

“Heisenberg Core” in Classical-Trajectory Monte Carlo Calculations of Ionization and Charge Exchange

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We present results of classical-trajectory Monte-Carlo calculations for bare ions on helium atoms. We impose the Heisenberg uncertainty principle on the classical system and obtain two important benefits. Firstly, the classical He atom is stabilized and thus full four-body classical-trajectory Monte Carlo calculations are possible. Secondly, this excludes from the classical phase space those regions where electron binding energies are below the quantum mechanical ground state. As a result, the range of agreement between calculations and experiment is increased considerably towards higher energies.

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Theoretical models for the prediction of ionization and charge exchange in ion-atom collisions have been quite successful in two domains: (1) in very asymmetric fast collisions where the field of the light projectile can be regarded as a perturbation on the target, and (2) in relatively symmetric slow collisions ($v \ll v_e$, with v the collision velocity and v_e the velocity of the relevant electrons), where the process can be represented by the formation of molecular orbitals during the collision.¹ In contrast, for medium-velocity ($v \sim v_e$) cases, existing quantum mechanical methods are difficult to apply,² although recently some results, mostly charge-exchange probabilities by time-dependent Hartree-Fock methods, have been reported.^{3,4} It is exactly in this region that the classical-trajectory Monte Carlo (CTMC) method has had some success. In this method initial conditions are chosen at random from a classical ensemble which resembles as much as possible the true quantum mechanical distribution. The time evolution is then calculated by solution of the classical Hamilton equations.

Calculations performed for the one-electron case (bare ion on hydrogen atom) by Abrines and Percival,⁵ Olson and Salop,⁶ and others, yield ionization and electron-capture cross sections in good agreement with experimental results in a very limited energy region around $v = v_e$. Calculations for multielectron targets have been performed in the independent electron approximation.⁷ Of course, only single-ionization and charge-transfer cross sections can be extracted directly and some reasonable agreement with data is obtained. Exact calculations for two-electron systems were attempted by Pfeifer and Olson⁸ for bare ion + He atom and by Becker and MacKellar⁹ for H + H; however, not all the problems which arose in these cases were solved.

We present here the first full four-body CTMC calculations for a bare ion + He atom over a relatively wide energy region and introduce a correction which should increase the range of validity of CTMC for oth-

er collision systems as well.

It is possible to regard the two electrons of the helium atom in a classical picture as revolving at constant angular velocity at the two ends of the diameter of a circular orbit. This is required in order to obtain equilibrium. However, even such an atom will autoionize when subjected to a small perturbation. The solution to this problem has been suggested by Kirschbaum and Wilets.¹⁰ According to them the quantum mechanical stabilization of the helium atom is essentially due to the Heisenberg uncertainty principle. Therefore, a constraint of the type $r_i p_i \geq \xi$, with the equality sign holding for the ground state, should be added to the classical picture. Here r_i, p_i represent respectively the spatial and momentum coordinates of the i th electron relative to the nucleus. (All physical quantities are given in atomic units.) Since the use of a constraint in the solution of a set of coupled differential equations is not practical, they approximate it by an analytic potential,

$$V(r_i, p_i) = \left(\frac{\xi^2}{4\alpha r_i^2} \right) \exp \left\{ \alpha \left[1 - \left(\frac{r_i p_i}{\xi} \right)^4 \right] \right\}. \quad (1)$$

α is called the hardness parameter. The value of ξ is determined by our requiring $r_0 p_0 = \xi$ for the ground state. From this follows¹⁰

$$r_0 = 4\xi^2/7, \quad p_0 = 7/4\xi. \quad (2)$$

We now set p_0 at the average value obtained from Hartree-Fock calculations: $p_0 = p_{\text{HF}} = 1.392$. This yields from Eq. (2) $\xi = 1.257$ from which, in turn, $r_0 = 0.903$. This is quite close to $r_{\text{HF}} = 0.93$.

