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LETTER TO THE EDITOR

Double electron transitions in collisions between multiply charged ions and helium atoms

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Abstract. A semiclassical model for He is applied to the study of multiply charged ions colliding with He at intermediate energies. Single and double electron capture and ionisation and capture-ionisation reactions are studied for incident-ion charge states $q = +1$ to $+50$ at 1 MeV amu^{-1}. For charge states $q \geqslant +10$, single electron capture is found to be dominated by a two-electron transition in which one electron is captured and the other is ionised.

The study of multiple electron capture or ionisation in collisions of multiply charged ions with many-electron target atoms is a difficult n-body problem with $n \geqslant 4$. The simplest such system involves fully stripped ions and He atoms where the following charge exchange (CEX) and ionisation (ION) reactions are possible:

$$A^{q+} + He \rightarrow \begin{cases} A^{q+} + He^{+} + e & \text{1-ION} \quad (1) \\ A^{q+} + He^{2+} + 2e & \text{2-ION} \quad (2) \\ A^{(q-1)+} + He^{+} & \text{1-CEX} \quad (3) \\ A^{(q-1)+} + He^{2+} + e & \text{CEX-ION} \quad (4) \\ A^{(q-2)+} + He^{2+} & \text{2-CEX.} \quad (5) \end{cases}$$

For collisions at intermediate energies, $200 \leqslant E \leqslant 2000$ keV amu^{-1}, it has been customary (Olson 1979, Willis *et al* 1985, Horbatsch 1986) to treat the above reactions within the independent-electron model (IEM) (Hansteen and Mosebekk 1972, McGuire and Weaver 1977). This model has recently been advanced to incorporate the Fermi statistics of the electrons (Becker *et al* 1984). In the IEM treatment, one simulates an electron shell with a single-electron model, and then computes the branching to the various channels using statistics based on a binomial distribution. To gain a more thorough understanding of the collision mechanisms responsible for reactions (1)-(5), we have applied a semiclassical model for the He atom which includes both electrons, to allow us to follow explicitly both the single and double electron transitions. The semiclassical description of the He atom allows the use of the classical-trajectory Monte Carlo (CTMC) method, which has demonstrated its validity in intermediate-energy collisions between multiply charged ions and atomic hydrogen (three-body processes) (Olson and Salop 1977, Olson *et al* 1978).

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In earlier work, we have performed four-body CTMC calculations (Pfeifer and Olson 1982) based on the Bohr model for the He atom with the use of restoring potentials (Kirschbaum and Wilets 1980) to enforce the Heisenberg uncertainty principle; other workers have recently expanded upon this procedure (Zajfman and Maor 1986). The semiclassical model presented here has several advantages over the Bohr and IEM models used previously, including an accurate electron momentum distribution and an allowance for the angular momenta of the electrons. Calculations based on the IEM employ a constant ionisation potential within an electron shell. For electron removal from inner shells of medium-Z elements, the ionisation energies are relatively close to one another and such an approximation is valid. However, outer-shell processes, where the sequential ionisation energies increase rapidly, severely strain the validity of the single-electron approximation and require the development of new theoretical methods.

Semiclassical quantisation rules have been given by Einstein, Brillouin and Keller (Keller 1958). Action integrals of the electron orbits are evaluated in terms of the conjugate momenta p_i and coordinates q_i:

$$I_j = (2\pi)^{-1} \oint p_i \, \mathrm{d}q_i \tag{6}$$

where

$$\begin{aligned} I_{ml} &= I_\phi \\ I_l &= I_\theta + I_\phi \\ I_n &= I_r + I_\theta + I_\phi. \end{aligned} \tag{7}$$

Applying the semiclassical quantisation rules, one finds

$$I_j = (v_j + \tfrac{1}{4}\alpha)\hbar \tag{8}$$

where the v_j are the n, l and m_l quantum numbers and $\alpha = 2$ for an oscillation integral (Maslov 1972). If one employs a self-consistent-field model for the He atom, where each electron experiences an averaged potential due to the nucleus and the other electron, one can show directly (Leopold and Percival 1980, Coveney and Child 1984) that the half-integer quantum numbers dictated by equation (8) require that the planes of the electron orbits are offset by 120° from one another and that the eccentricities of the orbits are equal to 0.866. Therefore, to some extent, the semiclassical model imparts correlation in the electron distribution of the He atom, since the electrons are primarily found on the outer portions of their elliptical orbits where their speed is lower.

