Electrons Ejected with Half the Projectile Velocity and the Saddle Point Mechanism in Ion-Atom Collisions

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Full three-dimensional ejected electron momentum distributions for proton impact ionization of atomic hydrogen are calculated for impact energies 10 through 50 keV. The distributions show a peak in the longitudinal momentum at half the projectile impact velocity: the $v/2$ peak. A quantitative assessment of saddle point ionization, based on quantum and classical analysis, reveals that the $v/2$ peak is a false indicator for this mechanism. The influence of the potential saddle on ionization is seen to decrease rapidly from 10 to 50 keV.

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"To understand hydrogen is to understand all of physics" [1]. This short statement is easily extended to summarize the motivation of studying proton-hydrogen collisions, in particular, for impact ionization which is a prototype of the breakup of three charged particles. For impact velocities much larger than the "matching" velocity (where the incoming projectile is moving at the same velocity as the average speed of the electron in the target atom), $v_0 = 1$ a.u., perturbative treatments of ionization work well. At impact velocities near or below the velocity matching, where ionization probability is comparable or smaller than the excitation and/or charge transfer probabilities, a nonperturbative treatment of the collision process is needed. With large scale close-coupling calculations, using atomic orbitals, one obtains total ionization cross sections within 10% to 20% of the experimental results [2], but from this theory no attempt has been made to extract the distributions of the ejected electrons. The subject is full of controversial issues, as summarized by a recent monograph [3]. One recent issue is the discrepancy between two quantal calculations of the distributions of ejected electrons [4,5]. Much of the debate centers on a model proposed by Olson [6,7] which offers an explanation for the bulk of ionization in *near-matching velocity* ion-atom collisions—the "saddle point" (SP) mechanism. The SP mechanism is extremely interesting, because the idea should apply to ion-atom collisions in general.

The SP mechanism says that ejected electrons should collect around the point where the force from both protons cancels. In the laboratory frame, these electrons will then have a velocity of $v/2$, where v is the projectile ion velocity. Olson, performing calculations by the classical trajectory Monte Carlo (CTMC) method, assigned ejected electrons which have longitudinal velocity near the saddle velocity $v/2$ to SP ionization [7]. His calculations claimed that SP ionization was the dominant mechanism at intermediate energies of 40 to 60 keV. In this Letter we will show that a peak in longitudinal velocity near $v/2$ does not necessarily mean SP ionization mechanism is important. In fact, the saddle point mechanism is important only for collisions at low velocities.

To address the importance of the potential saddle in ionization with a classical calculation, one can analyze the evolution of the electron's energy as the two nuclei separate. Such an analysis was carried out by Bandarage and Parson [8] by defining an electron's energy with respect to a moving molecular frame. Their results supported Olson's conclusion and even predicted that the importance of the SP mechanism *decreases* at lower velocities. Illescas *et al.* [9] extended the classical theory of [8], but concluded that SP ionization is important only at low energies. Based on calculated total ionization cross sections using close-coupling theory, Winter and Lin [10] introduced the analogy between the SP mechanism and the Wannier mechanism for threshold electron impact ionization and concluded that the SP mechanism is important only at low energies. Using hidden crossing theory [11] Pieksma and Ovchinnikov [12] explored SP ionization at low energies, identifying the SP mechanism with the so-called T-series. Neither of these quantum theories predict electron momentum distributions.

Many experiments have been carried out to search SP electrons, with conflicting results. Most of these experiments were carried out for 60–200 keV protons colliding with helium, searching for $v/2$ electrons. Based on our analysis the $v/2$ electron peak in the longitudinal velocity distribution is not a signature of the SP mechanism. A low energy collision experiment for proton-hydrogen system by Pieksma *et al.* did not resolve the issue [13–15]. With the advent of the cold target recoil ion momentum spectroscopy, where detailed ejected electron momentum distributions (EEMDs) are fully mapped out in three dimensions, the need to understand the mechanism of ionization becomes much more urgent. EEMDs have been measured for a number of collision systems [16,17] at low to intermediate impact velocities.

