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Impact parameter dependence of K-shell ionisation in slow collisions of near-symmetric relativistic atoms

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Abstract. The total K vacancy production probability of both collision partners in a heavy-ion–atom collision is calculated, assuming that two processes contribute, a single-collision process where $2p\sigma$ and $2p\pi$ vacancies are created by direct ionisation of the united atom and subsequently redistributed, and a double-collision process where a $2p\pi$ vacancy is created in the first collision and partially transferred in the second. The model used for the single-collision process is valid for slow collisions between partners of a combined charge greater than 70. Numerical results are presented for the (I, I) and (I, Ag) systems and show improved agreement with recent experiments.

1. Introduction

There has been much interest lately in the impact parameter distribution of the $2p\sigma$ vacancy production in adiabatic collisions. For light collision systems ($Z < 18$) when a $2p\pi$ vacancy is brought into the collision, the rotational $2p\pi$–$2p\sigma$ coupling model of Taulbjerg *et al* (1976) can describe the experimental results for gas targets well (Sackmann *et al* 1974, Luz *et al* 1979). However, in experiments on heavier systems discrepancies between the predicted and measured impact parameter dependences emerged (Annett *et al* 1979, Schuch *et al* 1979, Anholt *et al* 1980, Morenzoni *et al* 1982b). While the rotational coupling model predicts a double-peak structure, the 'kinematic peak' at small impact parameters b, and the 'adiabatic peak' at large b, it has been found that the adiabatic peak is strongly suppressed when no L-shell vacancies are brought into the collision. The intensity of the kinematic peak, on the other hand, lies far above the theoretical estimate, provided the theory is normalised to experiment so that the total cross sections agree. An extension of the $2p\pi$–$2p\sigma$ coupling model to relativistic systems (Jakubassa and Taulbjerg 1980) showed that relativistic effects were too small to account for these discrepancies.

It has been suggested that the additional intensity in the kinematic peak region, which is seen if the theory is subtracted from the data, can be accounted for by direct $1s\sigma$ or $2p\sigma$ ionisation, because of its strong fall-off with b (Nolte *et al* 1980). This would imply that the measured K vacancy production probability results from two independent processes, the coupling down of a $2p\pi$ vacancy produced in a previous

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collision, and direct ionisation in the second collision. On the other hand, it has also been suggested that in the first of these processes the impact parameter dependence of the mechanism creating the initial vacancy will influence the final distribution over b. The resulting b dependence will then deviate from a rotational coupling model where the number of L-shell vacancies is assumed to be constant (Morenzoni *et al* 1982b).

In this paper we examine these ideas quantitatively for a solid target. In order to calculate the single-collision contribution to the $2p\sigma$ vacancy production, we shall introduce a simplified model, the post-ionisation coupling model. In this model it is assumed that one can describe the K vacancy production as a two-step process. In the first step vacancies are created by direct Coulomb excitation to the continuum at small internuclear distances R. In the second step these vacancies are redistributed among the $2p\sigma$ and $2p\pi$ states during the outgoing part of the collision. This model is appropriate for slowly colliding relativistic systems, where the L-shell spin–orbit splitting causes the coupling to take place at comparatively large values of R.

An outline of the model is given in § 2. The direct ionisation is calculated within the Briggs model, i.e. using the MO picture with time-independent united-atom (UA) wavefunctions and energies (Briggs 1975, Amundsen 1978a). The redistribution of the ionisation amplitudes among the $2p\sigma$ and $2p\pi$ states is evaluated by means of a coupled-channel calculation, using first-order united-atom perturbation theory for the MO energies and wavefunctions, and a small-R expansion of the matrix elements (Jakubassa and Taulbjerg 1980).

For the calculation of the double-collision process (§ 3), the impact-parameter-dependent K-shell vacancy production probability obtained by transferring an initial $2p\pi$ vacancy to the $2p\sigma$ state, is folded with the experimental b-dependent L vacancy production probability from a previous collision (Anholt 1982). In order to demonstrate the importance of the folding procedure for the impact parameter distribution, this is compared with a simpler theory which includes the number of initial L vacancies by means of a multiplication with the (b-independent) total cross section for previous L vacancy production (Meyerhof *et al* 1977). In an appendix we show that the UA $2s\sigma$ ionisation does not contribute to the K-shell vacancy production.

