Role of Projectile Coherence in Close Heavy Ion-Atom Collisions

K. Schneider, M. Schulz, X. Wang, A. Kelkar, M. Grieser, C. Krantz, J. Ullrich, R. Moshammer, and D. Fischer

Phys. Rev. Lett. 110, 113201 — Published 12 March 2013
DOI: 10.1103/PhysRevLett.110.113201
We have measured fully differential cross sections for single ionization and transfer-ionization (TI) in 16 MeV O^{7+} + He collisions. The impact parameters mostly contributing to single ionization are about an order of magnitude larger than for TI. Therefore, the projectile beam was much more coherent for the latter compared to the former process. The measured data suggest that, as a result, TI is significantly affected by interference effects which are not present in single ionization.
Even after decades of research the description of the reaction dynamics in atomic fragmentation processes remains a challenging, but at the same time also important task in physics. Especially ionization of atoms by charged particle impact has been studied extensively (for reviews see e.g. [1,2]). Fully differential measurements [e.g. 3-10] played an important role in supporting theoretical efforts by providing data to sensitively test different models. At least in the case of electron impact the combined experimental and theoretical efforts have led to a satisfactory understanding of single ionization, although some quantitative discrepancies remain [5]. In the last 15 years various non-perturbative approaches were developed which achieve impressive agreement with experiment [e.g. 11-14] even at small projectile energies, which was considered the most challenging regime.

For ion impact the theoretical challenge proved to be significantly larger. Because of the difficulties associated with the much larger projectile mass fully quantum-mechanical, non-perturbative approaches incorporating the interactions within all particle pairs in the collision system have not been developed yet. Only recently, semi-classical non-perturbative calculations (for which the large projectile mass actually turns into an advantage) were reported [15,16]. Theoretical efforts were therefore focused on perturbative approaches [17-19]. At large perturbation parameters $\eta$ (projectile charge to speed ratio) these models were not able to reproduce fully differential experimental data [8] even qualitatively. Nevertheless, it was generally assumed that ionization at small $\eta$, which was regarded as a much less challenging regime, was basically understood. It was therefore surprising when considerable discrepancies between experiment and theory were reported for $\eta$ as small as 0.1 [7]. Even with non-perturbative models the experimental data could not be reproduced [15]. Only recently, some
qualitatively improved agreement was achieved, but significant quantitative discrepancies remained [16].

These discrepancies at small $\eta$ were vividly debated for many years [e.g. 15-27], but an important step towards resolving this puzzle was only achieved after nearly a decade by the consideration of the projectiles’ coherence properties [28 - 30]. For ionization of $\text{H}_2$ by proton impact it was demonstrated that interference, due to indistinguishable diffraction of the projectile from the two atomic centers in the molecule, was present in the scattering angle dependence of the cross sections if the projectile beam was coherent, but absent for an incoherent beam [28]. A requirement for observable interference is that the transverse coherence length of the projectile beam $\Delta r$ is larger than the separation of the diffraction centers, i.e. the internuclear distance $D$. $\Delta r$, in turn, is given by the dimensions of a collimating slit of width $a$ placed at a distance $L$ from the target by $\Delta r = (L/2a)\lambda$, where $\lambda$ is the de Broglie wavelength. In ref. [28] the experiments were performed for two different values of $L$ corresponding to $\Delta r > D$ and $\Delta r < D$, respectively. These studies showed that differential scattering cross sections can depend on the projectile transverse coherence properties.

It was further suggested that the discrepancies between experiment and theory in the fully differential ionization cross sections for atomic targets at small $\eta$ may also be related to the projectile coherence [28]. For example, the first-order transition amplitude and a higher-order amplitude involving the interaction between the projectile and the residual target ion (PT interaction) can contribute to the cross sections. In current theories, $\Delta r$ is assumed to be infinite and the coherent sum of both amplitudes can thus lead to interference. Here, the coherence requirement is $\Delta r > R_{\text{eff}}$, where $R_{\text{eff}}$ is the effective size of the diffracting object (this inequality implies that the term “coherence” only becomes meaningful for a given geometry of the
diffracting object). In the experiment, in contrast, $\Delta r$ was three orders of magnitude smaller than a typical atomic size so that no interference was observable. This interpretation was supported by fully differential data for small $\eta$ for a coherent projectile beam achieved by electron cooling at an ion storage ring [29]. Similar effects were reported for electron capture from atomic targets [30].

