Momentum Images of Continuum Electrons from He^{+} and He^{2+} on He: Ubiquity of π Structure in the Continuum

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Cold target recoil ion momentum spectroscopy has been used to image the momentum distributions of continuum electrons liberated in the impact of slow He^{+} and He^{2+} ions on He. The distributions were measured for fully determined vector impact parameter. The spectra show that the electron momenta lie mainly in the collision plane and display a structure which strongly suggests that it is π states which are dominantly promoted into the continuum in such collisions, but that interfering σ and perhaps δ amplitudes are also important. [S0031-9007(98)07407-9]

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It was established more than three decades ago that in slow collisions for which the projectile velocity v_p is much smaller than the classical electron orbital velocity, a molecular promotion picture of electronic excitation holds [1]. One of the earliest cases to be studied was the electronic excitation of He by extremely slow He^{+} projectiles [2]. The major excitation mechanism was found to be a rotational conversion of the initial $2p\sigma$ orbital into a $2p\pi$ orbital, which was subsequently promoted to an excited $2p$ state of the projectile/target. It was suggested, but not established, that this process might also provide a route for promotion to the continuum. How high in velocity such a promotion picture might continue to be valid has remained a substantial question over the years. In recent years, much effort has been focused on identifying the mechanism for collisional target ionization of simple systems in the "quasithreshold" velocity region where continuum electron production first begins to compete with the dominant electron capture process (roughly, for v_p between about 0.01 and 1 a.u.). The discussion initially focused on the role of "saddle-point" [3,4] electron production, whereby electrons liberated from the target ride on the potential saddle formed by the Coulomb potentials of the receding centers. Experimental attempts to identify a saddle-point feature in the continuum electron spectra resulted in both positive and negative evidence for such a mechanism but at velocities so high that the whole concept of saddle-point electrons was questionable. On the theoretical side, the theory of hidden crossings, whereby an electron can be promoted into the continuum via series of hidden crossings in the complex R plane of the quasimolecular states [5–7], has allowed the identification of two major mechanisms for ionization, the T and the S processes. In this language, the T process corresponds to a saddle-point promotion. Pieksma *et al.* [8] studied p on H at very low projectile velocity where the molecular language is most appropriate. They were able to deduce, from soft electron spectra extending down to zero energy, evidence for the operation of both T and S processes. More recent electron momentum imaging of the continuum electron spectra showed that, while the soft electron continua tend to gather about the saddle for slow singly charged projectiles on He, they do not do so for higher charged projectiles [9–11].

The whole subject of soft electron production has been profoundly changed over the last few years through momentum imaging techniques which produce complete "pictures" of the momentum spectra. A major advance came from the recent work of Dörner *et al.* [12] who used cold target recoil ion momentum spectroscopy (COLTRIMS) [13,14] to study electron ejection in the p-He system at energies of 5, 10, and 15 keV. Their technique allowed momentum-space images of the continuum electron spectra to be obtained for *experimentally determined vector impact parameter* (collision plane and transverse momentum transfer). The ejected electrons were found to be concentrated in the scattering plane and preferentially emitted with velocities between zero and v_p, that is, in the vicinity of the sadde-point velocity. The distribution in the collision plane displayed a two-finger structure, with the electron momenta lying preferentially above or below the beam axis. Furthermore, whether the preference was toward or away from the recoil ion depended on the projectile velocity. It was suggested that this behavior could be explained as due to the dominance of a π amplitude in the continuum, caused by the promotion to the continuum of an initially excited π molecular orbital. The oscillation with v_p was attributed to interference between π and σ amplitudes to the continuum. This suggestion has now been examined quantitatively for p on H by Macek and Ovchinnikov [15], who have obtained a theoretical value for the oscillation frequency consistent with that found by Dörner *et al.*, and who point out that this oscillation frequency can be interpreted as a direct measure of the real part of the energy difference between the relevant π and σ molecular orbital energies.

