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LETTER TO THE EDITOR

Orientation parameters and dipole moments of $He^+(n=2)$ states in $He^{2+}+H$ collisions: comparison of CTMC and close-coupling results

N Toshima†, R Shingal‡ and C D Lin‡

† Institute of Applied Physics, University of Tsukuba, Tsukuba, Ibaraki, 305, Japan
‡ Department of Physics, Kansas State University, Manhattan, KS 66506, USA

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Abstract. The orientation parameter and the dipole moment of the $n=2$ states of He^+ resulting from electron capture in $He^{2+}+H$ collisions are examined using the classical trajectory Monte Carlo (CTMC) method and the close-coupling expansion using two-centre atomic orbitals at 10, 25 and 50 keV amu^{-1}. It is shown that the orientation parameters calculated from the two theories are in good agreement but large discrepancies exist for the dipole moments. Together with previous similar comparisons for p-H collisions, we conclude that the CTMC method is not reliable in predicting the coherence parameters of excited states formed in atomic collisions.

In a recent paper, Toshima and Lin (1991) examined the coherence parameters of excited states formed in collisions of hydrogen atoms with electrons, positrons, protons and antiprotons using the classical trajectory Monte Carlo (CTMC) method and compared the results with those obtained from quantal calculations. The excited states examined were the $n=2$ states formed by electron capture or by excitation processes. In particular, the orientation parameter $\langle L_y \rangle$, which is defined to be the average of the electronic angular momentum perpendicular to the scattering plane, and the dipole moments, $\langle D_x \rangle$ and $\langle D_z \rangle$, on the xz collision plane, were calculated and compared. It was found that the classical and the quantal calculations are in good agreement in some cases but not in others.

The results from this previous work appear to indicate that the CTMC method, despite its wide applications in ion–atom collision, does not provide reliable predictions for the finer details that are readily calculable from quantal theory. However, in the collision systems studied in that paper, the excitation and capture probabilities are quite small, usually less than a few per cent. It is not clear whether the discrepancy is not partly due to the statistical nature of the CTMC which is less reliable for processes where the transition probabilities are small. Before one can conclude the deficiency of the CTMC for predicting these coherence parameters, it is desirable to make comparison for processes where the transition probabilities are large. In this letter, we report the orientation parameters and the dipole moments for the excited $n=2$ states of He^+ resulting from collisions of He^{2+} on H at laboratory energies of 10, 25 and 50 keV amu^{-1}. The electron capture probabilities are quite large at these energies. We again compare the CTMC results with those obtained from the close-coupling calculations using the two-centre travelling atomic orbital expansion method. We find that the orientation parameters obtained from the two calculations are in good agreement, but large deviations occur for the dipole moments.

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The geometry is defined such that the beam direction is the $+z$ direction and that the incident particle always enters on the $+x$ side of the target. This defines the xz scattering plane and the y is in the direction of $z \times x$. According to this definition, the quantal expression for the orientation parameter is $\langle L_y \rangle = 2\sqrt{2}\,\mathrm{Im}(a_{2p_0} a^*_{2p_1})$ and the two components of the dipole moments are $\langle D_x \rangle = -3\sqrt{2}\,\mathrm{Re}(a_{2s} a^*_{2p_1})$ and $\langle D_z \rangle = 3\,\mathrm{Re}(a_{2s} a^*_{2p_0})$ for $He^+(n=2)$ states. The electron capture amplitudes a_{2s}, a_{2p_0} and a_{2p_1} to the 2s, $2p_0$ and $2p_1$ states of He^+, respectively, are calculated from the two-centre close-coupling method using two-centre atomic orbitals (see Fritsch and Lin 1991).

In the CTMC calculations, the electron is defined to have a principal quantum number n if its energy E (in au) lies between $-Z^2/(n-0.5)^2 < 2E < -Z^2/(n+0.5)^2$. In the present case, $Z=2$. The definition of the orbital angular momentum l and its projection along the y direction is not unique in the classical theory, as to be addressed below. One more ambiguity about the CTMC is the choice of ensemble average for the initial state. In the previous study (Toshima and Lin 1991) only the microcanonical ensemble was considered since excitation processes were examined there and it is desirable to have a well defined energy for the initial state. In the present paper, we consider the electron capture channels only. Thus it is also possible to carry out CTMC calculations using the ensemble adopted by Hardie and Olson (1983) (see also Cohen 1985). Their ensemble was chosen to give a better description of the spatial distribution of the electron density in the initial atomic hydrogen, but at the price that the initial state is a weighted collection of states with different binding energies. We note that in the standard microcanonical ensemble the initial atomic hydrogen has a binding energy of -0.5 au. It gives the correct description of the momentum distribution of the electron but not the spatial distribution.