Within the context of a self-consistent-field model, the orbit of each electron is hydrogenic. The problem of autoionisation in a classical description of a many-electron atom is therefore removed owing to the lack of the r_{ij}^{-1} electron–electron repulsive potential. Recalling the quantum mechanical two-term spatial wavefunction for the He atom,

$$\Psi = N[\exp(-\alpha r_1)\exp(-\beta r_2) + \exp(-\alpha r_2)\exp(-\beta r_1)] \tag{9}$$

it is possible to relate classically the parameters α and β to the effective charge experienced by each electron. Using Slater's rules, $\alpha = \beta = 1.6875$, and the total electronic energy calculated (Clementi and Roetti 1974) for He is -2.848 au. The experimental value is -2.903 au. However, such values for α and β classically require each electron to be bound by 1.424 au ($\alpha^2/2$ and $\beta^2/2$), which is unacceptable for collision studies because the experimental first and second ionisation potentials are

0.903 and 2.000 au, respectively. Since classical electrons are distinguishable, it is possible to use effective charges of $\alpha = 1.344$ and $\beta = 2.000$ for the two electrons. With these values, the experimental ionisation potentials are reproduced, and quantum mechanical calculations using the split-shell wavefunction given by equation (9) yield an electronic energy of -2.866 au, one third closer to the experimental value than the single-parameter result. We also find that the electronic momentum distribution for the He atom with the two electrons in their classical hydrogenic orbits very closely reproduces recent extensive quantum mechanical calculations (Regier and Thakkar 1985); see figure 1. The latter comparison is crucial for the application of the classical model to scattering calculations where an accurate description of the momentum transfer is essential (Percival and Richards 1975).

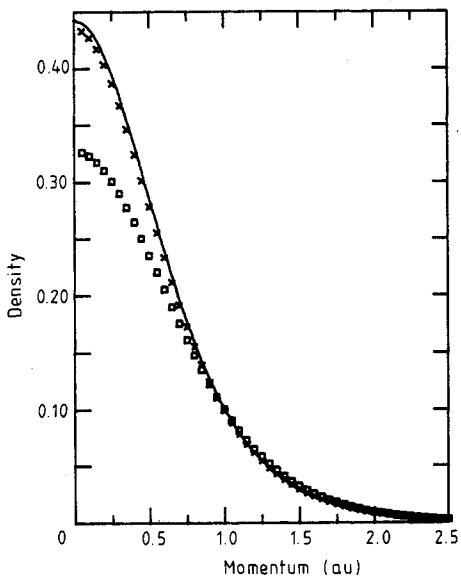

Figure 1. Electron momentum density for the He atom. Full curve, 40-term Gaussian geminal calculation (Regier and Thakkar 1985); ■, classical calculations for effective charges $\alpha = \beta = 1.6875$; ×, $\alpha = 1.344$ and $\beta = 2.000$.

Calculated cross sections obtained from the application of the split-shell semi-classical He model in the CTMC method are shown for 1000 keV amu^{-1} in figure 2. As expected, the dominant collisional process at this energy is single ionisation. The calculated single ionisation cross sections agree well with experimental data (Hvelplund *et al* 1980, Shah and Gilbody 1985) that differentiate between single ionisation, reaction (1), and 'gross' ionisation, in which only the total charge produced in the collision region is measured. The calculated double ionisation (reaction (2)) cross sections for the higher charge states are also in accord with the available data.

Of special interest, however, are the single and double electron capture processes, reactions (3)-(5). At 1000 keV amu^{-1} data are available on double electron capture from studies on the O^{8+} + He (MacDonald and Martin 1971, Hippler *et al* 1985) and S^{13+} + He (Tanis *et al* 1986) systems. Good agreement between theory and experiment is obtained for the S^{13+} system, while for the O^{8+} system the calculations lie between the two experimental studies. The calculations assume that the product ion will stabilise by radiative decay rather than through the Auger process. Such an assumption becomes

