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# Origin of the structures in the excitation cross sections in ion-atom collisions

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It is demonstrated that the structures in the low-energy cross sections for excitation to certain levels of atoms by ions originate from a shift from direct excitation at high and intermediate energies to pronounced two-center molecular mechanisms at lower energies. The molecular mechanisms dominate at small internuclear separations, whereas the direct mechanism is driven by the familiar dipole long-range coupling. The demonstration is based on maps of the excitation probabilities over the collision velocity and impact parameter, which display two typical regions. System specific details determine the separation between the two regions and give rise to a plateau, an oscillation or a hidden shoulder in the excitation cross sections. Maps of classical-trajectory Monte Carlo method results show similar qualitative features, but the details of these maps differ considerably from those of the quantal calculations. [S1050-2947(98)50506-9]

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Most of the cross sections for excitation in ion-atom collisions increase with energy up to a maximum for projectile velocity $v_{\max}$, and decrease in the high-energy region. For many collision systems, however, it has been established [1,2] that a plateau, or in some cases an oscillation, in the excitation cross section, appears roughly at $v_{\max}/2$ or slightly below. These plateaus have been observed in a series of collision systems involving $2s$ and $2p$ excitation of hydrogen [1] as well as in $3p$ excitation in $Z^+$-Na($3s$) collisions [2]. Such plateaus have also been reported in several theoretical calculations, e.g., [3,4]. In Ref. [2] it was shown that for the fixed Na($3s$) target, a plateau appears in the same velocity range for all the studied projectiles, and the authors concluded that the excitation process is coupled to the more dominating capture process. This conclusion is in agreement with Fritsch [5] who first pointed out that the structure disappears if the projectile centered states are removed from the expansion basis.

Very recently, Schultz *et al.* [6] found oscillatory structures in classical-trajectory Monte Carlo method (CTMC) calculations for excitation to the $n=2$ level in $\alpha$-H collisions. They related these structures to the number of times the active electron changes its location from one of the collision partners to the other in a classical description of the electron's motion during the collision (so-called swaps). Such swaps have previously been used to explain the observation of the oscillations in capture cross sections in collisions between ions and Rydberg atoms [7].

In this Rapid Communication we want to point out that the plateaus for excitation from ground-state atoms have a very simple *quantal* origin. We also want to indicate that although the mechanisms involve swaps of the quantal probability of the electron's location, their description by classical calculations might be fortuitous.

A typical quantal calculation will exhibit a number of different mechanisms, related to various regions of the energy curves of the quasimolecular energy diagrams. At large distances, the flat regions of the correlation diagram correspond to the direct mechanism, and at smaller distances avoided crossings and other ''flat'' regions are competing. In general, each of the mechanisms contributes to the cross section by a broad peak on the plot of the cross section versus collision velocity or energy. Depending on the relative position of the maxima (and the number of distinct mechanisms), the total cross section develops one or more plateaus (when the maxima are close), an oscillationlike structure if the maxima are well separated, or just a small variation in the slope of the curves if the new maximum is hidden under a broader and larger peak. In the following we will refer to all such structures as ''plateaus'' for simplicity.

In this Rapid Communication we study the two simplest collision systems that show the discussed behavior, the $2p$ excitation in hydrogen by protons and by $\alpha$ particles. These two collisional systems are sufficiently different for showing our point, that common features may be found in widely different collision systems.

We have chosen to demonstrate the mechanism on the minimal basis-set calculations, since they show the features of interest, while they remain easy to analyze and perform. Calculations with large basis sets are used to show that inclusion of many more states does not change the basic features of interest, i.e., the occurrence of the plateaus.

In Fig. 1 the cross sections for $2p$ excitation in $p$-H($1s$) and $\alpha$-H($1s$) collisions obtained from simple eight atomic orbital-state (AO) calculations containing the $n=1,2$ shell on each center are plotted together with representative experimental data and some more precise calculations as well as our CTMC calculations. For both systems the minimal basis AO calculations compare rather well with experiments and with calculations with larger basis sets. In particular, the plateaus around $v=0.6$ a.u. (10 keV/amu) are well reproduced by the eight-state calculations.