We now write the full four-body Hamiltonian for the two nuclei and two electrons and add to it the potential of Eq. (1) for each electron relative to the He nucleus:

$$H = \sum_{k=1}^4 \frac{p_k^2}{2m_k} + \sum_{k=1}^3 \sum_{j=k+1}^4 \frac{q_k q_j}{r_{kj}} + \sum_{i=1}^2 V(r_i, p_i). \quad (3)$$

The next step is the determination of the initial con-

ditions. At present we fix the initial absolute values of r_i and p_i at the above-calculated r_0 and p_0 , respectively. Thus we are left with five initial parameters to be chosen by the Monte Carlo sampling: the three Euler angles of the plane of the circular electron orbit relative to the plane defined by the target nucleus and the initial velocity of the projectile, the initial position of one of the electrons on the orbit (the other is opposite it), and the impact parameter. For each set of initial conditions, the classical Hamilton equations are then integrated. For our four-body case they constitute a set of 24 first-order coupled differential equations. The number of equations could be decreased by 6 by transformation into center-of-mass coordinates; however, it seemed to be difficult to find a transformation for which the benefit of this decrease would not be significantly counteracted by the much more cumbersome form to which the equations transform.

The integration is started with the projectile at a distance of 20 a.u. from the target nucleus. When the nuclei are again about 35 a.u. apart, tests are started to check whether each electron is free or bound to one of the nuclei, or whether additional integration is necessary. Integration is only stopped after two consecutive tests, performed at 5-a.u. intervals, give the same unambiguous results. Further details will be given in a subsequent publication.¹¹

Next, the dependence of the results on the hardness parameter α and indeed on the whole arbitrary shape of V in Eq. (1) has been tested. For a few hundred random sets of initial conditions, the equations were integrated for a whole series of values of α , with V of Eq. (1).

It was found that once α exceeds a certain value, the results do not change at all when it is further increased, until V becomes so steep that the integration program cannot handle it any more. Although the limiting values of α depend on the projectile energy, a typical range where the result is independent of it is $40 < \alpha < 100$.

Other functional shapes for V having, of course, the same features of being practically zero for $p_i r_i > \xi$ and increasing very steeply around $p_i r_i = \xi$ were also tried. Integration with the same initial conditions as before again yielded practically identical results once the potential was steep enough and increased to high enough values. However, the integration was considerably faster when V of Eq. (1) was used and therefore it was adopted for all further calculations.

The first collision system investigated by the method described above was $\text{He}^{++} + \text{He}$. Figure 1 shows a typical trajectory for the four particles in a very small-impact-parameter collision. The oval shape of the initial electron orbit is due to the different scales chosen along the x and z axes. For the same reason the apparent scattering angles are much larger

than the actual ones. In this particular case, the projectile has captured one electron into a rather strongly bound state while the second electron stayed on the target nucleus, in an excited state.

Since Eq. (3) sets the constraints on the electrons only relative to the target nucleus, we calculate, at this stage, only the total single- and double-ionization cross sections for the target atom, σ_1 and σ_2 , disregarding the question of whether the removed electrons are free or have been captured by the projectile. Unfortunately, almost no experimental data for σ_1 and σ_2 are available. We therefore compare our results with the data of Puckett, Taylor, and Martin¹² who measured the apparent target ionization cross section $\sigma_{\text{app}} = \sigma_1 + 2\sigma_2$. Figure 2 displays these data together with our calculated values, for the energy range from 50 to 250 keV/amu. The error bars on the calculated values are due to the statistical character of the CTMC calculation. Agreement between experiment and theory is very good.

Next, the even simpler system $p + \text{He}$ is considered. Here one can expect to calculate also the electron-capture process, using the Hamiltonian of Eq. (3). Figure 3 displays the cross section of the charge-exchange process: $p + \text{He} \rightarrow \text{H} + \text{He}^+$. The two sets of available experimental data are displayed by the filled symbols, while the open circles are the results of

