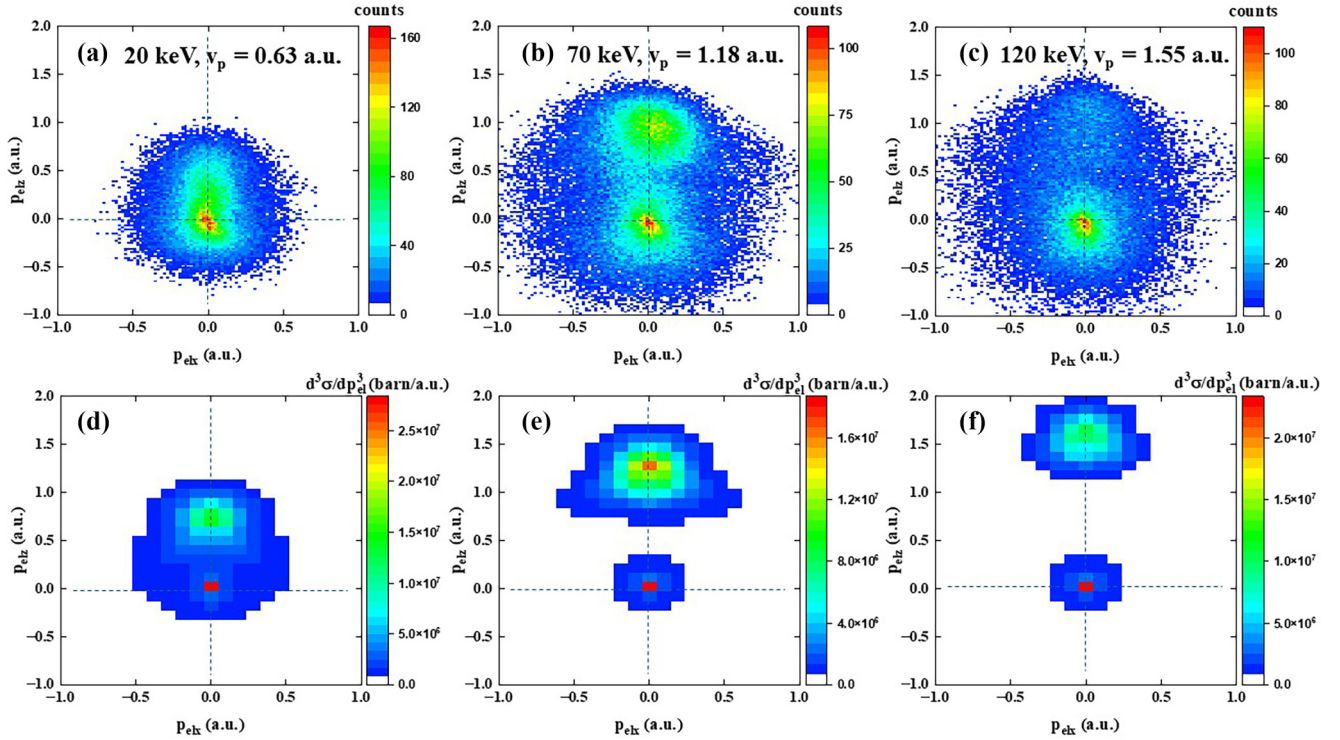


FIG. 3. Triple-differential momentum distribution of electrons ejected in (a) 20 keV, (b) 70 keV, and (c) 120 keV $D^- + \text{He}$ collisions and measured in coincidence with the neutralized projectiles and singly charged recoil ions as a function of the x and z components. Furthermore, an anticondition was applied to the recoil-neutralized projectile coincidence (see text). The y component was restricted to $|p_{ely}| < 0.05$ a.u. (d)–(f) The same spectra and at the same energies calculated with our second-order model.

This double-coincident electron momentum distribution is qualitatively very similar to what we observed for 300 keV $\text{Si}^- + \text{Ar}$ collisions [8], in that two structures are quite prominent in Fig. 2: first, a ringlike shape with a radius approximately equal to v_p , where along this ring a strong maximum is observed in the forward direction; second, a pronounced maximum is seen at zero momentum. The ring structure can be understood within the QEM [8,11,23], which neglects the initial binding of the projectile electron because the detachment energy of D^- is small (about 0.75 eV). With this approximation, the detachment process is viewed as elastic scattering of a free electron off the target. The ring shape then simply reflects conservation of kinetic energy, which is characteristic to elastic scattering [11].

