J. Phys. B: At. Mol. Opt. Phys. 25 (1992) 5431-5437. Printed in the UK

Inclusive probability calculations for the K-vacancy transfer in collisions of S^{15+} on Ar

P Kürpick, W-D Sepp and B Fricke

Fachbereich Physik, Universität Kassel, Heinrich-Plett Str. 40, 3500 Kassel, Federal Republic of Germany

Received 15 May 1992, in final form 7 September 1992

Abstract. Using the single-particle amplitudes from a 20-level coupled-channel calculation with *ab initio* relativistic self consistent LCAO-MO Dirac-Fock-Slater energy eigenvalues and matrix elements we calculate within the frame of the inclusive probability formalism impact-parameter-dependent K-hole transfer probabilities. As an example we show results for the heavy asymmetric collision system S^{15+} on Ar for impact energies from 4.7 to 16 MeV. The inclusive probability formalism which reinstates the many-particle aspect of the collision system permits a qualitative and quantitative agreement with the experiment which is not achieved by the single-particle picture.

1. Introduction

During the last decade intensive experimental work has been done to study K-vacancy transfer in heavy asymmetric collision systems. Both double and triple coincidence techniques were applied to provide impact-parameter-dependent probabilities for many-particle states after the collision [1-4]. Measurements have been carried out over a wide range of projectile energies from 10 keV u^{-1} up to 3 MeV u^{-1} and numerous experimental $P(b)$ curves are available to test the different theoretical approaches. In this paper we take up the case of hydrogen-like S^{15+} ions colliding with Ar where both experimental and theoretical impact-parameter-dependent probabilities for Ar K vacancy are available [3]. The probability of finding at least one Ar K vacancy in the Ar target after the collision was measured for projectile energies from 4.7 to 90 MeV. Using an *ab initio* relativistic self-consistent LCAO-MO Dirac-Fock-Slater description of the collision problem we focus on the low energy range from 4.7 to 16 MeV S^{15+} on Ar. To take into account the many-particle aspect of the collision system the experimental question is answered by using the inclusive probability formalism. Starting from the single-particle amplitudes we compute the inclusive probability to find at least one Ar K vacancy after the collision and we get an improved agreement compared to simple theories as discussed below.

2. Method

Starting from the semiclassical approximation we need to solve the time-dependent many-particle Dirac equation for the electrons involved in the collision system

$$\hat{H}_{\mathrm{el}}(\boldsymbol{R}(t))\Psi_{\mathrm{el}}(\boldsymbol{r}_1,\ldots,\boldsymbol{r}_N,t)=\mathrm{i}\hbar\frac{\partial}{\partial t}\Psi_{\mathrm{el}}(\boldsymbol{r}_1,\ldots,\boldsymbol{r}_N,t). \tag{1}$$

The classical trajectory of the nuclei can either be given by a model potential (like the Coulomb or Bohr potential) or a SCF potential obtained by solving the static Dirac–Fock–Slater equation. We use an effective many-particle Hamiltonian given as a sum of single-particle Hamiltonians

$$\hat{H}_{\mathrm{el}}^{\mathrm{eff}} = \sum_{i=1}^{N} \hat{h}_i^{\mathrm{DFS}}.$$

Then we need to solve a set of time-dependent single-particle equations

$$\left(\hat{h}_i^{\mathrm{DFS}} - \mathrm{i}\hbar\frac{\partial}{\partial t}\right)\psi_i(t) = 0 \qquad \text{with } i = 1, \ldots, N. \tag{2}$$

The wavefunctions $\psi_i(t)$ must satisfy the initial conditions for the N electrons

$$\lim_{t\to-\infty} (\psi_i(t) - \psi_i^0(t)) = 0 \qquad \text{with } i = 1, \ldots, N. \tag{3}$$

The solution of the many-particle equation (1) is given as a determinant built-up from the single-particle wavefunctions $\psi_i(t)$

$$\Psi_{\mathrm{el}}(\boldsymbol{r}, t) = \frac{1}{\sqrt{N!}}\begin{vmatrix} \psi_1(1) & \cdots & \psi_N(1) \\ \vdots & & \vdots \\ \psi_1(N) & \cdots & \psi_N(N) \end{vmatrix}.$$