In this paper we examine two systems for which electronic excitation in the low velocity limit is well established to originate from the rotational excitation of the $2p\pi$ orbital. We have used COLTRIMS to record the continuum momentum-space images of the electron continuua for experimentally determined vector impact parameter. We find that the structure reported by Dörner *et al.* [12] is not specific to p-He but appears in both of our systems and appears to be quite general, although distinct differences of detail in the three systems are apparent. Our results suggest that the π promotion is likely to be a very common starting point for promotion to the continuum for any low-charged, light collision systems.

The experiment was carried out at the KSU CRYEBIS facility with a COLTRIMS system very similar to that described in [12]. The ion beam (z axis) collided with a supersonic gas jet (y axis). An electric field (x axis) of 35–60 V/cm perpendicular to both the ion beam and the gas jet was used to extract the recoil ions and slow ejected electrons in opposite directions. The electrons were accelerated by this field a distance of 10.2 mm, then drifted a distance of 17.7 mm to be detected by a position-sensitive detector. The recoil ions were accelerated in the opposite direction through a distance of 86.3 mm and then drifted a distance of 305 mm to be detected by another position-sensitive detector. The time of flight of the electron was small and had a small spread. This allowed us to use the electron time signal as a start for the time-to-amplitude converter which was stopped by the recoil time signal. The measured time of flight of the recoil ions was used in calculating one of the momentum components of the recoil ions. The two other components were calculated from the position on the recoil detector. Two of the three momentum components of the electrons were calculated from the electron position on the electron detector.

A typical recoil-ion transverse momentum spectrum and coordinate system schematic are given in Fig. 1. Recoil ions whose transverse momentum vectors lie within gate G1 occur when the recoil ion is scattered in the $+y$ direction. Selecting events in this gate thus defines a scattering plane parallel to the y, z plane, which is parallel to the electron detector ("top view"). Selecting events in gate G2 defines a scattering plane parallel to the x, z plane and therefore perpendicular to the electron detector ("side view"). The top view thus provides a view of the electron momentum image seen looking down on the scattering plane (yz) while the side view provides a similar view seen looking at the scattering plane edge-on (xz).

Electron momentum distributions for top and side views are given in Figs. 2(a) and 2(b), respectively, for the case of 0.64 a.u. He^{+}-He. Black rectangles represent the appropriate experimental resolutions in the images. The electrons are seen to be preferentially emitted in the forward direction, lying between the target and projectile velocities

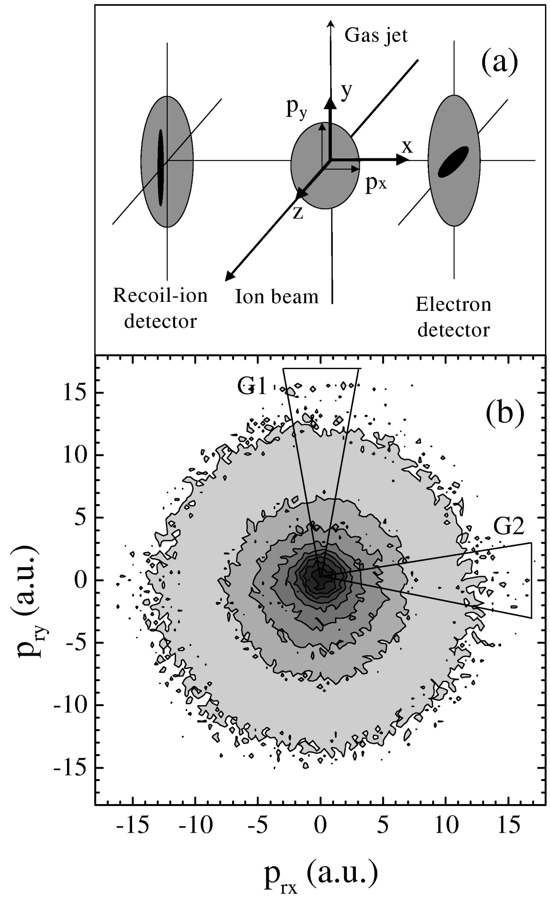

FIG. 1. (a) Geometry of target showing coordinate system. The extraction electric field is in the negative x direction. (b) Two-dimensional recoil-ion transverse momentum distribution for $v_p = 0.64$ a.u. He^{+}-He collisions. Gates G1 (top view) and G2 (side view) are used in data analysis.