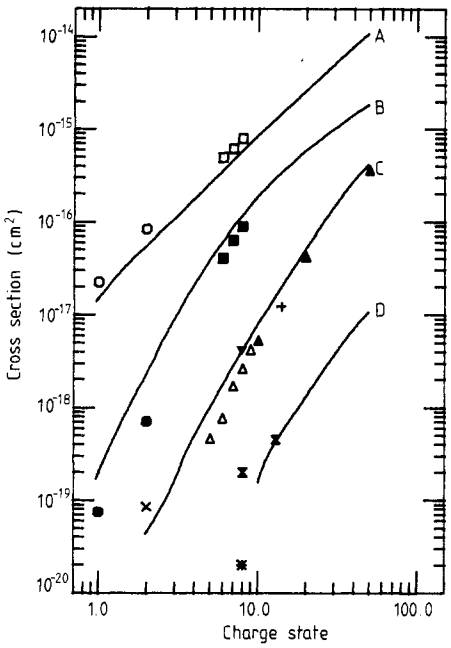

Figure 2. Experimental and theoretical cross sections at 1 MeV amu^{-1} on an He target. The full curves represent the calculated results for: A, single ionisation (reaction (1)); B, double ionisation (reaction (2)); C, single electron capture (reactions (3) and (4)); D, double electron capture (reaction (5)). The experimental data are as follows. Single ionisation: ○, Shah and Gilbody (1985); □, Hvelplund *et al* (1980). Double ionisation: ●, Shah and Gilbody (1985); ■, Hvelplund *et al* (1980). Single electron capture: ×, Hvelplund *et al* (1976); △, Guffey *et al* (1977); ▼, Knudsen *et al* (1981); ▲, Schlachter *et al* (1983); +, Schiebel *et al* (1977). Double electron capture: ⧖, MacDonald and Martin (1971); ⧗, Tanis *et al* (1986); *, Hippler *et al* (1985).

increasingly valid for $q > +10$. For low charge states, our calculations should lie above the experimental results.

For the single electron capture reaction, we have presented the results of the Schlachter *et al* (1983) scaling rule for comparison with high-charge-state ions, $q \geq +10$. The agreement between theoretical and experimental results is exceedingly good. For lower-charge-state ions the calculations are in reasonable accord with the existing data for highly stripped ions (Schiebel *et al* 1977, Knudsen *et al* 1981, Hvelplund *et al* 1976, Guffey *et al* 1977). It is significant that we find that single electron capture is complicated by a double electron transition, where one electron is ionised and the other is captured. At 1000 keV amu^{-1} and for charge states $q \geq +10$, more than 80% of the single electron capture is mediated by the ionisation process, reaction (4). At lower energies, experimental measurements also indicate the increasing importance of capture-ionisation in the single electron capture reaction. At 400 keV amu^{-1}, Shah and Gilbody (1985) find that the component of capture-ionisation in the single electron capture process rises from 3% for H^+ to 12% for He^{2+}, and then to 28% for Li^{3+}, for collisions on He. At 100 keV amu^{-1}, Damsgaard *et al* (1983) observe a similar trend in Au^{q+} + He collisions.

The single ionisation cross sections scale roughly as q^2 while the double ionisation values increase as q^3 for $q \leq +10$, followed by a lower dependence, roughly q^2, for

higher charge states. The charge state dependence of the double ionisation cross section is not in agreement with what one would naively predict using the Born approximation. Within the Born approximation, the double electron transition probabilities are proportional to the square of the single electron transition probabilities. Hence the predicted charge state dependence for the double ionisation cross section is q^4, in contrast to our results.

Both the single and double electron capture cross sections follow a q^3 dependence. This similarity in the charge state dependences is unexpected and has not been predicted by previous theoretical studies. An analysis of the calculated trajectories leading to capture-ionisation indicates that the ionised electrons scattered to small angles have velocities close to that of the projectile ion. This observation is consistent with the measurements of Andersen *et al* (1984) on 100 keV amu^{-1}Au^{15+} + He collisions. Double electron capture is a subset of these collisions where both electrons transfer to the rest frame of the moving projectile ion and are left in bound levels. We find that the partitioning between capture-ionisation and double electron capture is approximately constant, so similar dependences of the cross sections on the projectile-ion charge state are predicted. The double ionisation reaction contains the above components plus a large direct-impact ionisation contribution that causes its charge state dependence to differ from the capture processes.