The sharp maximum on the ring in the forward direction is reminiscent of so-called cusp electrons that are well known in collisions of cations with neutral atoms (e.g. [24]). It is usually associated with the long-range nature of the projectile potential. In the present case, the projectile gets neutralized in the detachment process, resulting in a short-range projectile potential. However, if detachment is accompanied by excitation of the neutralized projectile to the $n = 2$ level, the well-known accidental degeneracy between the $2s$ and $2p$ substates leads to a permanent electric dipole moment [25], which does not occur for many-electron atoms (e.g. [26]). The range of the corresponding potential of the D° atom is longer than without the mixing. Therefore, the electron is emitted from an effective two-center potential, which can result in the characteristic cusp-shaped peak in the electron momentum

spectrum or energy spectrum. Apart from cusp electrons, a shape resonance, due to double excitation to the $(2s2p)^1P$ state, can also contribute to the sharp peak in the forward direction [27].

Several processes contribute to the maximum at zero momentum. Probably the most important component comes from DI. Although this process is suppressed by the anticondition on the recoil ions, it is not completely eliminated from the electron momentum distribution of Fig. 2 because of the limited recoil-ion detection efficiency of about 35%. As we will show in the following paragraph and in Fig. 3, DI yields a strong maximum at zero momentum. Another contributor to this momentum is detachment with simultaneous target excitation (DE) [28].

In Fig. 3(b), we present the same type of electron momentum distribution as shown in Fig. 2, except a triple-coincidence condition on the electrons, singly charged recoil ions, and neutralized projectiles were applied. These triple coincidences select DI events. It should be noted that here the momentum spectra are only shown between -1 and 1 a.u. (horizontal axis) and -1 and 2 a.u. (vertical axis), while in Fig. 2 they are plotted between -2 and 2 a.u. for both axes. The momentum distribution for DI is actually significantly narrower than for pure detachment. In contrast to the double-coincident spectra, the triple-coincident data reveal a significant qualitative difference to those we reported for 300 keV $\text{Si}^- + \text{Ar}$ collisions: while in the latter the only structure we observed was a sharp peak at zero momentum [8], in the present data we find a second strong, but significantly

broader, maximum at an electron momentum corresponding to a speed somewhat below v_p . As outlined in Sec. I, we previously assumed that below the threshold, the signature of the C1 process is a single sharp maximum at zero momentum. The U2 process should lead to two components: a ring with radius corresponding to v_p , which reflects the ejected projectile electrons, while the target electrons should contribute mostly to $p_{el} = 0$. Although instead of a complete ring we only observe a maximum near v_p , this maximum is consistent with the U2 process as the electron momentum has the same magnitude as in pure detachment. The fact that we do not observe a complete ring merely means that the angular distribution of electrons ejected in DI is different from those ejected in detachment. Nevertheless, the process signified by the maximum near v_p is likely closely related to the one which, until now, we referred to as the U2 process. However, this discussion shows that this notation may not be completely justified: if the angular distribution of the electron detached from the projectile does depend on whether or not an electron is ejected from the target, then some correlation between the two electrons is present. For the remainder of this article, we therefore refer to the uncorrelated process as a sequential process and, correspondingly, to the correlated process as a nonsequential process.

In Figs. 3(a) and 3(c), we present the same momentum spectra as in the center panel, except for collision energies of 20 and 120 keV, respectively. Some similarity to the data for 70 keV is visible in so far as, apart from a sharp maximum at zero momentum, non-negligible contributions at $p_{el} \approx v_p$ are also present. However, the intensity ratio between the structures at $p_{el} \approx v_p$ and $p_{el} = 0$ is much larger at 70 keV. At first glance, this seems to suggest that the nonsequential process minimizes around this energy. In the following, we will analyze the relative excitation function for DI in order to examine this hypothesis in more detail as well as our conjecture expressed in [8] that at least for an Ar target, even well below the threshold, the nonsequential channel is dominant over the sequential channel.

If the nonsequential process was indeed dominant, we should observe the cross section for DI to step up relatively sharply at the threshold energy (scaled up by the projectile to electron mass ratio) of about 90 keV. This step should be “washed out” due to the projectile Compton profile. We can estimate the width of the step from the initial kinetic energy of the projectile electron in the target frame, which is given by $K_{el} = 1/2 M_p (\mathbf{v}_p + \mathbf{v}_i)^2$, where v_p is taken at the free-electron threshold and v_i is the momentum in the Compton profile. The width is determined by the cross term $M_p \mathbf{v}_p \mathbf{v}_i$, which is about 35 keV.