An alternative method of varying the coherence relation between $\Delta r$ and $R_{\text{eff}}$ is to change $\Delta b$, rather than $\Delta r$, where $\Delta b$ is the difference in impact parameter ranges mostly contributing to the interfering amplitudes. $\Delta b$ can be viewed as a reasonable measure of $R_{\text{eff}}$ if interference is caused by different $b$ leading to the same scattering angle [29,31]. The impact parameter is, of course, not an observable quantity and can not be determined experimentally. Nevertheless, it is possible to select small $b$ mostly contributing to the ejection of an electron by taking advantage of the presence of a second target electron. In the experiment, collision events can be selected in which the second electron also undergoes a transition, e.g. it can be captured by the projectile. At large projectile speeds capture is known to occur at much smaller $b$ than ionization [32-34]. Therefore, with the right choice of $\Delta r$ we can realize $\Delta r < \Delta b$ for pure single ionization and $\Delta r > \Delta b$ for ionization accompanied by capture (transfer-ionization, TI) simultaneously in the same experiment. In this letter we report results on such an experiment which suggest that indeed interference is present for TI, but not for single ionization.

The experiment was performed at the test storage ring (TSR) of the Max-Planck-Institute for Nuclear Physics in Heidelberg [35]. The pulsed 1 MeV/amu O$^{7+}$ beam ($\eta = 1.1$) was cooled by means of electron cooling so that an emittance of about 0.05 $\pi$ mm mrad and a beam width of 1-2 mm was achieved and no collimating slits were required. The coherence length is then determined by the source size of the beam and its distance to the target, rather than the geometry
of a collimating slit. Since in a storage ring the beam is circulating, the effective distance of the source from the target is difficult to specify. However, a reasonable value is provided by the storage ring circumference (55 m), which corresponds to a $\Delta r$ of about 0.5 a.u. The projectile beam was crossed with a cold (T $\leq$ 2K) helium beam from a supersonic jet. Charge-exchanged O$^{6+}$ ions were selected by a dipole magnet and detected by a scintillation detector.

The electrons and recoil ions were extracted by an electric field of about 5.5 V/cm in the longitudinal (projectile beam) direction and detected by two-dimensional position-sensitive channel plate detectors. The electrons were recorded in the forward direction and the recoil ions in the backward direction. Electrons with transverse momenta of less than 1.8 a.u. were guided onto the detector by a uniform magnetic field of 11 G oriented at an angle of about 10° relative to the longitudinal direction so that the detector could be located slightly offset from the projectile beam axis.

The detectors for all three collision fragments were set in coincidence. Furthermore, a timing signal from the projectile beam buncher was recorded. Single ionization events are identified by electron – He$^{1+}$ coincidences and TI events by triple coincidences between a charge-exchanged O$^{6+}$ projectile, a He$^{2+}$ ion, and an ejected electron. Using the two-dimensional position and coincidence time information the electrons and the recoil ions were fully momentum-analyzed. The momentum transfer from the projectile to the target atom (i.e. to the nucleus and to both electrons) $q = p_{\text{rec}} + p_{\text{el}} (+ v_o)$ (where $p_{\text{rec}}$ and $p_{\text{el}}$ are the recoil ion and ejected electron momenta and $v_o$ is the momentum of the captured electron) is then determined from momentum conservation. The captured electron momentum of course only occurs for TI and is thus set in parenthesis. The recoil-ion momentum resolution was about 0.2 a.u. full width at half maximum.
(FWHM) in the longitudinal and 0.35 a.u. FWHM in the transverse direction. For the electrons the resolution was better than 0.1 a.u. FWHM for both components.

In the following, we will present fully differential cross sections (FDCS) for electron ejection in single ionization and in TI. We use a coordinate system which is linked to the scattering (z,x) plane spanned by \( \mathbf{v}_o \) and \( \mathbf{q} \). The polar electron emission angle \( \theta_{el} \) is measured relative to \( \mathbf{v}_o \), which is along the z-axis, and the azimuthal angle \( \varphi_{el} \) is measured in the xy-plane (dubbed the azimuthal plane) about the \( \mathbf{v}_o \)-axis. The x-axis is defined by the direction of \( \mathbf{q}_{tr} \), the transverse component of \( \mathbf{q} \), and \( \varphi_{el} = 90^\circ \) and \( 270^\circ \) coincide with the direction of \( \mathbf{q}_{tr} \) and \( -\mathbf{q}_{tr} \), respectively. Our coordinate system is unconventional in so far as both \( \varphi_{el} \) and \( \theta_{el} \) run from 0 to 360°, i.e. part of the three-dimensional space is covered twice in the combination of the \( \varphi_{el} \)- and \( \theta_{el} \)-dependencies. The advantage is that the behavior of the FDCS in the hemispheres containing \( \mathbf{q} \) and \( -\mathbf{q} \) can be seen in a single plot for both dependencies. In Fig. 1 the FDCS are shown for electrons ejected into the scattering plane (left panel), i.e. for \( \varphi_{el} \) fixed at 90° and 270°, in single ionization (open symbols) and in TI (closed symbols) as a function of \( \theta_{el} \). The corresponding FDCS for electron emission into the azimuthal plane, i.e. for \( \theta_{el} \) fixed at 90°, are shown in the right panel of Fig. 1 as a function of \( \varphi_{el} \). The ejected electron energy and the magnitude of \( \mathbf{q} \) are fixed at 8 eV and 1.5 a.u. The data for TI were normalized to single ionization in the binary peak for a better comparison of the shape of the electron angular distribution.