Our CTMC calculations shown in Fig. 1 also compare quite well with experiments, except at the lowest energies for proton impact. The oscillations discussed by Schultz *et al.* [6] are, however, seen only for $\alpha$ impact. They are ascribed

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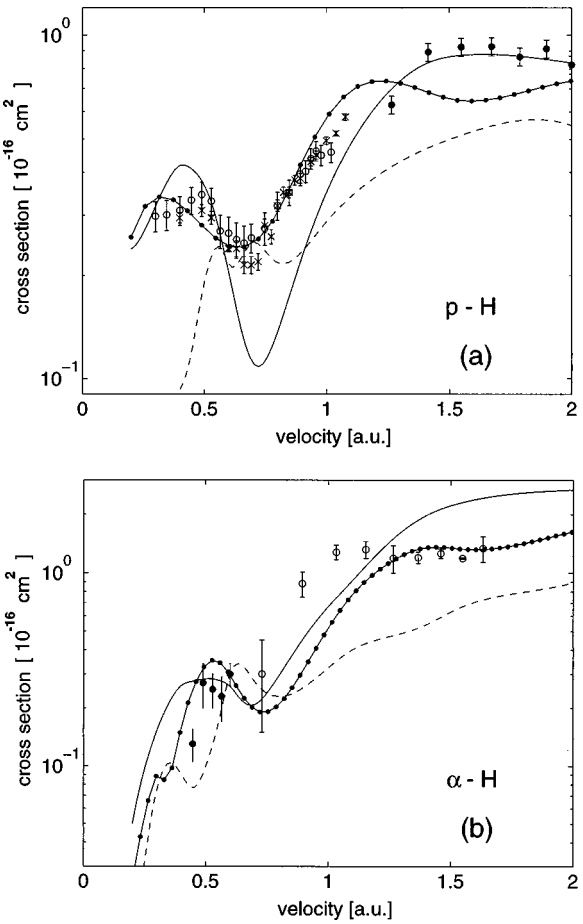


FIG. 1. Cross section for $2p$ excitation in $p$-H (a) and $\alpha$-H (b) collisions. Full line with dots, large-basis two-center AO expansion involving pseudostates for $p$-H [12], and for $\alpha$-H [13]; full line, AO eight-state calculations (present work); dashed line, CTMC calculations (present work). Experimental points: (a) $p$-H: open circles, Morgan *et al.* [14]; crosses, Kondow *et al.* [15]; filled circles, Detleffsen *et al.* [16]; (b) $\alpha$-H: open circles, Hughes *et al.* [17]; filled circles, Higgins *et al.* [18,19].

to two-swap and four-swap mechanisms at 10 and 3 keV/amu (respectively, $v=0.63$ and 0.35 a.u.). However, in our analysis, we have found that in the region around 10 keV/amu the two-swap mechanism contributes only about 50% of the total cross section, and at the direct-process peak around $v=2.83$ a.u., the two-swap mechanism still amounts to about 20% of the cross section.

In the quantal description, the relevant reaction dynamics might be understood by considering the energy curves of the instantaneous collision Hamiltonian, i.e., the correlation diagram of the quasimolecular states. Figure 2 shows a schematic plot of the energy curves obtained from our two-center atomic orbital collision code. The figure displays the eight lowest energy curves following from a diagonalization of the electronic Hamiltonian. For large distances, we see that both the $1s\sigma$ and the $2p\sigma$ states run more or less parallel with the higher states coupling to the separated atom's $n=2$ manifold. Because of the two-center ''promotion'' behavior [8,9] of the $2p\sigma$ orbital, a new region with smaller energy separation is formed at small internuclear distances. The part of the excitation process, which follows the $2p\sigma$ energy curve and couples to the final $2p$ state in this new region, is thus