The results of the close-coupling calculations are compared to the CTMC results in figures 1 and 2. In the close-coupling calculation, the basis set includes the $n = 1$–3 He^+ states and the 1s of the target, plus additional pseudostates on both centres (see Fritsch *et al* 1991 for the list of the basis functions). The electron capture probabilities to 2s and 2p states at 10, 25 and 50 keV amu^{-1} are shown along the left-hand column of figure 1. The CTMC results were calculated using the microcanonical ensemble and we note that they are in reasonable agreement with the close-coupling results. In the present CTMC calculation, 100 000 trajectories were used for each impact parameter. Since the capture probability is of the order of 0.1 or more, the statistical error associated with the present CTMC calculation is only of the order of 1% or less. The good agreement at large impact parameters is actually surprising since the outer part of the electron density distribution in the microcanonical ensemble and the quantal descriptions are quite different. We mention that the capture probabilities calculated using the ensemble of Hardie and Olson (not shown) are in poorer agreement with the close-coupling results.

We next compare the orientation parameter for the 2p states obtained from these calculations. Since angular momentum is not quantized in classical physics, an electron is assigned to have orbital angular momentum l if its classical value is in the range of l to $l+1$. In calculating L_y using the CTMC, one can sample all the $n=2$ states disregarding the value of l, or one can sample only the $l=1$ subset of the $n=2$ states. Along the right-hand column of figure 1 we compare the close-coupling results which are shown with full curves with those from three different CTMC calculations. The full and the open circles are from the microcanonical ensemble where the former sample all the $n=2$ states and the latter only the $l=1$ subset. The squares are from the model of Hardie and Olson and sample all the $n=2$ states. Except for the large impact

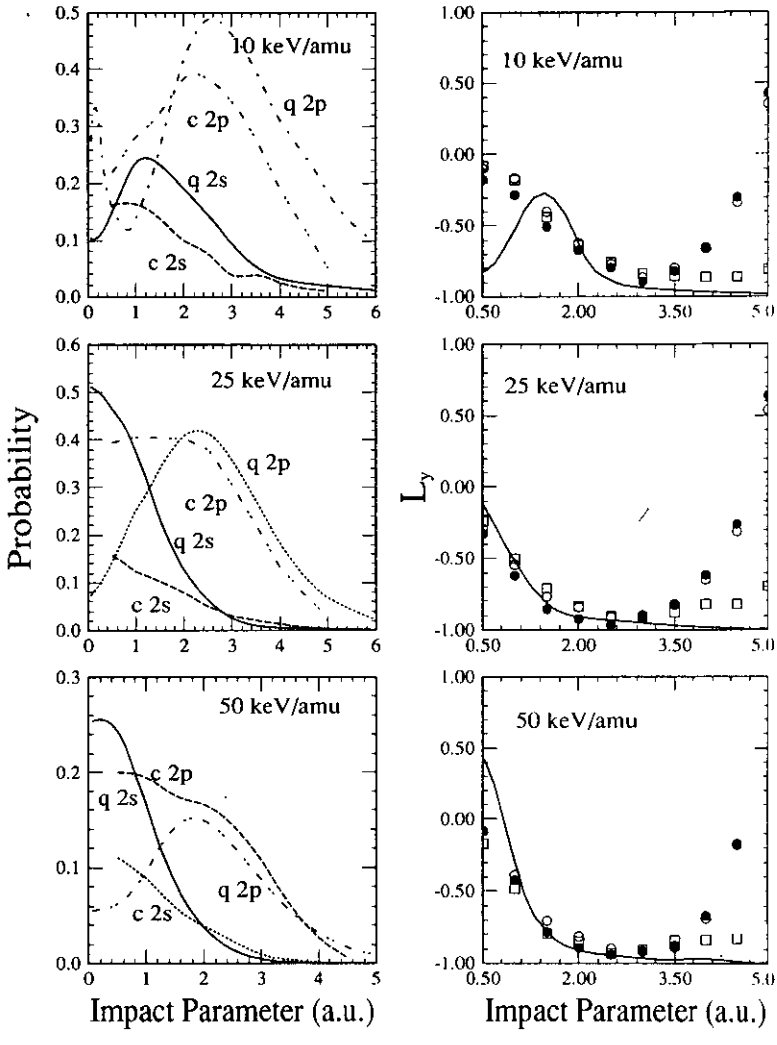

Figure 1. (*a*) Probabilities plotted against impact parameters for electron capture to 2s and 2p states for $He^{2+}+H(1s)$ collisions at 10, 25 and 50 keV amu^{-1} calculated using the close-coupling expansion method with two-centre atomic orbitals, labelled q, and the classical trajectory Monte Carlo (CTMC), labelled c. (*b*) Calculated orientation parameter for 2p states for the present system. The full curves are from the close-coupling calculations. The symbols are from the different versions of the CTMC method (see text).

parameters where the statistics is not good, we note that the close-coupling results and the CTMC results are in good agreement. Furthermore the CTMC results are not sensitive to the ensemble sampling. The orientation parameter tends to -1.0 which is in agreement with the propensity rule of Nielsen *et al* (1990).