Numerical calculations and a comparison with experiment are given in § 4. Concluding remarks follow in § 5. Atomic units ($\hbar = m = e = 1$) are used unless otherwise indicated. We denote the (adiabatic) MO states which correlate to the united atom $2p_{1/2,1/2}$, $2p_{3/2,1/2}$ and $2p_{3/2,3/2}$ states by $2p\sigma(1/2)$, $2p\pi(1/2)$ and $2p\pi(3/2)$, respectively, where the last number is the m quantum number of the total angular momentum along the internuclear axis.

2. The post-ionisation coupling model

2.1. General assumptions

The model is based on the usual semiclassical approximation for the collision system, where the internuclear motion is described by a classical Rutherford trajectory $\boldsymbol{R}(t)$, and an independent-particle description for the electrons is adopted. Since we shall only be interested in situations where the probability for a vacancy to be present at all in the L shell is much smaller than one, it is convenient to consider vacancies instead of electrons as the active 'particles' of the collision process. The ionisation is then described as the capture of a vacancy from the continuum. This vacancy is

subsequently redistributed among the MO correlated to the separated atom K and L shells. The Hamiltonian of the active vacancy can be written as

$$H = T + V_0 + V_1$$

$$V_1 = \frac{Z}{r} - \frac{Z_1}{|\boldsymbol{r} - \alpha \boldsymbol{R}|} - \frac{Z_2}{|\boldsymbol{r} + \beta \boldsymbol{R}|} \tag{2.1}$$

Here, T is the kinetic energy, $V_0 = -Z/r$, Z_1 and Z_2 are the projectile and target charges, respectively, $Z = Z_1 + Z_2$, and the centre of charge is chosen as the origin of the electron coordinates, implying $\alpha = Z_2/Z$ and $\beta = 1 - \alpha$. Let $\psi_n(t)$ and $\varepsilon_n(t)$ be the eigenstates and energies of H at fixed R, such that the solution ψ of the Dirac equation can be written as a superposition of these states with time-dependent coefficients $a_n(t)$. Insertion into the Dirac equation $i\dot{\psi} = H\psi$ leads to the well known system of differential equations for $a_n(t)$:

$$\dot{a}_n = \frac{1}{i} a_n \varepsilon_n - \sum_k a_k \left\langle \psi_n(t) \left| \frac{\partial}{\partial t} \right| \psi_k(t) \right\rangle - \sum_f a_f \left\langle \psi_n(t) \left| \frac{\partial}{\partial t} \right| \psi_f(t) \right\rangle$$

$$\dot{a}_f = \frac{1}{i} a_f \varepsilon_f - \sum_k a_k \left\langle \psi_f(t) \left| \frac{\partial}{\partial t} \right| \psi_k(t) \right\rangle - \sum_{f'} a_{f'} \left\langle \psi_f(t) \left| \frac{\partial}{\partial t} \right| \psi_{f'}(t) \right\rangle \tag{2.2}$$

where a_f and $a_{f'}$ are the amplitudes of the continuum states, while a_n and a_k denote bound-state amplitudes.

To solve these coupled-channel equations is a formidable task, in particular since couplings to continuum states are included. In order to make the problem numerically more tractable, we introduce a model, which we call the post-ionisation coupling model, where the following simplifying approximations are introduced.

(i) We retain only those bound states that correlate to the UA L shell, neglecting couplings to the UA K shell and to higher-lying states. This excludes the possibility of producing $2p\sigma$ vacancies via Coulomb transitions to high-lying vacant states. Unless the collision velocity is extremely low, the probability for such transitions is small compared with the ionisation probability (Walske 1951), essentially due to the small phase space of the discrete final states.

(ii) Back coupling from the continuum is neglected, since the ionisation amplitudes are small.

