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ON THE UNEXPECTED OSCILLATION OF THE TOTAL CROSS SECTION FOR EXCITATION IN He^{2+} + H COLLISIONS

D. R. Schultz, C. O. Reinhold, and P. S. Krstic
Oak Ridge National Laboratory*
Oak Ridge, TN 37831

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On the unexpected oscillation of the total cross section for excitation in He^{2+} + H collisions

D.R. Schultz, C.O. Reinhold, and P.S. Krstić

Physics Division, Oak Ridge National Laboratory,

Oak Ridge, TN 37831-6373

Abstract

Recent calculations and measurements have revealed unexpected oscillations of the total cross section for excitation in low- to intermediate-energy He^{2+} + H collisions. A physical explanation of this behavior is given here stemming from analysis of classical trajectory Monte Carlo simulations, molecular orbital close coupling calculations, and solution of the time-dependent Schrödinger equation on a numerical lattice. These results indicate that the observed behavior should be characteristic of a wide range of reactions in ion-atom collisions.

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In a recent study [1] of excitation to the n =2 and 3 levels of atomic hydrogen by Be^{4+} impact, utilizing the classical trajectory Monte Carlo (CTMC) method, we found a small local maximum in the cross section centered about a collision energy of approximately 10 keV/u in addition to the well-known, broad peak at a few hundred keV/u. Fritsch [2] had previously found this same behavior using an atomic orbital (AO) close coupling approach, which we found agreed very well with the CTMC result. However, no explanation for the physical mechanism underlying this new peak was evident. Further CTMC calculations for He^{2+}, Be^{4+}, and Ne^{10+} + H have shown that this feature is not unique to Be^{4+} impact, that there are further maxima and minima at still lower energies, and that similar oscillations exist in other channels, including ionization and non-resonant charge transfer.

In fact, both AO [3] and molecular orbital (MO) close coupling [4] calculations confirm the behavior found with the CTMC approach for $n = 2$ excitation in He^{2+} + H (see Figure 1) as does recent experimental measurements [5–7]. Clues to the origin of oscillations seem to be rare (e.g. Fritsch [8] noted that they were obtained only if projectile-centered states were included in the close coupling basis) and contradictory (e.g. Lin [9] has suggested that some may be artifacts of unconverged basis sets in the close coupling expansion). Yet, we find that they are present in quasi-classical (CTMC), quantum mechanical (solution of the time-dependent Schrödinger equation on a numerical lattice in addition to the AO and MO calculations), and experimental results, for at least several ion-atom collision systems. Other similar examples may even be found in textbooks (e.g. for H^+ + H [10]), yet without explanation.

To place this unexpected behavior into context, consider the overall shape of the total cross section as a function of collision velocity (or energy) for any reaction channel such as excitation, ionization, or non-resonant charge transfer in an ion-atom collision. The cross section is zero at or below a velocity corresponding to the reaction threshold energy. It then rises with increasing velocity as energy in excess of the threshold is available to drive the reaction, peaking at some point, and then dropping as velocity is further increased. In general terms, the peak occurs around the point when the velocity of the projectile ion

is close to that characteristic of a target electron, since then the greatest interaction time is available. The corresponding cross section is determined by the internuclear distance at which the potential barrier experienced by the electron drops, due to the projectile's approach, to the point that the reaction is possible classically [11]. As velocity is increased, the interaction time is shortened, lowering the probability of the reaction, and thus the cross section. Resonant charge transfer and elastic scattering, in contrast, plateau for small collision velocities since the reaction can take place with arbitrarily low energy transfer when the interaction time is large, but also fall off rapidly at high energy. These cross sections display narrow oscillations at very low collision velocity due to projectile-target orbital resonances, well separated from those discussed here.

Especially for slow collisions, defined by projectile velocity (v_p) comparable to or smaller than the orbital electronic velocity (v_e), this simple picture must be elaborated. In this regime, the electron or electrons interact strongly with both the target and projectile ions, and lowest-order perturbation theories (e.g. the Born approximation) or other "one-center" theories (so-called due to treating the electron as moving only within the field of one of the heavy particle centers of charge) are inadequate and do not display the present oscillations. Quasi-classical or quantum mechanical approaches are required which account for the dynamics possible in this slow collision regime.

That the oscillations of the excitation, ionization, and non-resonant charge transfer channels appear in the CTMC simulation of the collision gives hope that at least a classical interpretation of the physical mechanism underlying this behavior could be found. By examining individual trajectories, we find that excitation at low energies occurs after the electron first travels between the target and projectile a number of times. This is consistent with the observation by Fritsch that projectile-centered states are required in an AO calculation to obtain the 10 keV/u peak. Furthermore, MacAdam *et al.* [12] have noted a velocity-dependent oscillation in the charge transfer cross section for collisions of ions with Rydberg atoms, and reproduced these oscillations with CTMC calculations. By similar analysis of the trajectories leading to charge transfer, they found that each of the peaks they observed

corresponded to electrons "swapping" a number of times between the target and projectile.