We use a basis-set method to solve equation (2). The time-dependent single-particle wavefunction $\psi_i(t)$ is expanded in a set of M molecular wavefunctions $\{\varphi^{\mathrm{MO}}\}$

$$\psi_i(t) = \sum_{m=1}^{M} a_{im}(t)\varphi_m^{\mathrm{MO}}(\boldsymbol{R}(t)) \exp\left(-\frac{\mathrm{i}}{\hbar}\int^t \varepsilon_m(\boldsymbol{R}(t'))\,\mathrm{d}t'\right) \tag{4}$$

with $i = 1, \ldots, N$. The set of molecular wavefunctions $\{\varphi^{\mathrm{MO}}\}$ is taken as the solutions from the static DFS equation

$$\hat{h}^{\mathrm{DFS}}(\boldsymbol{R})\varphi_m^{\mathrm{MO}}(\boldsymbol{R}) = \varepsilon_m(\boldsymbol{R})\varphi_m^{\mathrm{MO}}(\boldsymbol{R}) \tag{5}$$

with $m = 1, \ldots, M$ [5]. Using this basis-set method the problem of solving the time-dependent single-particle equations (2) is equivalent to solving the single-particle matrix coupled-channel equations

$$\mathrm{i}\hbar\frac{\mathrm{d}}{\mathrm{d}t}a_{il} = \sum_{m=1}^{M} a_{im}\langle\varphi_l^{\mathrm{MO}}(R(t))| - \mathrm{i}\hbar\,\mathrm{d}/\mathrm{d}t|\varphi_m^{\mathrm{MO}}(R(t))\rangle$$
$$\times \exp\left(-\frac{\mathrm{i}}{\hbar}\int^t (\varepsilon_m(\boldsymbol{R}(t')) - \varepsilon_l(\boldsymbol{R}(t')))\,\mathrm{d}t'\right) \tag{6}$$

with $i = 1, \ldots, N$ for the single particle amplitudes a_{il} [11, 12]. During the solution of the static DFS equation the matrix elements are also calculated in an *ab initio* way.

Each of the N electrons in the collision system defines a new initial value problem for the M coupled channels taken into account in equation 6. One therefore ends up with N sets of single-particle amplitudes $\{a_{ij}\}$ with $i = 1, \ldots, N$ and $j = 1, \ldots, M$.

To match the many-particle aspect of the collision system one has to formulate the experimental questions as inclusive probabilities given in terms of single-particle amplitudes [6–10, 13]. Recently we developed a scheme to reduce the computational effort in calculating one of the most common experimental questions: finding a certain minimum number of vacancies or occupancies within a subset of states [13, 20].

3. Results and discussion

The asymmetric collision system 4.7 to 16 MeV S^{15+} on Ar was chosen as a good example to test both the full relativistic adiabatic molecular treatment of the electrons used in our Hamiltonian, and the inclusive probability formalism. Detailed double coincidence measurements between angle-resolved projectile and Ar K x-rays are available for this collision system [3]. In the experimental results the probability to find at least one hole in the target Ar K shell has an impact-parameter-dependent oscillation. The measurements were done for projectile energies from 4.7 to 90 MeV in an impact parameter range from 1000 to 20 000 fm.

To give a good description of the vacancy sharing and transfer including the effects of higher shells we solved the single-particle matrix coupled channel equations (6) for the 20 lowest relativistic channels $1(1/2)\pm$ to $8(1/2)\pm$ and $1(3/2)\pm$ to $2(3/2)\pm$. Figure 1 shows the corresponding correlation diagram which we obtain from solving the static DFS equation (5) for a large number of internuclear distances $R(t)$. Asymptotically the $1(1/2)\pm$ levels correlate to the S^{15+} 1s levels being occupied by one electron while the $2(1/2)\pm$ levels correlate to the Ar 1s levels being fully occupied. Owing to the high degree of ionization of the S^{15+} projectile the two K levels of target and projectile interchange. The occupation of the 18 remaining channels is chosen in correspondence to the asymptotic occupation of the separate atoms at $t=-\infty$.

To go beyond the single particle model we answer the experimental question $P_K^{Ar}(b)$ to find *at least one vacancy* in the Ar K shell within the frame of inclusive probabilities [10, 13]

$$P_K^{AR}(b)=P^{2(1/2)-,2(1/2)+}+P_{2(1/2)+}^{2(1/2)-}+P_{2(1/2)-}^{2(1/2)+}. \tag{7}$$

$P^{2(1/2)-,2(1/2)+}$ is the inclusive two-hole probability to find one vacancy in $2(1/2)-$ *and* one vacancy in $2(1/2)+$; $P_{2(1/2)+}^{2(1/2)-}$ is the inclusive one-hole one-particle probability to find one electron in $2(1/2)+$ *and* one vacancy in $2(1/2)-$; $P_{2(1/2)-}^{2(1/2)+}$ is the inclusive one-hole one-particle probability to find one electron in $2(1/2)-$ *and* one vacancy in $2(1/2)+$.