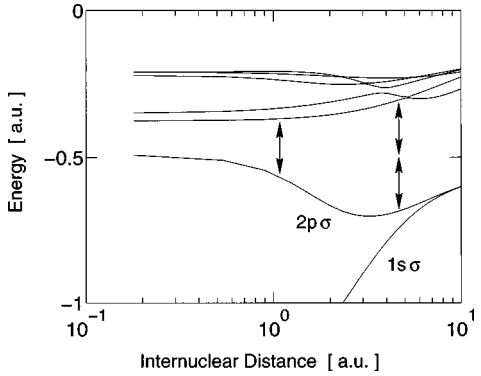


FIG. 2. Schematic plot of the $H_2^+$ energy levels, showing that at large distances the energy difference between $2p\sigma$ and the higher-lying levels is roughly twice as large as the same energy difference at small internuclear distances.

limited by a smaller collision range $R_{\mathrm{coll}}$ as well as smaller energy separation $\Delta E$. In view of the criterion for maximum $s$-$p$ transition probability [10].

$$\frac{\Delta E\, R_{\mathrm{coll}}}{v_{\mathrm{max}}} \approx \pi, \tag{1}$$

it is clear that the two-center process has its maximum at least on the order of $(v_{\mathrm{max}})/2$ below the direct process. Here $v_{\mathrm{max}}$ is the velocity at which the direct excitation process peaks. The parameters $R_{\mathrm{coll}}$ and $\Delta E$ are, however, empirical so the equation only indicates the plateau positions roughly.

Exactly the same argument may be put forward for excitation by multiply charged ions colliding with Na($3s$) [2]. Here, avoided crossings formed by highly excited states of the incoming ion set up a reaction path allowing for strong coupling to the Na($3p$) state *inside* the avoided crossing. Given that, the direct process is strong around and outside the avoided crossing, which is due to the long-range behavior of the related coupling [11]. Thus, the direct excitation and the two-center excitation peak at two different velocity regions also occur here.

The arguments above should be displayed in a plot of the impact-parameter and velocity-dependent probabilities in two distinct domains. At low energies the two-center mechanism dominates, which requires small internuclear separation and, thus, small impact parameters. At higher energies, where the long-range $r^{-2}$ coupling of the $s$-$p$ direct mechanism [11] dominates, larger impact parameters should be the most important.

For an analysis of the computations we use contour maps of the impact parameter and velocity-dependent probabilities of the $2p$ excitation, which are displayed in Fig. 3. The maps are used to support the arguments given above, i.e., they should show two distinct domains of contributions to the cross sections. At low energies the two-center mechanism dominates, which requires small internuclear separation and thus small impact parameters. At higher energies where the long-range $r^{-2}$ coupling of the $s$-$p$ direct mechanism [11] dominates, larger impact parameters should be the most important.

The maps of eight-state computations for $p$-H (upper left) and $\alpha$-H (upper right) collisions both show two distinct domains. At high energies and large impact parameters the dominant direct-process structure [we call it the ''Massey