The lack of theory capable of comparing with measured electron distributions has led us to make an *ab initio* study of the most basic ion-atom collision system, proton on

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hydrogen, over a wide range of impact velocities. From this study we offer in this Letter a new definition for saddle point electrons. Furthermore, we show for all impact energies that the ejected electron distribution divides into saddle point and kinetic components based on dominance of the potential and kinetic energy, respectively. At low energy our classification of saddle point electrons is similar to the hidden crossings T-series, and at high energy the kinetic electrons represent the direct impact ionization mechanism. For 10–50 keV collisions we find that *both* SP and kinetic electron distributions peak at $v/2$ in longitudinal momentum. Thus, the $v/2$ peak is *not* an experimental signature exclusive to the saddle point mechanism.

Investigating the proton-hydrogen system, we have recently developed an approach to solving the corresponding time-dependent Schrödinger equation in momentum space, the two-center momentum space discretization (TCMSD) method [5]. Extracting the EEMD revealed a peak at $v/2$ in the longitudinal momentum for 5 to 50 keV and a double-peak structure in the transverse momentum for the 5 to 15 keV range of impact energy. In contrast with the two-state Sturmian model of Ref. [4], an extension of hidden crossing theory, our prediction for the ejected electron distribution did not show strong projectile energy dependence from 5 to 15 keV.

With the aim of exploring both the importance of the SP mechanism and the discrepancy between TCMSD and the Sturmian theory, we compare our quantum mechanical results with CTMC calculations as implemented by Illescas *et al.* [9,18], employing the initial distribution of Hardie and Olson [19]. It has been shown that this version of CTMC does predict good total ionization cross sections compared with the most recent experimental data [20]. With complete quantum information from the TCMSD calculation we can provide an even more stringent check at the level of the probability distributions.

In Fig. 1 we show a comparison of the longitudinal and transverse EEMDs at impact energies 10, 15, 25, and 50 keV and an impact parameter $b = 1.2$ a.u. The ionization probabilities were calculated for an internuclear separation $R = 30$ a.u. The total ionization probabilities from TCMSD (CTMC) are 0.08 (0.04), 0.11 (0.08), 0.13 (0.14), and 0.22 (0.27) for 10, 15, 25, and 50 keV collisions, respectively. The main feature of the longitudinal momentum distribution is a peak at $v/2$. In transverse momentum one sees for 10 and 15 keV that *both* classical and quantum theories show a double-peak structure. At 25 keV the positive transverse momentum peak shrinks, and at 50 keV only the negative transverse momentum peak remains. The CTMC results are closer to the electron distributions of the TCMSD calculation than the results of Ref. [4].

The global agreement between classical and quantum calculations in the electron distribution encourages us to extract the mechanism for ionization based on the classical calculations, in particular, the role of the SP mechanism. The ejected electron spectra in Fig. 1 were calculated at