The FDCS for electron ejection into the scattering plane in single ionization exhibit a pronounced peak structure, the well-known binary peak, at \( \theta_{el} = 90^\circ \), which is approximately the direction of \( \mathbf{q} \). The recoil peak, which is usually observed at small \( \mathbf{q} \) in the direction of \( -\mathbf{q} \), is rather weak at this relatively large \( \mathbf{q} \). While we observe non-negligible intensity near \( -\mathbf{q} \) it does
not form a separate maximum. In the binary peak we observe a shoulder on the small-angle wing of the maximum, which can be explained in terms of post-collision interaction (PCI) between the outgoing projectile and the ejected electron. At large $\eta$ PCI is known to enhance the electron flux in the forward direction [e.g. 8].

In the FDCS for TI we find a peak structure which is significantly shifted in the forward direction compared to single ionization. This is the expected behavior if in TI electron-electron correlations do not play an important role and if it proceeds through two independent interactions of the projectile with the electrons. One might expect that the angular distribution of the ejected electrons should then not be affected by the capture step. However, this is not quite true in spite of the independence of the electron ejection form the capture step because the latter strongly selects small $b$. In the experiment this is reflected by $d\sigma/dq_{tr}$ for TI maximizing at a transverse momentum transfer $q_{tr}$ which is nearly an order of magnitude larger than for single ionization. For such close collisions most of the momentum transfer goes to the target nucleus, rather than the ejected electron. Therefore, for the same $q$ the momentum transferred to the electron, which determines the electron emission pattern, is smaller in TI than in single ionization. With decreasing magnitude of $q$ its direction moves increasingly to the forward direction, which is reflected in the observed shift of the binary peak in the forward direction for TI compared to single ionization. For correlated TI mechanisms, in contrast, peak structures are expected either at $\theta_{el} = 90^\circ$ [36] or in the backward direction [37-40]. Indeed, at this relatively large perturbation parameter ($\eta \approx 1.1$) a dominance of the uncorrelated TI mechanism is expected.

Ignoring possible coherence and interference effects the large fraction of the momentum transfer going to the target nucleus in TI should have a further effect: since the correlation
between the ejected electron momentum and \( q \) is weakened by the large recoil-ion momentum. The width of the binary peak should be much broader in TI than in single ionization. This width is further increased due to the Compton profile of the initial state, which on average is broader for TI because the electron is ejected either from He\(^+\) (if capture precedes ionization) or from He\(^0\) (if ionization precedes capture), while in single ionization it is always ejected from He\(^0\). Therefore, the characteristic binary/recoil double lobe structure, usually found in single ionization (at least for small \( \eta \)), should be “smeared out” in TI. Instead, in the azimuthal plane the width of the angular distribution in TI is nearly the same as in single ionization (for larger \( q \) it is even smaller). More importantly, in the FDCS for TI in the azimuthal plane we do observe a peak structure in the direction of \(-q_{tr}\) separated from the binary peak by a pronounced minimum.

For single ionization, in contrast, no minimum is found and the FDCS in the lower hemisphere of the polar plot containing \(-q_{tr}\) is nearly isotropic.