(iii) We shall assume that the ionisation process and the coupling between the UA L-shell states can be separated in time. Our justification for this approximation is the following: if no vacancy is brought into the collision, vacancies are only created by direct excitation, which takes place during a time $\Delta t \simeq (\Delta E)^{-1}$ when the two nuclei are near their point of closest approach, ΔE being the energy transferred to the electron, which is typically the UA binding energy. On the other hand, the $2p\pi$–$2p\sigma$ radial coupling mainly takes place at relatively large internuclear distances $R \simeq \frac{1}{2} a_L$ for relativistic systems, where $a_L = 2/Z$, i.e. at time scales $t_R \simeq R/v \simeq a_L/2v$, v being the collision velocity. Near $R = 0$ the rotational coupling is strongly suppressed by the spin–orbit splitting between the UA $2p_{1/2}$ and $2p_{3/2}$ states, in contrast to the non-relativistic case. Since $\Delta E \simeq \frac{1}{8} Z^2$, one finds that $\Delta t/t_R \simeq 8v/Z = 4v/v_L$, which leads to the condition

$$v/v_L \ll \tfrac{1}{4} \tag{2.3}$$

for the present model to be reasonable, $v_L = \frac{1}{2} Z$ being the UA L-shell velocity.

2.2. The ionisation process

Within the framework of the MO model, first-order perturbation theory leads to an ionisation amplitude at time t given by

$$A_{fi}(t)=\int_{-\infty}^{t} \mathrm{d}t \frac{1}{\varepsilon_f-\varepsilon_i}\left\langle \psi_f(R)\left|\frac{\partial}{\partial t}(V_0+V_1)\right|\psi_i(R)\right\rangle \exp\left(\mathrm{i}\int_{-\infty}^{t}(\varepsilon_f-\varepsilon_i)\,\mathrm{d}t\right). \tag{2.4}$$

where we used

$$\psi_n(t)=\psi_n(R)\exp\left(-\mathrm{i}\int_{-\infty}^{t}\varepsilon_n\,\mathrm{d}t\right)$$

When ionisation takes place at small internuclear distances (which is implied by the condition (2.3)), the wavefunctions $\psi_{f,i}(R)$ and energies $\varepsilon_{f,i}$ can be replaced by their time-independent united-atom limiting values. By means of a partial integration, (2.4) can then be cast into the standard form of the Briggs model (Briggs 1975). For the further evaluation one introduces the Fourier transform of V_0+V_1 and makes a partial-wave decomposition of the potential. The time and space integrals then separate and can be carried out (for details see, for example, Kocbach 1976).

According to assumption (iii) above, the ionisation process is essentially finished at a time t_c ($t_c \gg \Delta E^{-1}$), so that we can approximate $A_{fi}(t)$ by $A_{fi}\equiv A_{fi}(\infty)$ for $t \geqslant t_c$, and only A_{fi} will be needed in the calculations. In order to apply existing techniques we chose to calculate the ionisation amplitude in a space-fixed coordinate system. A_{fi} then follows from (Amundsen 1978a) (with $\hat{L}=2L+1$):

$$\begin{aligned} A_{fi}=\frac{4\mathrm{i}}{\pi^{1/2}}\sum_{LM}(\hat{L}\hat{j}_i\hat{j}_f)^{1/2}\begin{pmatrix} j_f & L & j_i \\ \frac{1}{2} & 0 & -\frac{1}{2}\end{pmatrix}(-1)^{m_f+1/2}\begin{pmatrix} j_f & L & j_i \\ -m_f & M & m_i\end{pmatrix}(-\mathrm{i})^{l_f} \\ \times\sum_{M'=-L}^{L} d^L_{M'M}(\theta)\int_0^\infty \mathrm{d}s\langle j_L(s)\rangle_{fi}(Z_1 B_{LM'}(b,q;\alpha s) \\ +(-1)^L Z_2 B_{LM'}(b,q;\beta s)) \end{aligned} \tag{2.5}$$

where the initial (final) states, being UA eigenstates, are characterised by the angular quantum numbers $j_i, l_i, m_i (j_f, l_f, m_f)$, $\langle\ \rangle_{fi}$ is a *radial* matrix element, and $B_{LM}(b,q;s)$ are the path factors resulting from the time integration with a hyperbolic internuclear path, which for monopole and dipole transitions can be found in Amundsen (1978b); an extension to arbitrary L will be published elsewhere. Furthermore, $q=(\varepsilon_f-\varepsilon_i)/v$ denotes the minimum momentum transfer, the symbols in parentheses are Wigner 3-j symbols, and the rotation matrices $d^L_{M'M}(\theta)$ are introduced to rotate the coordinate system from the one in which the path factors have been defined (with the internuclear axis at $t=0$ along the x axis) to that in which the internuclear vector at the beginning of the coupling calculations is along the z axis.