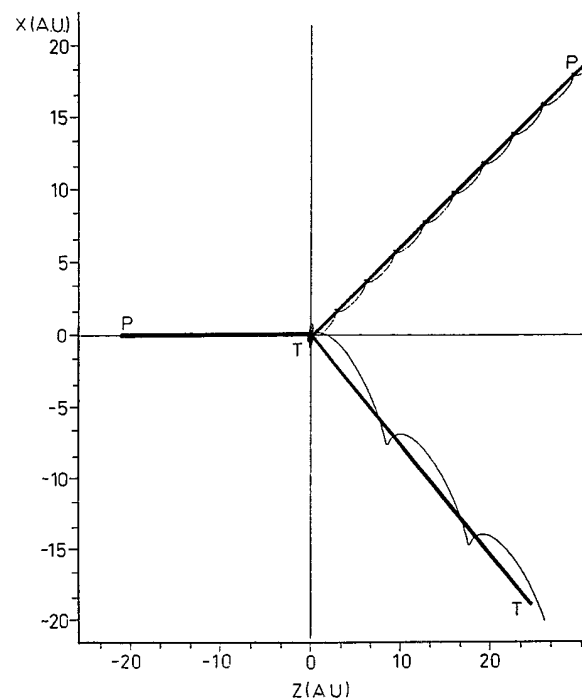


FIG. 1. A typical calculated trajectory for a small-impact-parameter $\text{He}^{++} + \text{He}$ collision. P and T designate the trajectories of the projectile and target nucleus, respectively.

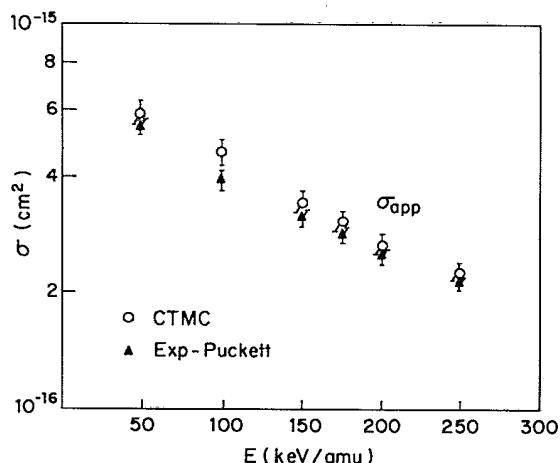


FIG. 2. Experimental data and CTMC calculations for the apparent target ionization cross section in $\text{He}^{++} + \text{He}$ collisions.

the CTMC calculations. The agreement is quite good in the very narrow energy region from about 25 to 50 keV. At higher energies, the calculations overestimate the experimental values by up to a factor of 2. Above 200 keV σ_{ce} becomes so small both for calculations and measurement that large inaccuracies cannot be avoided. Thus, the reasonable agreement between experiment and theory, at the one point at 200 keV, within the limits of relatively large error bars, might be coincidental. The very narrow range of agreement ob-

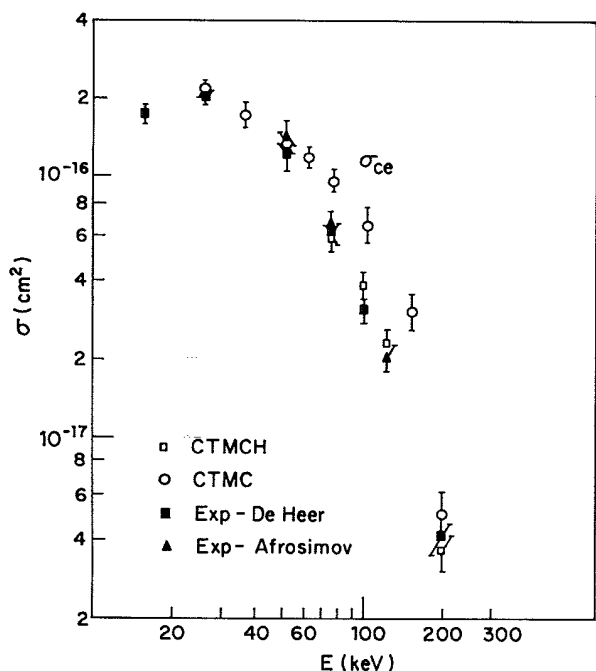


FIG. 3. Experimental data and two sets of calculations for the charge-exchange cross section in the $p + \text{He}$ collision. (For further details see text.)

tained here is similar to the respective range for the same process in the one-electron case: $p + \text{H} \rightarrow \text{H} + p$, as calculated by Olson and Salop⁶ and Cohen.¹³