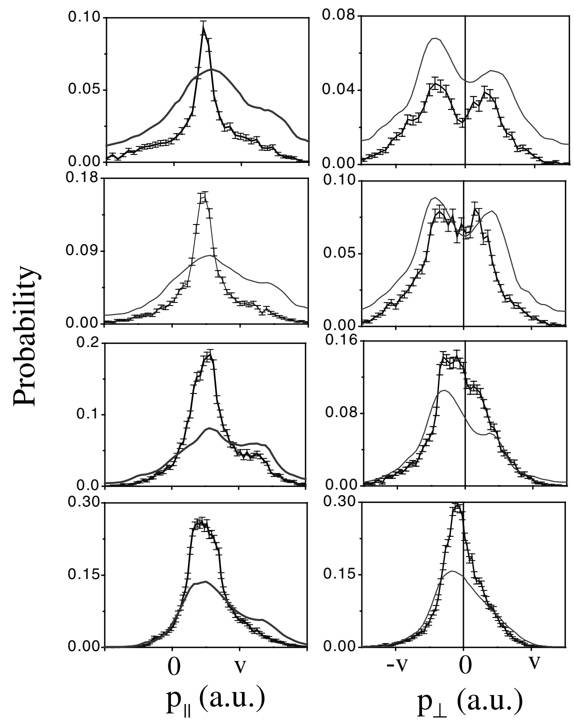

FIG. 1. Projection of classical (thin line with error bars) and quantum mechanical (thick line) ejected electron distributions parallel, $p_\parallel$, and transverse, $p_\perp$ (in collision plane) to projectile motion. The collision system is proton on hydrogen at 10, 15, 25, and 50 keV (from top to bottom).

$R = 30$ a.u. which may be considered insufficient to establish asymptotic behavior. The comparison is limited to this range of internuclear separation by difficulties in performing the TCMSD calculation at large R, as explained in Ref. [5]. There is no difficulty in extending the CTMC calculations to large R. To define the ionization more precisely, we identify the ionized electrons at an internuclear separation of 500 a.u. The ejected electrons were then propagated backwards to lower internuclear separations to examine the ionization mechanism, specifically, SP ionization as a function of R.

Figure 2 shows a slice of the EEMD, near the collision plane, at $R = 30$ for the 15 and 50 keV collisions. Each dot, gray or black, represents an electron trajectory that will be in the continuum at $R = 500$. The feature of Fig. 2 that immediately strikes the eye is the lack of electrons near both the target and projectile velocities. Both holes in the momentum distribution come from eliminating electrons that remain bound to the target or become captured by the projectile ion. To estimate roughly the minimum momentum that an ejected electron can have with respect to a nucleus of charge Z_c (where c refers to either the target or the projectile nuclear core), we assume that the ejected electrons primarily exhibit a free expansion [18]. In a free expansion the distance between two particles is proportional to their relative velocity. Thus, the distance r between electron and nucleus is tp, where p is the

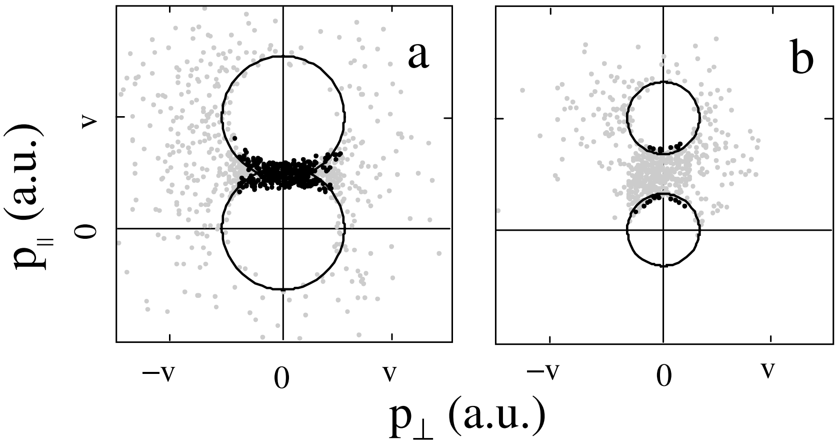

FIG. 2. Scatter plot of ejected electrons, in momentum space, within 0.1 a.u. of the collision plane. Gray and black dots represent kinetic and saddle electrons, respectively; see text. Circles of radius p_{ion} centered on the target and projectile protons indicate the minimum momentum of ejection based on a free expansion model. (a) 15 keV at $z = 30$ a.u. and (b) 50 keV at $z = 30$ a.u.