2.3. The coupling process

After the vacancy has been created, it is redistributed among the MO levels. In the present model, equation (2.2) reduces to

$$\dot{a}_n(t)=\frac{1}{\mathrm{i}}a_n\varepsilon_n-\sum_k a_k\left\langle\psi_n(t)\left|\frac{\partial}{\partial t}\right|\psi_k(t)\right\rangle \tag{2.6}$$

with the boundary condition

$$a_n(t_c) = A_{fn}(t_c) \tag{2.7}$$

where $t_c \ll t_R$ is chosen so that, according to assumption (iii) above, the coupling between the MO is still weak. The amplitudes $a_n(t)$ will, except for an unimportant common phase, not change much for $0 < t < t_c$, because the energy differences between the relevant states are small compared with ΔE, and the off-diagonal elements from radial and rotational couplings are also small in this region. Thus equation (2.7) is approximately equivalent to

$$a_n(0) = A_{fn} \tag{2.8}$$

which is taken to be the boundary condition for our numerical calculations. The corresponding value for θ in (2.5) is $\theta = -\frac{1}{2}\pi$.

Since it is crucial for the validity of our model that one can indeed find a time t_c, such that the ionisation process is practically ended while the redistribution of the vacancy among the bound states is not yet important at this time, it should be stressed that the theoretical estimates given above are confirmed by numerical calculations (discussed in § 4); if the condition (2.3) is fulfilled, such a t_c does exist, with a typical value $t_c \simeq 2(\Delta E)^{-1}$.

For establishing and solving the system of differential equations we proceed similarly as in Jakubassa and Taulbjerg (1980). There are eight states correlating to the united-atom L shell to be coupled, namely $2p\sigma(\pm 1/2)$, $2p\pi(\pm 1/2)$, $2p\pi(\pm 3/2)$ and $2s\sigma(\pm 1/2)$, which coincide for $R \to 0$ with the $2s\sigma(\pm 1/2)$ state. We start with calculating the diabatic states using united-atom perturbation theory, i.e.

$$E_{ljm}(R) = E^{\mathrm{UA}}_{ljm} + V_{ljm,ljm}$$

where

$$V_{ljm,l'j'm'} = \langle \psi^{\mathrm{UA}}_{ljm} | V_1 | \psi^{\mathrm{UA}}_{l'j'm'} \rangle \tag{2.9}$$

and retain only the lowest expansion term in R. This can be justified because the level crossings where the main contribution to the transitions between these states comes from, occur at internuclear distances which are smaller than the united-atom L-shell radius. The approximation is probably not so good for relativistic systems as for non-relativistic ones (where the error in the vacancy distributions is smaller than 25% as estimated by Taulbjerg *et al* (1976)), since the couplings take place at larger R than in the non-relativistic case. Recently, relativistic Hartree–Fock MO correlation diagrams have become available for the (I, I) system (Morović *et al* 1982), indicating that calculations based on the small-R expansion may not be very accurate. However, the use of coupling matrix elements derived from these correlation diagrams in our calculations would imply a large numerical effort. Anyhow, we judge that the present matrix elements will give a qualitatively correct estimate of the vacancy production rate.

The adiabatic states are obtained from the diabatic ones by means of diagonalisation. As the potential coupling $V_{ljm,l'j'm'}$ mixes only states with $m = m'$, the states $2p\pi(\pm 3/2)$ are decoupled and in this approximation identical with the diabatic ones, while the resulting adiabatic states with $m = \frac{1}{2}$ are degenerate in energy to those with $m = -\frac{1}{2}$.

In the appendix it is shown that for our purpose only the coupling between the $2p\pi$ and $2p\sigma$ states is important when solving the system of differential equations, while the $2s\sigma$ state can be neglected.