Figures 2 to 4 show the experimental values and different theoretical results for $P_K^{Ar}(b)$ versus our inclusive probability calculations presented as full curves for projectile energies from 4.9 to 16 MeV. All these inclusive probabilities of the three different energies show good agreement with the experiment for absolute size and oscillatory structure. A slight shift in the minima and maxima, probably due to the conversion from scattering angle to impact parameter, can be observed. The filling up of the minima and the damping of the maxima, to be seen in the experimental $P_K^{Ar}(b)$ curves for 4.7 and 7.9 MeV S^{15+} on Ar, is reproduced in our calculations.

To show the effect of inclusive probabilities we also calculated the single particle probability for K–K charge transfer. For the case of 4.7 MeV S^{15+} on Ar, figure 5 presents the results of the single-particle probability (broken curve) and the inclusive probability (full curve) versus the experimental results.

Four additional dynamic coupling calculations are available for the scattering system S^{15+} on Ar. The broken curves in figures 2–4 show results from Stolterfoht [18, 19] who used a fitted Hamiltonian and model matrix elements to calculate the charge transfer probability. Lin and Tunnell [14] used an *ab initio* two-state atomic expansion method to calculate the K–K charge transfer. The narrow dotted curve shows the results for 16 MeV S^{15+} on Ar. Fritsch and Lin [15, 16] improved the two-state atomic expansion model by adding united-atom orbitals to the basis set used. Results

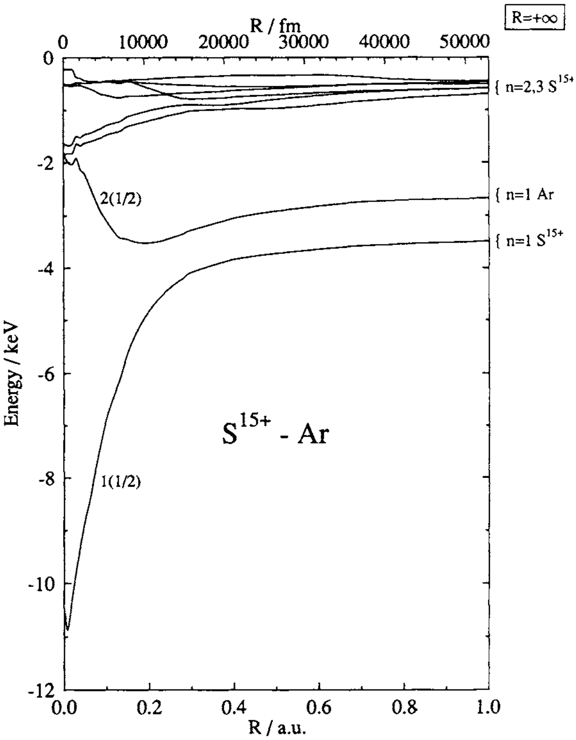

Figure 1. Correlation diagram for S^{15+}-Ar.

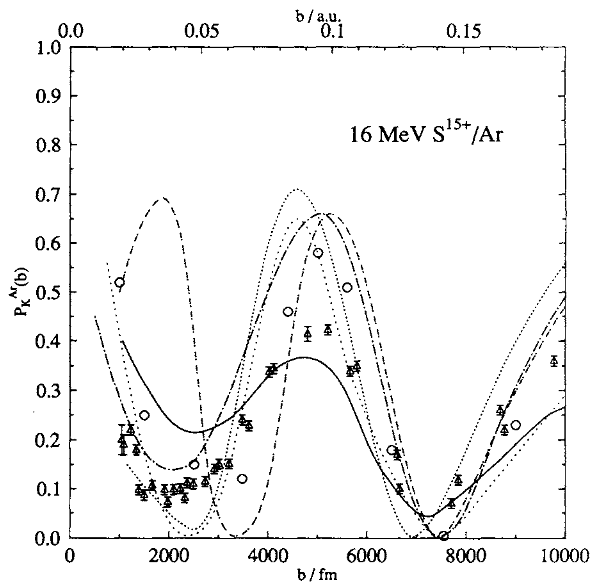

Figure 2. Experimental values: $P_K^{Ar}(b)$ in collision of 16 MeV S^{15+} on Ar. Broken curve, Stolterfoht *et al* [18, 19]; narrow dotted curve, Lin and Tunnell [14]; chain curve, Fritsch and Lin [15]; open circle, Grün *et al* [17]; wide dotted curve, Schuch *et al* [3]; full curve, our inclusive probability.