momentum of the electron and t is time measured from the projectile's closest approach, the beginning of the expansion. Rewriting t in terms of the internuclear separation R and projectile velocity v, we obtain $r \approx (R/v)p$. Evaluating the total energy of the nucleus-electron system, we find a radial momentum p_{ion} which divides bound and free electrons:

$$\frac{p_{\text{ion}}^2}{2} - \frac{Z_c}{(R/v)p_{\text{ion}}} = 0; \qquad p_{\text{ion}} = \left(\frac{2vZ_c}{R}\right)^{1/3}. \quad (1)$$

In Fig. 2 circles of radius p_{ion} are drawn in about both target and projectile centers. In Fig. 2b, most of the electrons are outside the two circles, indicating that electrons which are ejected at $R = 500$ a.u. are already in the continuum at $R = 30$ a.u. for collisions at 50 keV. In Fig. 2a, there is still a nontrivial fraction of electrons, represented by black dots, which lie inside the circles. These electrons have $p_{\parallel}$ near $v/2$—an indication that the potential saddle may play an important role.

To isolate the effect of the saddle potential as the two nuclei recede from each other, we classify the ejected electrons for $t > 0$ into two groups at each z, where $z = vt$: (1) "kinetic" electrons—energy is positive with respect to both target and projectile protons, *individually,* and (2) saddle point electrons—energy is negative with respect to *either* proton. The kinetic electrons represent any electron that is considered ejected at the current z, and they are represented by gray dots in Fig. 2. The saddle point electrons are not free at the current z and require the long-range two-center interaction to reach the continuum. These electrons are represented by the black dots in the figure. Clearly, this definition includes electrons promoted into the continuum on the internuclear potential saddle and from Fig. 2a these electrons are mostly located near the potential saddle.

Following these definitions, we have evaluated the ratio P_S of SP electrons as a function of z as the two nuclei recede for 5, 10, 15, and 50 keV; see Fig. 3. The curves for 5–15 keV exhibit similar behavior. The saddle fraction exhibits a plateau after $z = 8$, where the top of the barrier is the same as the average energy of the initial state. Subsequently, as the saddle pushes upward the SP electrons are promoted to the continuum, and thus the SP fraction decreases monotonically. For low energy collisions this process is stretched out, since the electrons entering the saddle region have a low kinetic energy with respect to the saddle point. Figure 3 shows that the SP mechanism contributes a maximum of 33% to the total ionization probability, decreasing slowly from 5 to 15 keV. On the other hand, the range of SP ionization decreases rapidly with impact energy until finally, at 50 keV, the saddle fraction just drops monotonically to zero.

So far in the literature the SP electrons have been analyzed based solely on the criterion that the longitudinal momenta of the ejected electrons are in proximity of $v/2$. If the potential saddle does indeed play a role in the *dynamics* of the ejection of the electron, then the effect should be reflected in the z dependence of the electron's transverse momentum. For this purpose, we isolated all the electrons that have negative $p_{\perp}$ at $z = 500$ a.u. The momentum distributions of these electrons at $z = 12$ and $z = 30$ are then displayed. In Fig. 4 we show these distributions for the 10 keV collision. The kinetic electrons—moving too fast to be significantly affected by the saddle—remain at negative $p_{\perp}$ from $z = 12$ to 500. The SP electrons, on the other

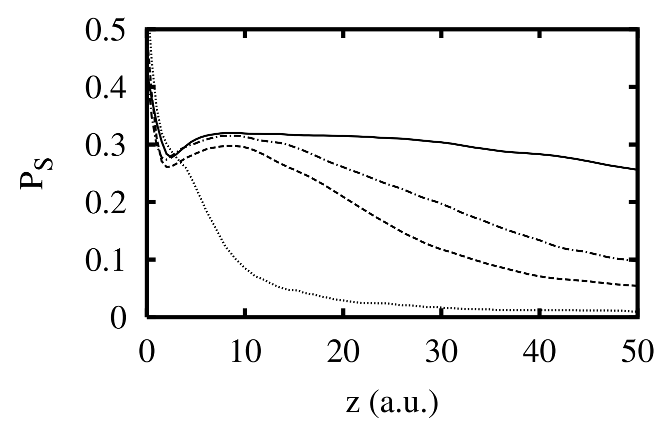

FIG. 3. Fraction of saddle electrons as a function of z. Curves correspond to impact energies 5 keV (solid line), 10 keV (dash-dotted line), 15 keV (dashed line), and 50 keV (dotted line).