Instead of expanding the wavefunction ψ directly in terms of the adiabatic states which, after insertion into the Dirac equation, leads to coupled equations for the coefficients a_n (cf equation (2.2)) an expansion in the following linear combination of the adiabatic states

$$\psi = \sum_{n=\pm 1} \Big(c_1^n \frac{1}{\sqrt{2}} (\psi_{2p\sigma(1/2)} + in\psi_{2p\sigma(-1/2)}) + c_2^n \frac{1}{in\sqrt{2}} (\psi_{2p\pi(1/2)} + in\psi_{2p\pi(-1/2)}) + c_3^n \frac{1}{\sqrt{2}} (\psi_{2p\pi(3/2)} + in\psi_{2p\pi(-3/2)}) \Big) \tag{2.10}$$

results in the reduction of the 6×6 system of differential equations for a_n to two decoupled 3×3 systems of differential equations for the coefficients c_i^n as given in equation (A.8) of Jakubassa and Taulbjerg (1980). The a_n are linear combinations of c_i^n (for example, $a_{2p\sigma(1/2)} = 2^{-1/2}(c_1^+ + c_1^-)$ and $a_{2p\sigma(-1/2)} = i2^{-1/2}(c_1^+ - c_1^-)$). It is convenient to add a differential equation for the internuclear path $R(t)$.

The initial conditions for c_i^n at time $t = 0$ are obtained from (2.8) and (2.10)

$$\begin{aligned} c_1^{\pm} &= (A_{f,2p_{1/2,1/2}} \mp iA_{f,2p_{1/2,-1/2}})/\sqrt{2} \\ c_2^{\pm} &= (A_{f,2p_{3/2,-1/2}} \pm iA_{f,2p_{3/2,1/2}})/\sqrt{2} \\ c_3^{\pm} &= (A_{f,2p_{3/2,3/2}} \mp iA_{f,2p_{3/2,-3/2}})/\sqrt{2} \end{aligned} \tag{2.11}$$

with the ionisation amplitudes taken from (2.5). Due to the selection rule that $l + l_i + l_f$ is even, the amplitudes show the following symmetry

$$A_{fi}(-m_f, -m_i) = (-1)^{j_f + m_f + j_i + m_i}(-1)^{l_f + l_i} A_{fi}(m_f, m_i) \tag{2.12}$$

such that the initial conditions for negative m_f can immediately be found from

$$\begin{aligned} c_i^{\pm}(-m_f) &= \pm c_i^{\pm}(m_f) \qquad i = 1, 3 \\ c_2^{\pm}(-m_f) &= \mp c_2^{\pm}(m_f) \end{aligned} \tag{2.13}$$

where an irrelevant common phase has been dropped. The coupled equations must be solved separately for each different final state. The total probability for a vacancy to be present in the $2p\sigma$ state is then

$$P_{2p\sigma}^{IC} = \int_0^\infty d\varepsilon_f \sum_{j_f l_f m_f} (|c_{1f}^+(t=\infty)|^2 + |c_{1f}^-(t=\infty)|^2 \tag{2.14}$$

where it has been summed over the magnetic quantum number of the $2p\sigma$ state, and we have indicated that $c_1^{\pm}$ depends on the continuum state $|f\rangle$ through the boundary conditions. Note that due to the Hermiticity of the interaction the total number of L vacancies is conserved, and we have

$$\sum_n P_n^{IC} \equiv P_{L,UA}^{ION} = \sum_n \int_0^\infty d\varepsilon_f \sum_{j_f l_f m_f} |A_{fn}|^2 \tag{2.15}$$

where the sum over n extends over all p states correlating to the united-atom L shell.

3. The double-collision process

Let us assume that a projectile impinges on a target atom 1 with impact parameter b_1 and is thereby scattered through a (solid) angle Ω_1 (in the laboratory system). During this collision, an L vacancy is produced with a probability $P_L(b_1)$. Let us further assume that the lifetime, τ_x, of this vacancy is long enough and the density n_2 of the target high enough such that the vacancy is still present when the next collision occurs. If the impact parameter with respect to target atom 2 is b_2 then the projectile is scattered through the corresponding angle, Ω_2. In this collision, the vacancy approaches the target atom in a $2p\pi$ state and is, by means of radial and rotational couplings, transferred to the $2p\sigma$ state with a probability $P_{RC}(b_2)$.