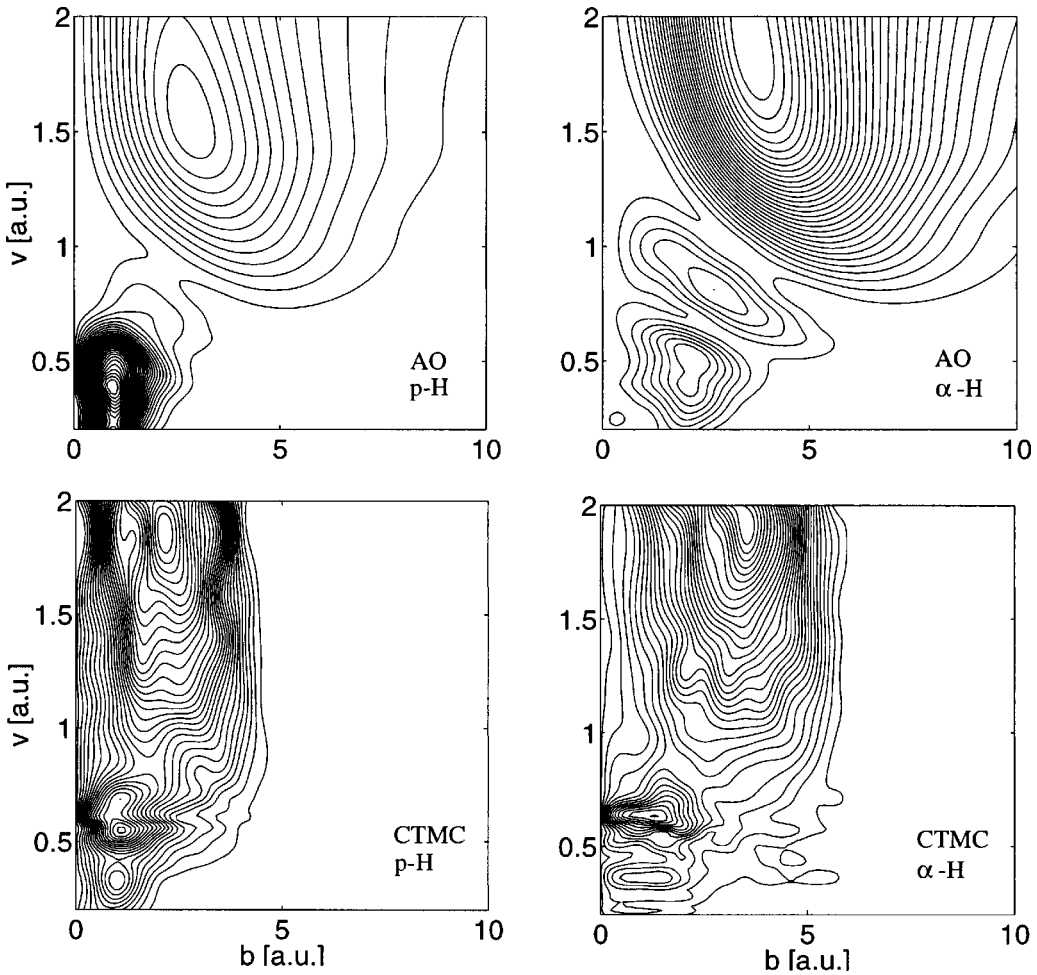


FIG. 3. Contour maps of reduced $2p$ excitation probabilities $P(b)\times b$. The contour interval is linear with 30 contours displayed between zero and the peak value of $P(b)\times b$ given as follows. Upper parts, quantal results; lower parts, CTMC results. Left: $p$-H, $P(b)\times b$; peak value, 0.17 (AO) and 0.12 (CTMC); right: $\alpha$-H, $P(b)\times b$; peak value, 0.21 (AO) and 0.19 (CTMC).

mountain,'' because of Massey criterion, Eq. (1)], peaks at $(v,b)=(1.6,3)$ and (1.8,4), respectively, and a ''two-center region'' at small $(v,b)$. For $p$-H, the two-center region is well separated and contains one single ''mountain.'' for $\alpha$-H, the two-center region is more structured following the projection of the asymptotic atomic wave functions onto the quasimolecular levels.

Corresponding maps of the CTMC calculations are shown in the lower parts of Fig. 3. Surprisingly, the two main regions identified above are roughly present also in the classical calculations, though the details, as well as the underlying mechanisms are different.

The maps are used because they indeed display the common features of the computations, in terms of the differential quantities leading to the total cross sections. They may be a useful tool to also analyze other collision processes.

In conclusion, we have given a simple explanation for the plateaus of excitation in ion-atom collisions. They arise as a crossover between two-center and direct excitation in which each process dominates at different velocity and impact-parameter regions. The nature of the structures can be displayed using contour maps of these contributions to the cross sections. System-specific details may, in some cases, cause the structure to look more like an oscillation, for example, if the two mechanisms are well separated in energy.

The existence of the discussed structures is thus one of the examples where the quantal nature of the electron dynamics manifests itself, and where the classical picture cannot satisfactorily describe the collision processes. The quantal mechanisms involve swaps of the quantal electronic density, well known from two-state calculations, but their description by classical calculations might become realistic only for very large quantum numbers, i.e., in collisions involving Rydberg atoms.

In fact, even for certain such processes (cf., the experimental results of MacAdam *et al.* [7]), one could suggest a quantal interpretation similar to the one discussed here, since also for the large $n$ states the molecular mechanisms may give rise to three different reactions paths, where each path has its peak at a distinct projectile velocity (cf., Fig. 3). This point however, must be considered in detail in a future work.

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