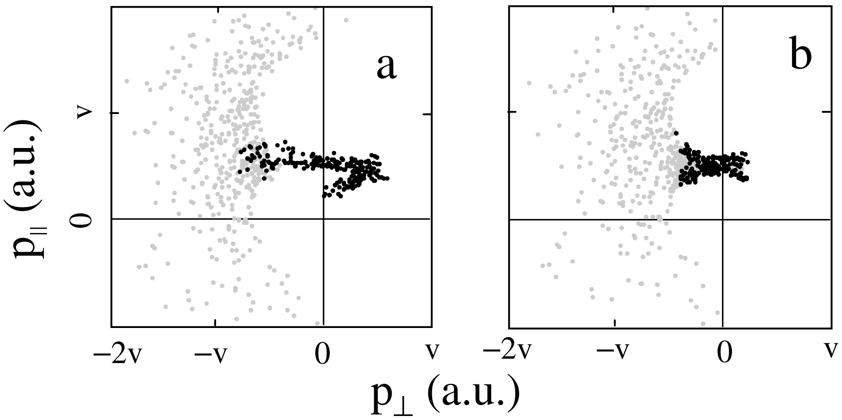

FIG. 4. Scatter plot of ejected electrons, in momentum space, within 0.1 a.u. of the collision plane. Gray and black dots represent kinetic and saddle electrons, respectively. Collision energy is 10 keV. Ejected electrons with $p_{\perp} < 0$ at $z = 500$ a.u. were propagated back to (a) $z = 12$ a.u. and (b) $z = 30$ a.u.

hand—trapped by the transverse binding potential of the saddle—are distributed at both positive and negative transverse momenta before they are ejected to the continuum. Thus, the role of the potential saddle on the SP electrons is firmly established.

Having analyzed SP ionization as a function of internuclear separation, we have established the role of potential saddle for ionization at different collision energies. The analysis of the classical calculations indicates that at $z = 30$ a.u. the kinetic electron fraction is 90% and 99%, respectively, for 15 and 50 keV collisions. Accordingly, we can interpret the longitudinal and transverse momentum distributions of Fig. 1 for 15 and 50 keV with the scatter plots of the kinetic electrons in Figs. 2a and 2b. For 15 keV the ejected electron cloud for $p_{\perp} = 0$ is thinned out simply because of the holes left by removing electrons bound to either proton, leading to the double-peak structure in Fig. 1. For 50 keV the peak at $v/2$ in the longitudinal momentum results again from exclusion of electrons near either proton. As can be seen in Fig. 2b, the $v/2$ peak for this collision does not come about from saddle point electrons. Thus, an experimentally measured $v/2$ peak in the longitudinal momentum does not uniquely identify the SP mechanism. We have also checked using the CTMC calculation that the basic asymmetry in the transverse momentum distributions and the $v/2$ peak in the longitudinal momentum, shown in Fig. 1 for $z = 30$ and impact energies 10–50 keV, remain for larger z.

In summary, we have calculated ejected electron momentum distributions for the proton-hydrogen system near velocity matching. We have provided a classification of ejected electrons into saddle point and kinetic electrons—valid for a broad range of impact energies. The saddle point contribution to ionization is largest at low energy, yet the ejected electron distributions show a peak in the longitudinal momentum at $v/2$ for 10–50 keV collisions. Thus, we conclude that experimental measurement of the $v/2$ peak in the longitudinal momentum distribution of ejected electrons does not necessarily imply the saddle point mechanism.

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