It is convenient to define an equivalent single-collision impact parameter b corresponding to the total scattering angle $\Omega = \Omega_1 + \Omega_2$. This is the 'apparent' impact parameter one will deduce if one assumes that the process under consideration is a single-collision process. The K vacancy production probability $P_{MC}(b)$ from this multicollision process can then be expressed as (Anholt 1982)

$$P_{MC}(b)\frac{d\boldsymbol{b}}{d\Omega} = c_0 \int \frac{d\boldsymbol{b}_1}{d\Omega_1} d\Omega_1 P_L(b_1) \frac{d\boldsymbol{b}_2}{d\Omega_2} P_{RC}(b_2) \tag{3.1}$$

where the integration is performed over all scattering angles Ω_1 in the first collision. The weight coefficient c_0 is the average number of $2p\pi$ vacancies brought into the collision. Meyerhof *et al* (1977) find that is to a good approximation given by

$$c_0 = N n_2 v \tau_x w_{2p} \tag{3.2}$$

where $n_2 v \tau_x$ is the number of 2p vacancies per projectile inside the target material, and w_{2p} is the vacancy sharing fraction between the 2p states of the target and the projectile (being one half for symmetric systems and near one for asymmetric systems with $Z_1 > Z_2$). N is a statistical factor which is determined by the redistribution of the L vacancy over the molecular states and equals two thirds if P_{RC} is normalised to one incident vacancy, and if one can assume that the vacancies are distributed statistically among the magnetic substates (Jakubassa and Taulbjerg 1980). The validity of the latter assumption may be somewhat uncertain.

In order to use formula (3.1) one needs P_L. A theoretical calculation of this quantity would be even more involved than the calculation of P_K described in the previous section. However, there exist measurements of P_L of a sufficient quality for our purpose for at least one system of interest, as will be discussed in the next section.

Using the relation between impact parameter and laboratory scattering angle for pure Rutherford scattering

$$b = \frac{R_0}{2}\left(\frac{2}{1-\cos\vartheta[1-(m_1/m_2)^2\sin^2\vartheta]^{1/2}+(m_1/m_2)\sin^2\vartheta} - 1\right)^{1/2} \tag{3.3}$$

where m_1 and m_2 are the masses of projectile and target, respectively, (3.1) can be written as

$$P_{MC}(b) = \left(\frac{b\,db}{\sin\vartheta\,d\vartheta}\right)^{-1} c_0 \int_0^{\vartheta_{max}} d\vartheta_1 b_1 \frac{db_1}{d\vartheta_1} P_L(b_1) 2 \int_0^{\varphi_{max}} d\varphi_1 P_{RC}(b_2) \frac{b_2\,db_2}{\sin\vartheta_2\,d\vartheta_2} \tag{3.4}$$

with

$$\cos\vartheta_2 = \cos\vartheta_1\cos\vartheta + \sin\vartheta_1\sin\vartheta\cos\varphi_1.$$

Here the upper integration limits have to be chosen such that b_1, b_2 and ϑ_2 attain physical values, which for $m_1 \geqslant m_2$ leads to $\vartheta_{\max} = \sin^{-1}(m_2/m_1)$ and $\varphi_{\max} = \cos^{-1}\lambda$ with

$$\lambda = ([1-(m_2/m_1)^2]^{1/2} - \cos\vartheta_1\cos\vartheta)/\sin\vartheta_1\sin\vartheta$$

if λ lies between -1 and 1, π if $\lambda < -1$ and 0 if $\lambda > 1$. Note that the weight factor

$$\frac{\mathrm{d}\boldsymbol{b}_2}{\mathrm{d}\Omega_2}\left(\frac{\mathrm{d}\boldsymbol{b}}{\mathrm{d}\Omega}\right)^{-1}$$

i.e. the ratio between the elastic differential cross sections, tends to one for small b but produces the fall-off of $P_{\mathrm{MC}}(b)$ at large b.

4. Numerical results and comparison with experiment

We have calculated the probability for the sum of projectile and target K vacancy production, which corresponds to the probability of producing a vacancy in the $2p\sigma$ state, for the (I, I) and (I, Ag) collision systems. We used screened relativistic hydrogenic wavefunctions with an effective charge of Z_1+Z_2-2 (Taulbjerg *et al* 1976) and experimental UA binding energies. Apart from the modified path factors, the amplitudes for direct ionisation were evaluated from (2.5) by the method described in Aashamar and Amundsen (1981). We included transitions up to multipolarity $L = 2$. The only other multipole that can contribute significantly to L-shell ionisation is $L = 3$, but for near-symmetric or symmetric systems this is, like the $L = 1$ amplitude, almost completely cancelled between the projectile and target terms of the perturbing potential. The coupling between the $2p\sigma$ and $2p\pi$ states was calculated from the system of differential equations (A.8) in Jakubassa and Taulbjerg (1980). The vacancy production probability $P^{\mathrm{IC}}(b)$ of the single-collision process was obtained from (2.14) with the initial conditions (2.11). The probability for creating a K vacancy in a collision with one incident $2p\pi$ vacancy,

$$P_{\mathrm{RC}}(b) = |c_1^+(t=\infty)|^2 + |c_1^-(t=\infty)|^2 \tag{4.1}$$

is found by solving the differential equations with the initial conditions $c_1^n = 2^{-1/2}$, $c_2^n = c_3^n = 0$, $n = \pm 1$ at $t = -\infty$.