Our collision studies for multiply charged ions impinging on He indicate reasonable agreement with existing experimental data for the electron removal reactions. However, data are sparse since coincidence measurements are required to deduce the cross sections for reactions (1)-(4) directly. An unexpected result of the calculations was that a large fraction of the single electron capture process for high-charge-state ions, $q \geq +10$, is due to a double electron transition.

Studies of the impact parameter dependence of the transition probabilities indicate there is almost unit probability of removing one or both of the electrons for small impact parameters, $b \leq 1.0\ a_0$ ($q = +5$) and $b \leq 10.0\ a_0$ ($q = +50$). At these impact parameters, the double electron processes are found to be comparable with the single electron removal processes. Our calculations therefore indicate that the eventual quantum mechanical treatment of this problem cannot be based on perturbation approximations and must have enough flexibility in the basis set and coupling terms to accurately describe both the discrete and continuum channels.

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References

Andersen L H, Frost M, Hvelplund P and Knudsen H 1984 *Phys. Rev. Lett.* **52** 518-21
Becker R L, Ford A L and Reading J F 1984 *Phys. Rev.* A **29** 3111-21
Clementi E and Roetti C 1974 *At. Data Nucl. Data Tables* **14** 177-478
Coveney P V and Child M S 1984 *J. Phys. B: At. Mol. Phys.* **17** 319-32
Damsgaard H, Haugen H K, Hvelplund P and Knudsen H 1983 *Phys. Rev.* A **27** 112-6
Guffey J A, Ellsworth L D and MacDonald J R 1977 *Phys. Rev.* A **15** 1863-70
Hansteen J M and Mosebekk O P 1972 *Phys. Rev. Lett.* **29** 1361-2
Hippler R, Datz S, Miller P D and Pepmiller P L 1985 *Proc. 14th Int. Conf. on Physics of Electronic and Atomic Collisions (Palo Alto) 1985* Abstracts p 505
Horbatsch M 1986 *J. Phys. B: At. Mol. Phys.* **19** L193-8
Hvelplund P, Haugen H K and Knudsen H 1980 *Phys. Rev.* A **22** 1930-40

Hvelplund P, Heinemeier J, Horsdal-Pedersen E and Simpson F K 1976 *J. Phys. B: At. Mol. Phys.* **9** 491-6
Keller J B 1958 *Ann. Phys., NY* **4** 180-8
Kirschbaum C L and Wilets L 1980 *Phys. Rev.* A **21** 834-41
Knudsen H, Haugen H K and Hvelplund P 1981 *Phys. Rev.* A **23** 597-610
Leopold J G and Percival I C 1980 *J. Phys. B: At. Mol. Phys.* **13** 1037-47
MacDonald J R and Martin F W 1971 *Phys. Rev.* A **4** 1965-74
McGuire J H and Weaver L 1977 *Phys. Rev.* A **16** 41-7
Maslov V 1972 *Theorie des Perturbations* (Paris: Dunod) part II, ch 2
Olson R E 1979 *J. Phys. B: At. Mol. Phys.* **12** 1843-9
Olson R E Berkner K H, Graham W G, Pyle R V, Schlachter A S and Stearns J W 1978 *Phys. Rev. Lett.* **41** 163-6
Olson R E and Salop A 1977 *Phys. Rev.* A **16** 531-41
Percival I C and Richards D 1975 *Advances in Atomic and Molecular Physics* vol 11, ed D R Bates and B Bederson (New York: Academic) pp 1-82
Pfeifer S J and Olson R E 1982 *Phys. Lett.* **92A** 175-8
Regier P E and Thakkar A J 1985 *J. Phys. B: At. Mol. Phys.* **18** 3061-71
Schiebel U, Doyle B L, MacDonald J R and Ellsworth L D 1977 *Phys. Rev.* A **16** 1089-92
Schlachter A S, Stearns J W, Graham W G, Berkner K H, Pyle R V and Tanis J A 1983 *Phys. Rev.* A **27** 3372-4
Shah M B and Gilbody H B 1985 *J. Phys. B: At. Mol. Phys.* **18** 899-913
Tanis J A *et al* 1986 *Bull. Am. Phys. Soc.* **31** 955
Willis S L, Peach G, McDowell M R C and Banerji J 1985 *J. Phys. B: At. Mol. Phys.* **18** 3939-50
Zajfman D and Maor D 1986 *Phys. Rev. Lett.* **56** 320-3