In Fig. 4, we present the projectile-energy dependence of the ratio R between the content of the peak at $p_{el} = 0$ and the content of the ring containing the maximum at $p_{el} \approx v_p$ for DI, which we refer to as the relative excitation function. The peak at $p_{el} = 0$ was integrated up to a radius r at which the minimum separating the two peak structures (except for 20 keV) occurs, which increases with E_p . This radius for the position of the minimum as a function of E_p is well described by the expression $0.26 + 2.6 \times 10^{-3} \times E_p$ (keV). The ring was integrated from r to $p_{el} = 1.5$ a.u. since beyond this value we did not detect any electrons. It should be noted that the statistical

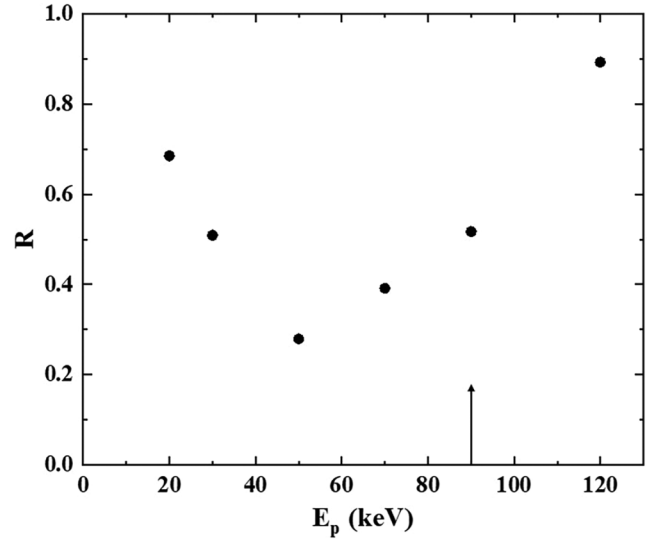


FIG. 4. Projectile-energy dependence of the ratio R between the content of the peak at $p_{el} = 0$ and the content of the ring containing the maximum at $p_{el} \approx v_p$ for DI seen in the momentum distributions of Fig. 3. The arrow indicates the threshold energy for the nonsequential channel.

error bars are smaller than the size of the data points. For 20 keV, no clear minimum can be seen and R is therefore afflicted with a larger uncertainty. The arrow in Fig. 4 indicates the threshold energy for the nonsequential channel. The region of $p_{el} = 0$ does not exclusively select the nonsequential process because the sequential channel also produces, apart from projectile electrons with $p_{el} \approx v_p$, target electrons preferentially with very small momenta. However, assuming that the region of the ring strongly favors the sequential mechanism, R can still serve as a qualitative measure of the relative importance of the nonsequential channel. Indeed, at large E_p , R exhibits a step occurring at approximately the expected threshold energy and with the estimated width.

Before we conclude that the rise of R near 90 keV reflects the threshold for the nonsequential process, we should discuss one other possible explanation. Just like pure detachment, DI can also be accompanied by projectile excitation of the residual D^+ projectile. The strength of the mixing between the $2s$ and $2p$ states, which increases the range of the projectile potential and thereby the electron flux near $p_{el} \approx v_p$, is expected to be projectile-energy dependent. It could thus affect the shape of R as a function of E_p and possibly lead to a rise of R at large E_p . However, for the following reasons, we believe that the nonsequential channel is more important than this projectile-excitation channel: first, the latter represents at least a third-order process, while the nonsequential mechanism is first order. In addition, the excitation of the neutralized projectile requires an energy of 10 eV, which is more than an order of magnitude larger than the detachment energy of D^- . Therefore, although this third-order process should exist, it is likely significantly weaker than the second-order channel without excitation, at least at large E_p . Furthermore, the maximum near $p_{el} = v_p$ observed for DI is much broader than the one seen in the pure detachment process. Thus, it does not match the cusplike shape expected for electrons produced