For charge transfer to the quasi-resonant level from initial states of low principal quantum number, we find that oscillations are not present. However, this mechanism is what drives the oscillation of the other channels. That is, at an internuclear separation at which the saddle point of the potential experienced by the target electron is below its ionization potential, the electron can freely travel between the target and projectile centers. For the He^{2+} + H system, this distance is approximately 7.7 a.u. The swapping occurs between the states H(1s) and He^{+}(n=2) which are quasi-resonant.

Obviously at high impact velocity there is insufficient time for a swap to occur and the reaction is direct. At lower velocities (when $v_p < v_e$), one or more swaps can take place. Thus, in our classical picture, the target electron swaps between quasi-resonant states of the two centers once the saddle of the potential drops. Then excitation occurs at some small internuclear distance, turning off the swapping. Furthermore, these two internuclear distances, where the saddle point drops and where excitation occurs (about 1 a.u.), set the distance over which the swapping can occur. If we assume that the orbital velocity is roughly preserved since the states are quasi-resonant, we can determine that the impact velocities for which integral numbers (N) of swaps can occur is given by $v_p = (R_{saddle} - R_{excit})/(2\pi N) = 6.7/(2\pi N)$. For $N = 1 - 6$ the positions predicted by this crude model are indicated in Figure 1 by arrows. The arrow labeled "1/2" indicates the position where the electron is just promoted as it reaches the saddle point, similarly as in Irby's model of ionization [13], which seems to predict well the position of the overall peak of the cross section.

Our independent MO calculations indicate that at velocities below the peak at about 2 keV/u, the oscillations cease since the swapping levels become non-quasi-resonant due to Stark splitting. Since the quasi-classical model of the atom in the CTMC method has an infinite number of levels there always exists a pair of resonant levels. Thus, the CTMC cross section continues to show the succession of peaks extending to even lower energies. Further, our MO calculations show that the peak near 2 keV/u occurs due to strong channels for excitation by rotational coupling. This further emphasizes the distinction between the

classical and quantum models, even though the basic picture is the same in each. One- and two-dimensional lattice solutions of the Schrödinger equation also display oscillations present at the same collision energies, but since rotational coupling is not possible in these models, the promotion is through radial coupling, again emphasizing the basic picture that at certain velocities, the swapping occurs which is then shunted by a promotion at small internuclear distance, leading to a peak in the cross section. These lattice calculations have also been performed for systems where there is no initial quasi-resonant channel and show a strong suppression of the effect at intermediate-energies.

Thus, this work leads to the implication that such oscillations should be present in the low- to intermediate-energy cross sections for many ion-atom pairs. Since we have found that they also are predicted by the CTMC method for ionization and non-resonant charge transfer channels, they should indeed be an unexpected but ubiquitous phenomena.

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REFERENCES

[1] D.R. Schultz, P.S. Krstić, and C.O. Reinhold, Physica Scripta **T62**, 69 (1996).

[2] W. Fritsch, in *Proc. VIth International Conference on the Physics of Highly Charged Ions*, edited by P. Richard *et al.* (American Institute of Physics, New York, 1993) p. 24.

[3] W. Fritsch, R. Shingal, and C.-D. Lin, Phys. Rev. A **44**, 5686 (1991).

[4] L.F. Errea *et al.*, J. Phys. B **27**, 3603 (1994).

[5] D.P. Higgins, J. Geddes, R.W. McCullough, and H.B. Gilbody, Nucl. Instr. Methods Phys. Res. B **103**, 120 (1995).

[6] M.P. Hughes, J. Geddes, and H.B. Gilbody, J. Phys. B **27**, 1143 (1994).

[7] R. Hoekstra and J.P.M. Beijers cited in [3].

[8] W. Fritsch, in *Review of fundamental processes and applications of atoms and ions*, edited by C.-D. Lin (World Scientific, Singapore, 1993) p. 239.

[9] C.-D. Lin, in *Two-center effects in ion-atom collisions*, AIP Conference Proceedings Number 362, edited by T.J. Gay and A.F. Starace (American Institute of Physics, New York, 1996) p. 135; J. Kuang and C.-D. Lin, J. Phys. B **29**, 1207 (1996).

[10] B.H. Bransden and M.R.C. McDowell, *Charge Exchange and the Theory of Ion-Atom Collisions* (Clarendon Press, Oxford, 1992) see e.g. theories and experiments on page 196.