We next consider $\langle D_z \rangle$ and $\langle D_x \rangle$ at the same three energies. The results from the two CTMC calculations and from the close-coupling calculations are shown in figure 2. First note that there is large difference between the two CTMC results where the full symbols are from the microcanonical ensemble and the open symbols from the model of Hardie and Olson. Both CTMC results disagree with those from the close-coupling

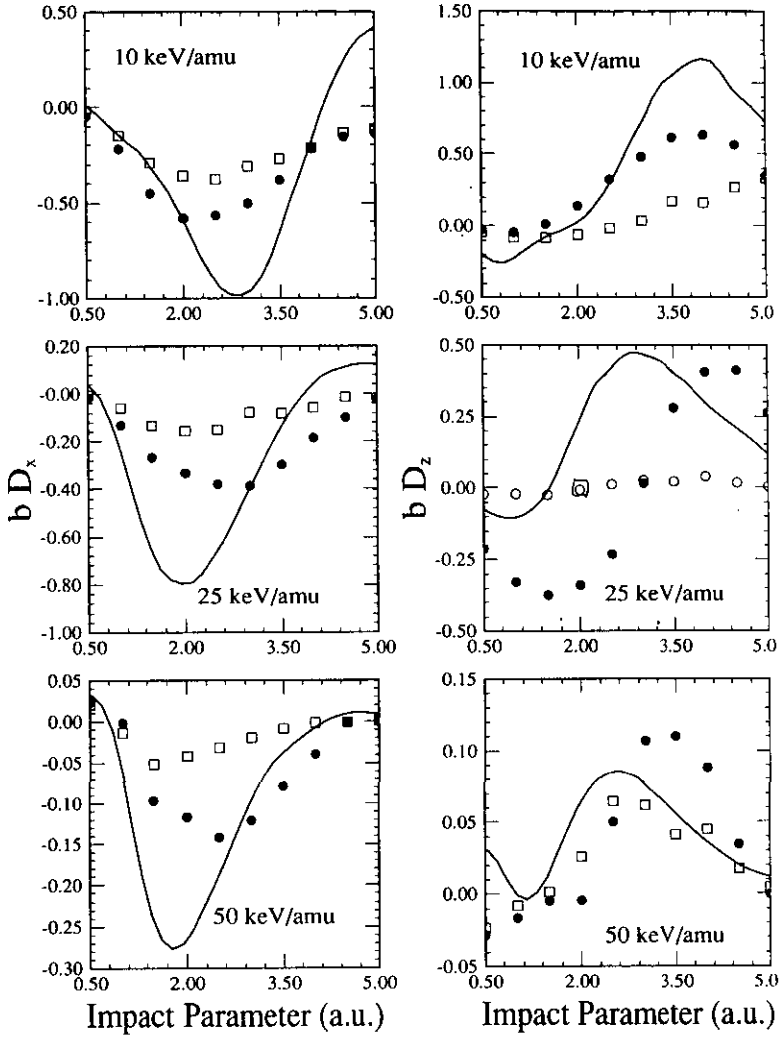

Figure 2. Impact parameter-weighted dipole moments along the x and the z directions for the $N = 2$ states of He^+ from He^{2+}-H collisions at 10, 25 and 50 keV amu^{-1}. The full curves are from the close-coupling calculations, the full circles are from the CTMC calculations using the microcanonical ensemble and the open squares are from the CTMC calculations using the ensemble of Hardie and Olson.

calculations. Based on these results, we conclude that the CTMC method fails to predict the dipole moments of the present collision system.

The different parameters shown in figures 1 and 2, whether close coupling or CTMC, were obtained from a single calculation. Thus in evaluating the validity of the CTMC method, all of these coherence parameters should be compared simultaneously. From the results in figures 1 and 2, it is clear that there exist discrepancies between the CTMC and the close-coupling predictions for the coherence parameters even when the transition probabilities are large. The difference is thus not due to the statistical average of the CTMC method, but rather to the inability of classical mechanics in describing quantal systems in detail. Together with the results from the systems studied by Toshima

and Lin (1991) earlier, we conclude that the CTMC method is incapable of predicting the coherence parameters of excited states formed in atomic collisions.

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