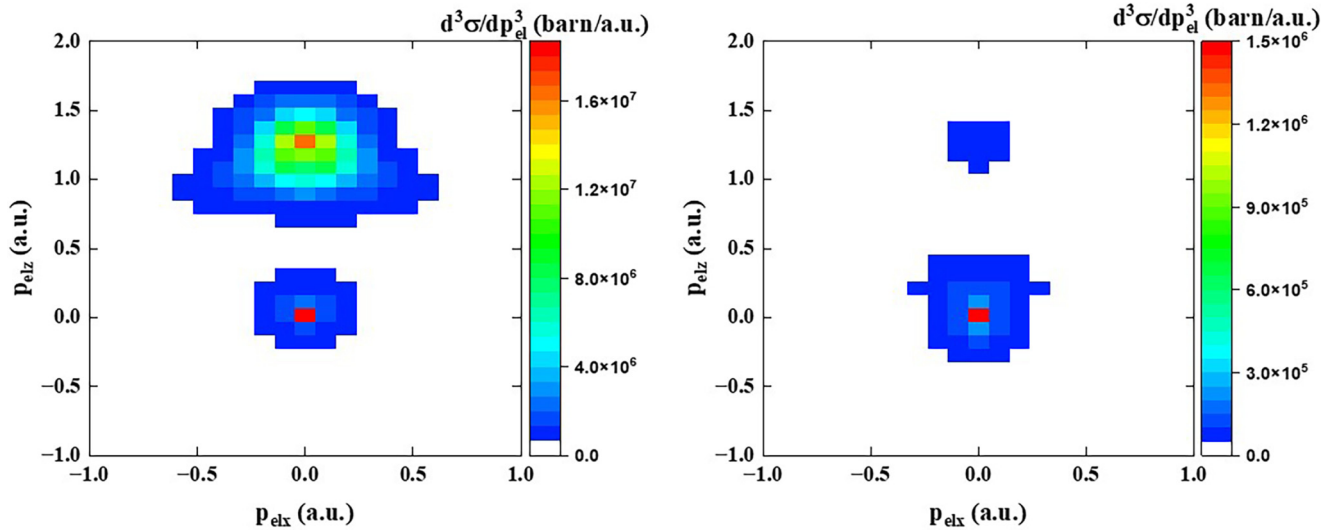


FIG. 5. Left panel: Same as Fig. 3(e). Right panel: Same as Fig. 3(e), but calculated with the first-Born approximation, i.e., only the nonsequential channel is accounted for.

by the dipole moment resulting from projectile excitation. We also note that the rise of R at large E_p matches the expectation for the threshold of the nonsequential channel both in position and width. In contrast, for the process involving projectile excitation, the expected energy dependence of R is not clear and a match with the threshold behavior would be coincidental. Finally, our theoretical model does not account for DI with simultaneous projectile excitation to degenerate states, but it does reproduce the rise of R at large E_p (see Fig. 6).

A surprising observation is that after a pronounced minimum at about 50 keV, R rises again with decreasing E_p . It is difficult to understand why the importance of the nonsequential relative to the sequential channel would increase well below the threshold. The probability to find a sufficiently large momentum in the Compton profile to overcome the ionization potential diminishes rapidly with decreasing projectile energy. Of course, the target electrons ejected in the sequential channel leading to projectile electrons with $p_{el} \approx v_p$ mainly contribute (like the nonsequential process) to $p_{el} = 0$. However, in the absence of the nonsequential process, it would lead to a flat behavior of R because, for each target electron ejected in this channel, there should be one projectile electron with high probability contributing to the structure near v_p . The rise of R below the minimum could possibly be explained by the DI plus projectile excitation channel because higher-order processes tend to gain in importance relative to lower-order mechanisms with decreasing E_p .

Before we return to the discussion of the energy dependence of R , in Figs. 3(d) and 3(f), we present the calculated two-dimensional electron momentum distributions for the same conditions as for the experimental data shown in Figs. 3(a) to 3(c). Considering the complexity of the problem, there is encouraging qualitative agreement between experiment and theory. Like the data, the calculations also exhibit two peak structures, one at $p_{el} = 0$ and one near $p_{elz} = v_p$. Furthermore, the calculation correctly predicts that the intensity of the maximum near $p_{elz} = v_p$ relative to the one at zero is larger at $E_p = 70$ keV than both at 20 and 120 keV.