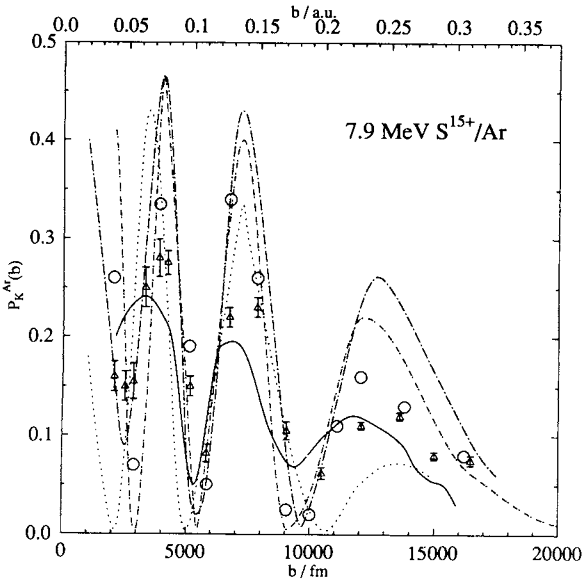

Figure 3. Experimental values: $P_K^{Ar}(b)$ in collision of 7.9 MeV S^{15+} on Ar. Broken curve, Stolterfoht *et al* [18, 19]; chain curve, Fritsch and Lin [15]; open circle, Grün *et al* [17]; wide dotted curve, Schuch *et al* [3]; full curve, our inclusive probability.

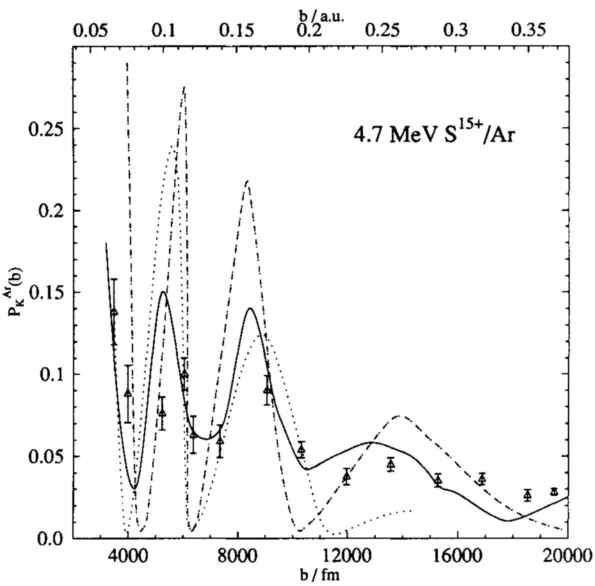

Figure 4. Experimental values: $P_K^{Ar}(b)$ in collision of 4.7 MeV S^{15+} on Ar. Broken curve, Stolterfoht *et al* [18, 19]; wide dotted curve, Schuch *et al* [3]; full curve, our inclusive probability.

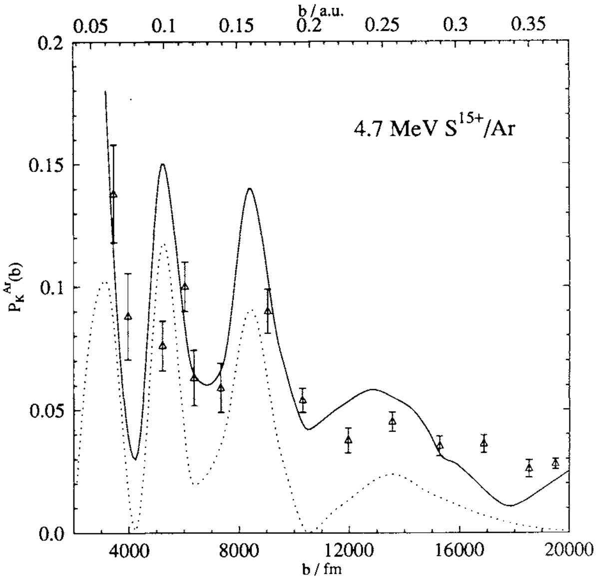

Figure 5. Experimental values: $P_K^{Ar}(b)$ in collsion of 4.7 MeV S^{15+} on Ar. Broken curve, single-particle K–K charge transfer probability without inclusion of Pauli principle; full curve, inclusive probability.