It could be argued that the comparison between the FDCS for TI and single ionization is not adequate because for a fixed \( q \) a very different amount of momentum is transferred to the electron in both processes. This is manifested, for example, in a smaller FDCS (relative to the binary peak) in the direction of \(-q_{tr}\) in single ionization compared to TI. The observation that the minimum, leading to a separate binary/recoil double lobe structure in TI, is missing in single ionization, is therefore not necessarily significant. For this reason we have also analyzed the FDCS integrated over all \( q_{tr} \), which also has the pleasant side-effect of much improved statistics compared to the FDCS. It should be noted that this integration only refers to the magnitude of \( q_{tr} \), but not to its direction. The azimuthal angle \( \phi_q \) of \( q \) is still determined and fixed for each event and \( \phi_{el} \) is still measured relative to \( q_{tr} \). These integrated cross sections are differential in the solid angle \( \Omega_{el} \) and \( E_{el} \) as well as in \( \phi_q \), i.e. they are fourfold differential (considering that...
\(d\Omega_{el}\) is a double differential), and we refer to them as 4DCS. In Fig. 2 the 4DCS for \(E_{el} = 8\) eV are shown for the scattering plane (left panel) and for the azimuthal plane (right panel), where open and closed symbols again represent single ionization and TI, respectively.

In the scattering plane the 4DCS for single ionization and TI resemble each other much more than the FDCS. Furthermore, in the azimuthal plane the ratio between the 4DCS in the directions of \(q_{tr}\) and \(-q_{tr}\) is identical for single ionization and TI, in contrast to the FDCS. These features illustrate that indeed comparing 4DCS for both processes is more meaningful than FDCS. Otherwise, the basic features of the 4DCS are very similar to what we discussed already in the context of the FDCS: the angular distribution for TI is remarkably narrow and, in fact, this time even significantly narrower than for single ionization. More importantly, again we find a pronounced minimum, separating the binary and recoil peaks from each other, in the TI data, but not in the single ionization data. These features we interpret as due to interference effects in TI, which are not present (or at least weaker) in single ionization. Without interference, one would expect that the large fraction of the momentum transfer going to the recoil ion in TI should lead to a broadening in the angular dependence of the ejected electrons. The stronger PT interaction, due to the smaller \(b\) (compared to single ionization), should then weaken the correlation between the electron momentum and \(q\) one gets in a first-order process, where \(q\) goes exclusively to the electron.

The comparison between TI and single ionization is similar to the FDCS measured for single ionization at small \(\eta\) for a coherent and incoherent projectile beam [29]: in the azimuthal plane the data for TI look like those for the coherent beam and the data for single ionization like those for the incoherent beam in ref. [29]. This similarity further suggests that the difference between the present TI and single ionization data is also due to the coherence properties of the projectile.
beam. The impact parameter range which mostly contributes to single ionization is at this relatively large $\eta$ larger than the size of the target atom ($\approx 1$ a.u.), while for capture at this high projectile speed it is much smaller than the target atom size, as mentioned above. For the coherence length in the present experiment ($\approx 0.5$ a.u.) the projectile beam is then indeed incoherent for single ionization, but at least partly coherent for TI. The differences between the TI and single ionization cross sections are therefore consistent with expectations based on the projectile coherence properties and we interpret the surprisingly narrow width of the FDCS for TI and the minimum between the binary and recoil peaks as a destructive interference effect.

In summary, we have performed a comparative study of single ionization and transfer ionization TI. Because single ionization mainly takes place at relatively large and TI at small impact parameters the coherence length of the projectile beam relative to the effective size of the diffracting object was much larger in the latter than in the former case. This can lead to interference effects between different amplitudes in the TI cross sections, which are absent (or weaker) in single ionization. The present data thus provide once again strong indications that atomic scattering cross sections can sensitively depend on the projectile coherence. However, an important advantage over our previous studies on this topic is that the projectile beam can be simultaneously coherent and incoherent, depending on the process. We can therefore safely rule out that experimental artifacts, introduced e.g. by the projectile beam profile or by the experimental resolution, can lead to differences between the cross sections for different processes, i.e. between a coherent and an incoherent beam.

We thank the MPIK accelerator and TSR team for their experimental support. This work was funded through the Emmy-Noether program of the German Research Council (DFG), under grant No. FI 1593/1-1. We are grateful for the support by the Alliance Program of the Helmholtz
Association, under grant No. HA216/EMMI. M.S. acknowledges the support by the National Science Foundation, under grant No. 0969299, and a Mercator scholarship by the Deutsche Forschungsgemeinschaft.

Figure Captions

Fig. 1: Measured fully differential cross sections (FDCS) for electrons with an energy of 8 eV ejected into the scattering plane (top panel) and into the azimuthal plane (bottom panel). The open symbols represent single ionization data and the closed symbols transfer-ionization data. The momentum transfer was fixed at 1.5 a.u.

Fig. 2: Same as Fig. 1, but the FDCS were integrated over all magnitudes of the momentum transfer.