One important question to address is to what extent theory confirms that the nonsequential channel is characterized by a sharp maximum at $p_{el} = 0$ and the sequential channel by a ring with radius close to v_p or at least a maximum near v_p . To this end, we calculated the momentum spectra within the first-Born approximation (FBA). Here, only the mutual interaction between the two active electrons can lead to a DI process, i.e., the sequential channel is completely excluded. This calculation is shown in the right panel of Fig. 5 for 70 keV in comparison with the full calculation (left panel). First, we note that the full calculation exhibits cross sections which are about an order of magnitude larger than those predicted by the FBA. Therefore, the dominance of the nonsequential channel, which appeared to be present for 300 keV $\text{Si}^- + \text{Ar}$ collisions even below the threshold, is not predicted by our theory for the He target. Nevertheless, this process is not negligible.

A surprising feature of the FBA calculation is that apart from the expected sharp maximum at $p_{el} = 0$, it also predicts a maximum at v_p . However, it is more than an order of magnitude weaker than in the full calculation. Theory thus confirms that the contributions at v_p are mostly a signature of the sequential channel. Nevertheless, based on the argument that below the threshold the nonsequential process should force both ejected electrons to give up their entire kinetic energy in the target frame, we did not anticipate any maximum at all at v_p . Its presence can be explained as follows: two factors which determine the cross section for the nonsequential process need to be considered. First, this channel needs to pick a minimum momentum from the projectile Compton profile in order to bring the projectile electron energy above the threshold. Beyond this minimum momentum, this factor decreases strongly because the Compton profile drops rapidly with increasing momentum. This is basically the argument which made us anticipate a single sharp maximum at $p_{el} = 0$. However, a second factor is that a relatively large force is required to decelerate the projectile electron to a zero momentum. Since a final momentum at v_p does not require any force, this factor

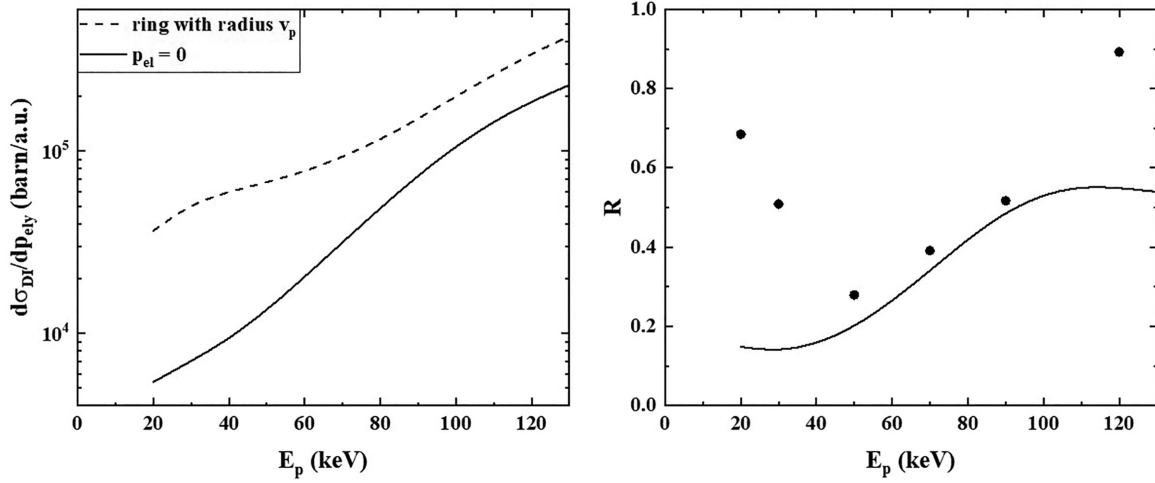


FIG. 6. Left panel: Calculated content of the ring with radius v_p (dashed curve) and of the maximum at $p_{el} = 0$ (solid curve) as a function of projectile energy. Right panel: Ratio between the calculations represented by the solid and dashed curves in the left panel in comparison with the experimental data of Fig. 4.

would favor electrons with such a final momentum. However, this is only possible if the nonsequential process selects a momentum from the Compton profile which is even much larger than what would be required by the need to overcome the threshold. The combination of both factors leads to the double-peak structure observed in the FBA.