are shown in figures 2 and 3 as chain curves for 16 and 7.9 MeV S^{15+} on Ar. Finally the open circles in figures 2 and 3 show results from Grün *et al* [17] who solved the time-dependent Schrödinger equation for the scattering system 7.9 and 16 MeV S^{15+} on Ar.

An analytical formula used by Schuch *et al* [3] gives the K–K charge transfer probability presented in figures 2 to 4 as wide dotted curves.

All four *ab initio* theories are in good agreement with the experimental results but the filling up of the minima and smearing out of the oscillatory structure that is most pronounced in the low energy collisions seems to be best reproduced by our inclusive probability calculation.

The *ab initio* relativistic LCAO–MO description of the collision problem and the additional application of the inclusive probability formalism to reinstate the many-particle aspect allows the agreement of shape and absolute height with the experimental measurements to be improved especially for low collision energies.

Our results for the heavy asymmetric collision system S^{15+} on Ar show that even in inner shell processes like K–K charge transfer the many-particle aspect plays an important role. For low energy collisions on has to go beyond the single-particle model to improve the agreement with the experimental results.

Acknowledgments

This work was partially supported by the Deutsche Forschungsgemeinschaft (DFG) and the Gesellschaft für Schwerionenforschung (GSI). Computer time on the CRAY-YMP from the Höchstleistungsrechenzentrum (HLRZ) Jülich is gratefully acknowledged.

References

[1] Schuch R, Nolte G, Schmidt-Böcking H and Lichtenberg W 1979 *Phys. Rev. Lett.* **43** 1104

[2] Hagmann S, Cocke C L, Macdonald J R, Richard P, Schmidt-Böcking H and Schuch R 1982 *Phys. Rev.* A **25** 1918

[3] Schuch R, Ingwersen H, Justiniano E, Schmidt-Böcking H, Schulz M and Ziegler F 1984 *J. Phys. B: At. Mol. Phys.* **17** 2319

[4] Schulz M, Justiniano E, Konrad J, Schuch R and Salin A 1987 *J. Phys. B: At. Mol. Phys.* **20** 2057

[5] Sepp W-D, Kolb D, Sengler W, Hartung H and Fricke B 1986 *Phys. Rev.* A **33** 3679

[6] Reading J F 1973 *Phys. Rev.* A **8** 3262

[7] Reinhardt J, Müller B, Greiner W and Soff G 1979 *Phys. Rev. Lett.* **43** 1307

[8] Reading J F and Ford A F 1980 *Phys. Rev.* A **21** 124

[9] Becker R L, Ford A L and Reading J F 1984 *Phys. Rev.* A **29** 3111

[10] Lüdde H J and Dreizler R M 1983 *J. Phys. B: At. Mol. Phys.* **18** 107

[11] Kürpick P, Thies B, Sepp W-D and Fricke B 1991 *J. Phys. B: At. Mol. Opt. Phys.* **24** L139

[12] Kürpick P, Heinemann D, Sepp W-D and Fricke B 1991 *Z. Phys.* D **22** 407

[13] Kürpick P, Lüdde H J, Sepp W-D and Fricke B 1992 *Z. Phys.* D accepted

[14] Lin C D and Tunnell L N 1980 *Phys. Rev.* A **22** 76

[15] Lin C D, Winter T G and Fritsch W 1982 *Phys. Rev.* A **25** 2395

[16] Fritsch W and Lin C D 1985 *Phys. Rev.* A **31** 1164

[17] Grün N, Mülhaus A and Scheid W 1982 *J. Phys. B: At. Mol. Phys.* **15** 4043

[18] Stolterfoht N 1980 *J. Phys. B: At. Mol. Phys.* **13** L559

[19] Stolterfoht N 1983 *J. Phys. B: At. Mol. Phys.* **16** 2385

[20] Kürpick P and Lüdde H J 1993 *Comput. Phys. Commun.* accepted