in agreement with earlier investigations of similar systems [9–12]. Figure 2(b) shows that electrons are concentrated in the scattering plane. The most striking feature of these spectra is the two-fingered structure seen in spectrum (a) showing a local minimum on the internuclear axis. (In all top-view figures the recoil ion moves down after the collision.) This local minimum can be seen more clearly by making a transverse projection of a slice of the two-dimensional distribution near $v_{ez}/v_p = 0.5$. Such a projection of Fig. 2(a) is shown in Fig. 2(c). A similar projection of Fig. 2(b) is shown in Fig. 2(d). This structure is very similar to that seen by Dörner *et al.* for the p-He system. To investigate the generality of this structure, we also performed measurements for He^{2+} on He and show spectra corresponding to (a) and (b) for this system in Figs. 2(e) and 2(f). Both the concentration of events in the collision plane and the quasinodal line along the internuclear axis are seen to appear for this system also. It is interesting to note, however, that, while for He^{+} on He more electrons are emitted away from the recoil direction, the electrons follow the recoil for He^{2+} on He.

The interpretation of the two-fingered structure seen by Dörner *et al.*, and now reinforced by the calculation

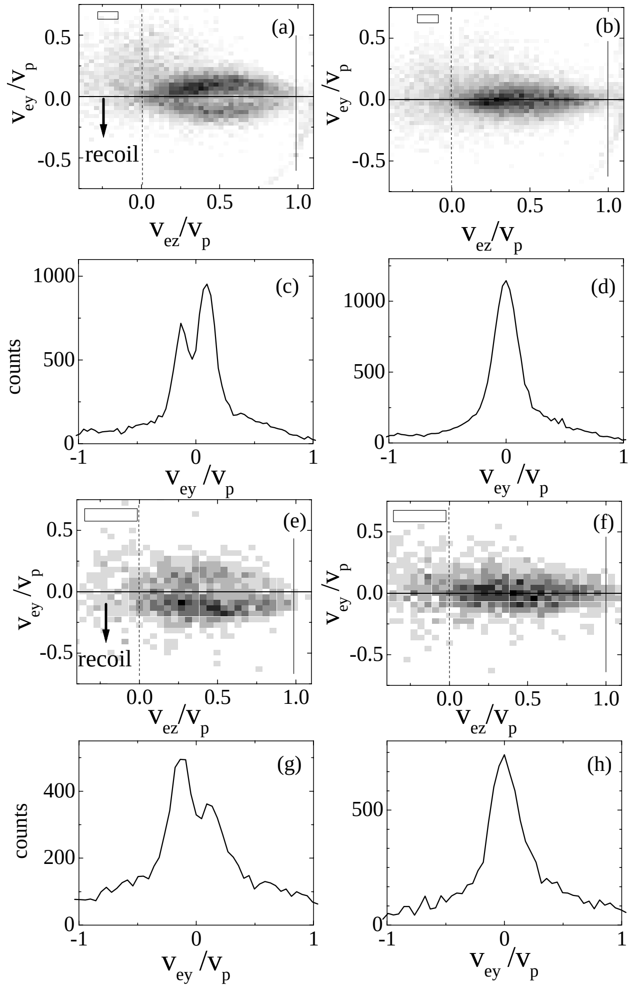

FIG. 2. (a) Top view and (b) side view of the emitted electrons in the collision $v_p = 0.64$ a.u. He^+-He. (c) and (d) are transverse projections of (a),(b) near $v_{ez}/v_p = 0.5$. (e) and (f) are, respectively, top view and side view of the emitted electrons in the collision $v_p = 0.55$ a.u. He^{2+} with He, and (g) and (h) are the corresponding projections. The approximate resolution functions in the two-dimensional distributions are represented by black rectangles.