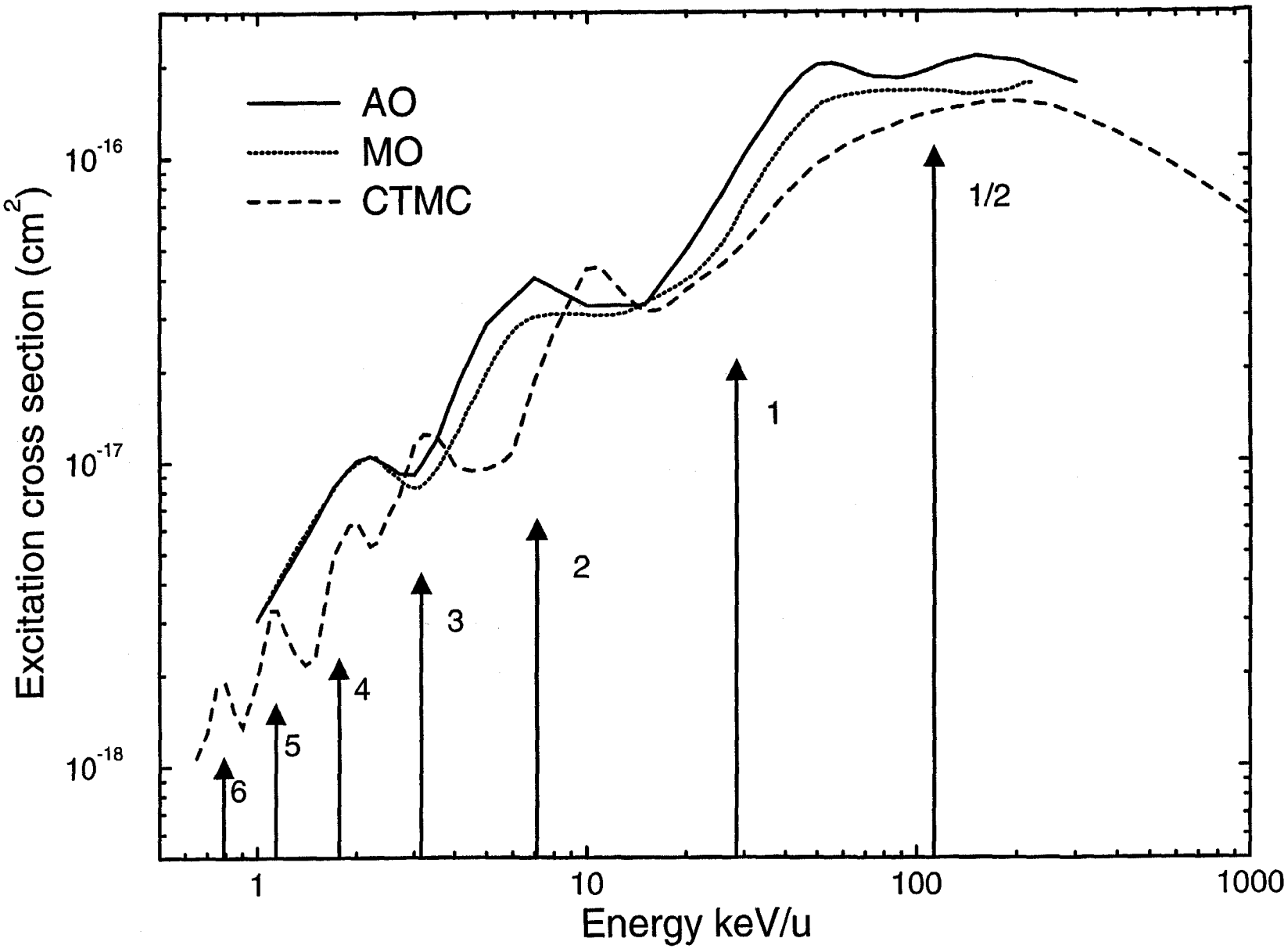
[11] This is the basic idea of the Bohr-Lindhard or classical over-the-barrier model, and that of Irby [13] who generalized this concept considering the motion of the saddle point.

[12] K.B. MacAdam *et al.*, Phys. Rev. Lett. **75**, 1723 (1995).

[13] V.D. Irby, Phys. Rev. A **39**, 54 (1989).

Figure Captions

Figure 1. Excitation to the n=2 level of H by He^{2+} impact. Along with the present CTMC (dashed line) results, AO (solid line, [3]), and MO (dotted line, [4]) calculations are shown. Arrows indicate the peak positions predicted on the basis of the simple timing model described in the text.


AO
MO
CTMC
Excitation cross section (cm^2)
10^{-16}
10^{-17}
10^{-18}
1/2
1
2
3
4
5
6
1
10
100
1000
Energy keV/u