The solid curve in the right panel of Fig. 6 shows R calculated with the full theory in comparison to the experimental data as a function of projectile energy. There is satisfactory agreement between experiment and theory for $E_p > 40$ keV, where theory reproduces the expected threshold behavior. However, the calculation only yields a very shallow minimum separating the two peak structures, which furthermore is shifted to smaller E_p compared to the measured data. In addition, in the calculation, the magnitude of R is smaller by about a factor of 5 at small E_p than the experimental values. On the other hand, our perturbative model is not well suited for very small E_p and larger discrepancies are thus not surprising. Furthermore, one should recall that the calculation does not account for DI with projectile excitation, which could make a significant difference at small E_p . The left panel of Fig. 6 shows the calculated content of the ring with radius v_p (dashed curve) and of the zero-momentum peak (solid curve) separately. Here, for both regions, the calculation increases

monotonically with increasing E_p . For the zero-momentum region, the shape is consistent with the threshold behavior expected for the nonsequential channel. In contrast, for the $p_{el} = v_p$ region, the excitation function exhibits a “shoulder” structure, which is not seen for $p_{el} = 0$, and, overall, increases significantly slower. Here, the increasing excitation function is not primarily due to a threshold behavior, but rather reflects the general tendency of cross sections for inelastic processes to increase with E_p before reaching a maximum at E_{max} . For the sequential DI channel, E_{max} is larger than the highest energy for which data were taken.

The calculated cross sections for the regions $p_{el} = 0$ and $p_{el} = v_p$ suggest that the minimum in the energy dependence of R is not necessarily due to a minimum for the cross section for the $p_{el} = 0$ region nor due to a maximum for those for the $p_{el} = v_p$ region. Rather, the increase of the theoretical R at small E_p could merely be a reflection of the flatter behavior of the curve for the ring compared to the one for $p_{el} = 0$ in the region between approximately 30 and 60 keV.

B. $D^- + Ar$

The discussion of the preceding section shows that the non-sequential process is an important contributor to DI, at least at

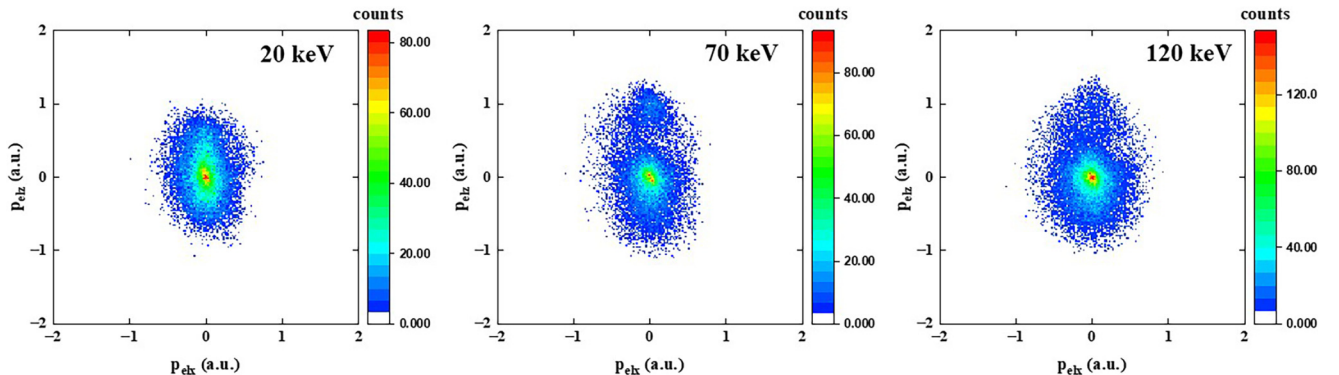


FIG. 7. Same as Fig. 3, but for $D^- + Ar$ collisions.

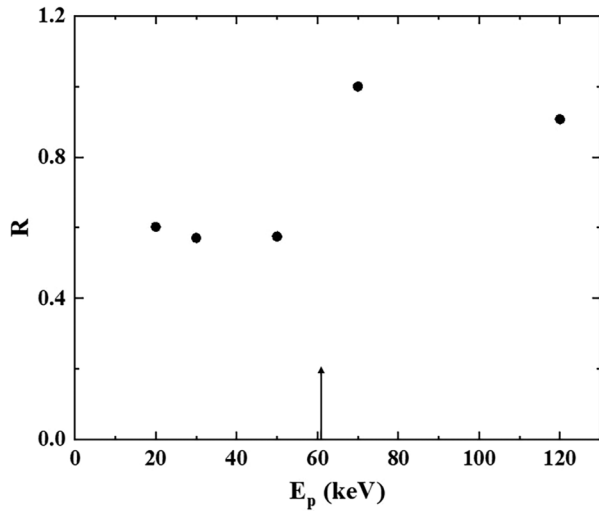


FIG. 8. Same as the right panel of Fig. 4, but for $D^- + Ar$ collisions.