by Macek and Ovchinnikov, attributes this structure to the promotion of molecular orbitals of π character into the continuum. Such an explanation seems natural for the present system as well. Specifically, the electron initially occupying the $2p\sigma$ orbital in the He-He^+ molecule is first rotationally coupled to a $2p\pi$ state at small internuclear distances and then promoted by a T-type process into the continuum, keeping its π character. The nodal line along the internuclear axis is a signature of that character. The asymmetry in intensity about this quasinodal line, showing more intensity in the direction away from the recoil, can be interpreted as due to the interference of the π amplitude with a weaker σ amplitude, which would both produce an asymmetric distribution and weaken the node.

To study the impact-parameter dependence of the emitted electron momentum distribution in the scattering plane, the electron distribution was gated on different ranges of recoil-ion transverse momentum. The results are shown in Fig. 3. In these figures the recoil-ion transverse momentum increases in moving from Fig. 3(a) to Fig. 3(e) and, therefore, the impact parameter decreases by moving the same way. The corresponding transverse projections of slices from each distribution near $v_{ez}/v_p = 0.5$ are given in Figs. 3(f)–3(j). The figures show that the distribution exhibits the two fingers for all ranges of impact parameter. However, the distribution becomes more symmetric at smaller impact parameters (larger recoil-ion transverse momenta). This suggests that the relative contributions of σ and π amplitudes depend on the impact parameter. At small impact parameters the π orbital is dominant over σ orbital. This leads to a symmetric distribution around the internuclear axis. At large impact parameters the σ orbital contribution increases relatively, leading to the asymmetry of the distribution.

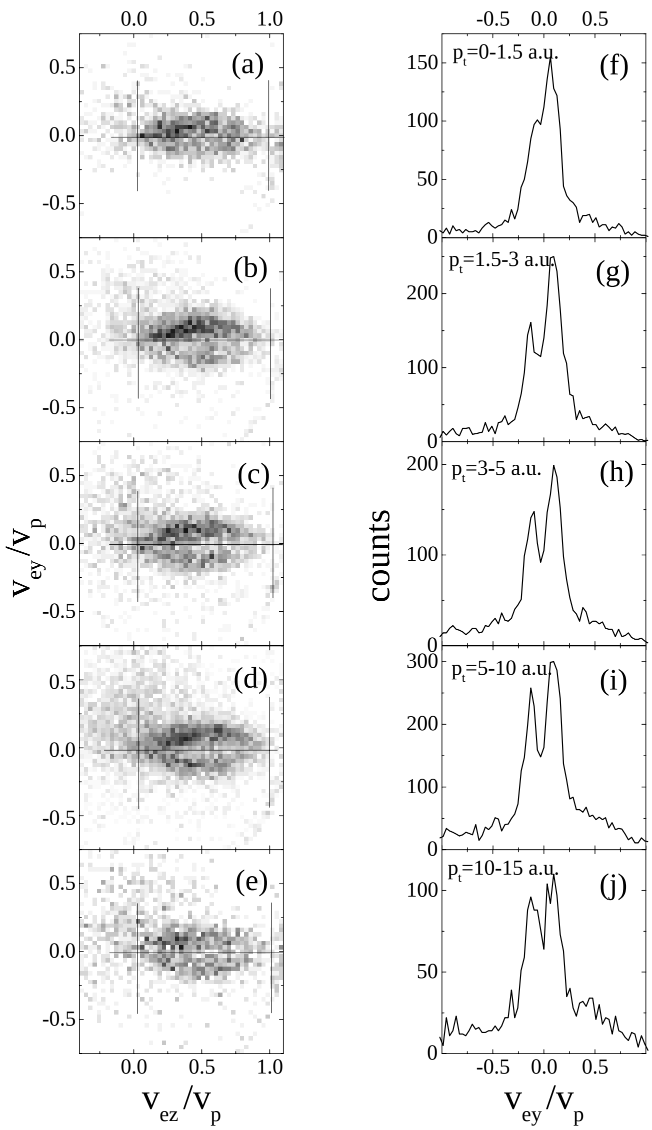

FIG. 3. (a)–(e) Top view of the ejected electron momentum distribution in the collision 0.64 a.u. He^+ on He for different windows on the magnitude of the recoil transverse momentum transfer (p_t). (f)–(j) Vertical slices from (a)–(e), centered at $v_{ez}/v_p = 0.5$, projected onto the v_{ey}/v_p axis.