The reason lies, of course, in the limitations of the correspondence of the classical picture. In trying to overcome some of these severe limitations, we make the following qualitative consideration: It is well known both classically and quantum mechanically that ionic projectiles favor the capture of electrons into orbitals where the electron orbital velocity is comparable to the velocity of the ion. Thus, at energies above 50 keV, the protons tend to capture electrons whose orbital velocities will be considerably larger than 1 a.u. The corresponding binding energies of these classical orbits are below the hydrogen ground state, and thus it is not surprising that at higher energies the CTMC calculations overestimate the capture cross section. Other quantum mechanical aspects seem to be less important. For example, the quantization of binding energies in the actual atom should not affect the results very much at these energies, both because of the above-mentioned velocity-matching effect and because of collision broadening.

Considering a possible inclusion of this restriction into the classical picture, one is brought back to the Heisenberg principle: Just as it holds the helium atom together, it requires a finite-binding-energy ground state for the quantum mechanical system. Thus, the constraint $r_i p_i \geq \xi$, with the equality in the ground state [or its analytical alternative given by V of Eq. (1)], exactly excludes for the electron the region in

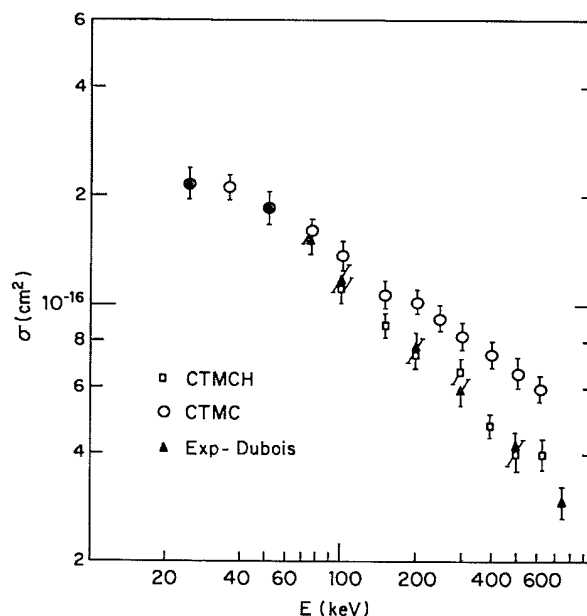


FIG. 4. Experimental data and two sets of calculations for the target single-ionization cross section in the $p + \text{He}$ collision.

phase space which corresponds to binding energies below the quantum mechanical ground state. We therefore designate it as the "Heisenberg core" since its function is similar to the Pauli core which is frequently used to impose the Pauli principle on fermions in semiclassical representations.

Following the above considerations, we repeated the calculation for $p + \text{He}$, adding for each electron an additional potential V of the form of Eq. (1), centered on the proton. For the proton, $\xi = 1$.¹⁰ The open squares in Fig. 3, designated CTMCH, represent the results of these calculations. It is clear that agreement between calculations and experiment is much improved over the whole range of displayed energies: 25 to 200 keV.

Figure 4 displays the total experimental and calculated target single-ionization cross sections for $p + \text{He}$. Again, the open circles designate CTMC calculations, while the squares display results from calculations with the Heisenberg core added on the proton. We see that while the agreement between experiment and CTMC calculations is reasonable from 25 to about 100 keV only, for CTMCH it extends over the whole energy range of 25 to 600 keV. For this system, time-dependent Hartree-Fock calculations were performed by Stich, Lüdde, and Dreizler³ for lower energies (10–40 keV), and their agreement with experiment is also good.

To conclude, we have presented full four-body CTMC calculations for bare ions on He atoms and have shown how to increase considerably their region of validity towards higher energies by introducing the Heisenberg core, without any adjustable parameters. We obviously intend now to return to the even simpler $p + \text{H}$ case and test the effect of adding the Heisenberg core on both protons. It is reasonable to assume that again the region of agreement between calculations

and experiment will increase considerably. After this, additional results such as cross sections for simultaneous electron capture and ionization, impact-parameter-dependent probabilities for all processes, cross sections for free-electron production as a function of their final energy and direction, etc., will readily be available for comparison with experiment.

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