the larger energies studied here. However, it also suggests that the conclusion of [8], i.e., that the nonsequential channel is dominant over the sequential channel in 300 keV $Si^- + Ar$ collisions, even far below the threshold, does not hold to the same extent for a helium target. According to theory, for 20 keV $D^- + He$ (and even for 70 keV), which corresponds to nearly the same v_p as 300 keV Si^- , the sequential process is dominant. In the following, we will therefore analyze the projectile-energy dependence of the electron-momentum distributions for D^- colliding with Ar, which was the target investigated in [8].

In Fig. 7, we present the same electron-momentum distributions as in Fig. 3, but for $D^- + Ar$ collisions. These spectra are not dissimilar to those for the He target insofar as structures at $p_{el} = 0$ and $p_{el} \approx v_p$ are visible. However, for 70 keV (center panel), the maximum near v_p is much less pronounced than for He, barely visible at 120 keV (right panel), and not visible at all for 20 keV (left panel). This may be taken as a first hint that the nonsequential channel, relative to the sequential channel, is much more important than for the He target.

This is further supported by the projectile-energy dependence of R , which is plotted for Ar in Fig. 8. Once again, the statistical error bars are smaller than the size of the data points. Like for He, $R(E_p)$ also exhibits an increase near the threshold energy (60 keV, marked by the arrow in Fig. 8). However, the step has a narrower width (about 20 keV) than for He. This is expected based on the smaller binding energy of Ar: the kinetic energy of the (quasifree) projectile electron is given by $K_{el} = 1/2 M_p (v_p + v_i)^2$, where v_i is the electron velocity in the initial bound projectile state. At the threshold, the required v_p is determined by the binding energy ϵ_i of the target. Since the width of the threshold is given by the cross term $M_p v_p \cdot v_i$ it increases with ϵ_i . Below the threshold energy, the behavior

is rather different from He. Here, R is nearly flat and only a shallow (if any) minimum is visible below the threshold. Overall, for Ar, R ranges from 0.6 to 1.1 compared to 0.25 to 0.9 for He. This is also consistent with a stronger role played by the nonsequential channel in Ar compared to He. Finally, no clear signatures of the sequential channel could be identified in the momentum spectrum for 20 keV $D^- + Ar$. Therefore, the present data for Ar support the conclusion of [8] that for 300 keV $Si^- + Ar$, which corresponds to the same v_p as 20 keV $D^- + Ar$, the nonsequential process plays an important role in DI even well below the threshold for the nonsequential process.

V. CONCLUSIONS

We have investigated the projectile-energy dependence of triple-differential momentum spectra of electrons ejected in detachment with simultaneous target ionization (DI) in collisions of D^- with He and Ar. For both targets, a threshold behavior was observed signifying the importance of the non-sequential DI channel in which both electrons are ejected through a single mutual interaction. In contrast to 300 keV $Si^- + Ar$, the electron momentum spectra also revealed, apart from the peak at $p_{el} = 0$, a structure at $p_{el} \approx v_p$. Its intensity relative to the one of the peak at $p_{el} = 0$ maximizes just below the threshold and it is more important for the He target. Previously we assumed that contributions near $p_{el} \approx v_p$ are essentially entirely due to the sequential process. However, our first-order calculations suggest that the nonsequential process can also contribute to a structure around v_p , although not nearly as strongly as the sequential channel. At small projectile energies, the data for both targets differ considerably: while for Ar the energy dependence of the ratio R between the contents of the maxima at $p_{el} = 0$ and $p_{el} \approx v_p$ is nearly flat, it strongly increases with decreasing energy for He.

In the case of the Ar target, the data provide further support for the conclusion that the nonsequential process plays an important role even well below the threshold, as discussed in [8]. For the He target, the situation is more complicated. The strongly rising R with decreasing projectile energy is very difficult, if not impossible, to explain in terms of the non-sequential process. Rather, it is due to a “shoulder structure” in the cross sections for the $p_{el} = v_p$ region at small E_p . It is thus more closely related to the sequential channel.

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DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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