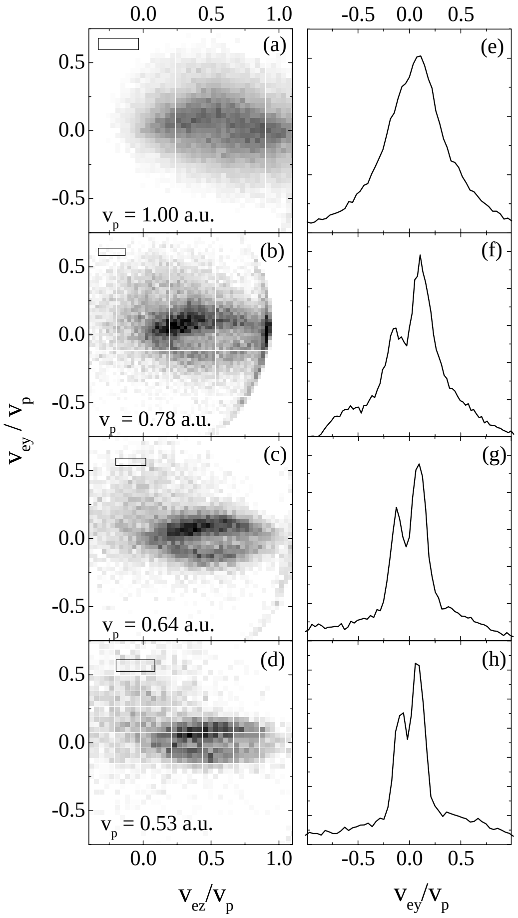

FIG. 4. (a)–(d) Top view of the ejected electron momentum distributions for He^+ on He with $v_p = 0.45-1.00$ a.u. The black rectangles represent the resolution functions. (e)–(h) Vertical slices of (a)–(d) centered at $v_{ez}/v_p = 0.5$, projected onto the v_{ey}/v_p axis.

For the p-He case, a strong oscillation with v_p of the relative intensity of the two "fingers" was seen and interpreted by Macek and Ovchinnikov in terms of the $\pi - \sigma$ interference. We do not observe such an oscillation for the present cases. Figure 4 shows electron momentum images similar to that of Fig. 2(a) for several projectile velocities. The stronger finger is insistently that on the side opposite that in which the recoil goes. Thus it appears that the relative phase of the π and σ amplitudes is not highly v_p dependent, as it was for the p-He system, perhaps suggesting that the energy difference between the π and σ promotion routes is smaller for the He-He^+ case. The fact that the stronger intensity is towards the recoil for the He-He^{2+} case also remains without explanation. We also note that, at the highest velocity for the He-He^+ system, the intensity actually seems to be moving preferentially toward the projectile, suggesting that a δ amplitude may also be coming into play. It is clear that these data contain information on the participating amplitudes which remains to be explained by theoretical treatments.

In conclusion, we have used electron momentum-space imaging, together with vector impact parameter determination, to show that the electrons emitted into the continuum for slow He^+ and He^{2+} ions on He have structure with at least two very general characteristics: First, the electrons are concentrated in the collision plane and lie roughly between the projectile and the target in longitudinal velocity space. Second, the distributions viewed perpendicular to the collision plane display a structure reminiscent of a "textbook" π orbital. This behavior suggests strongly that the promotion of π orbitals dominates the low-energy continuum electron production for all of these systems. However, details of the spectra differ for the three systems. From such detail we would hope that quantitative information concerning the relative strengths and phases of amplitudes representing different promotion paths might be deduced and interpreted.

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