One of the important approximations in the present model is the replacement of the time t_c by zero in the coupling amplitude a_n, and by infinity in the ionisation amplitude A_{fn}, equation (2.8). In order to check this, i.e. to test the stability of our calculation of $\mathrm{P}^{\mathrm{IC}}(b)$ against changes in t_c, we solved the coupled equations with the initial condition (2.8) up to times $t_c = 2\Delta t$ where Δt is taken as the inverse united-atom $2p_{1/2}$ binding energy. We found that for the collision systems considered here, the change of the MO occupation amplitudes with respect to their UA values was at most about five per cent (at the highest velocity). Within the present computational scheme it is not so easy to check numerically that the ionisation process is really terminated at this time. This would require the evaluation of the path factors, B_{LM}, in equation (2.5) at a finite time, which is presently not numerically feasible. The only relevant calculations we have found in the literature, although employing somewhat different

wavefunctions, are those of Soff *et al* (1979) for 1sσ ionisation in very heavy collision systems (Pb–Pb and U–U). These calculations show that, although the ionisation probability has approached its final value at $t_c = 2\Delta t$, it continues to oscillate with a rather large amplitude, about $\pm 30\%$ for the Pb+Pb system at $v/c \simeq 0.1$. These oscillations are even larger for the U+U system, and this may be a systematic effect connected with the approach of the 1sσ orbital to the negative energy continuum, in which case these results are not very relevant to our case of 2p ionisation in much lighter systems. Additionally, for large values of t_c the wavefunctions are no longer well represented by one-centre (UA) wavefunctions, but are distorted into proper molecular orbitals, which should also be equipped with appropriate translational factors. These changes in the wavefunctions introduce additional time dependences which will generally tend to damp the oscillations in the ionisation matrix elements. Concerning the translational factors alone, this effect is illustrated by Vaaben and Taulbjerg (1981). Whether this additional damping is already important at $t_c \sim 2\Delta t$ is uncertain, as relevant numerical evidence is not available. However, even with a 15% residual oscillation in the ionisation amplitude, as indicated by the work of Soff *et al* (1979), we do not find the assumption of our model—that the ionisation process and the coupling can be separated in time—to be unreasonable.

Figures 1 and 2 show the K vacancy production probability for the single-collision process in (I, I) collisions as a function of impact parameter, $P^{IC}(b)$, calculated with

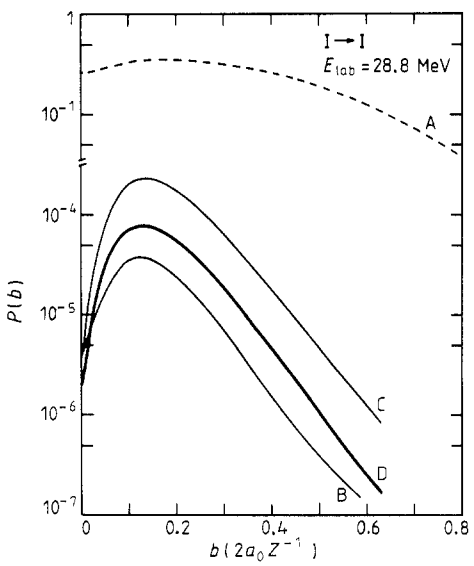

Figure 1. Probability for K vacancy production in (I, I) collisions as a function of the impact parameter ($a_0 = 5.3 \times 10^4$ fm is the Bohr radius) at a collision velocity of $v/c = 0.022$. Curve A denotes the coupling probability $P_{RC}(b)$, curves B and C are the ionisation probabilities $P^{ION}_{2p_{1/2}}$ and $P^{ION}_{2p_{3/2}}$, respectively, and curve D is the probability P^{IC} from the post-ionisation coupling model.

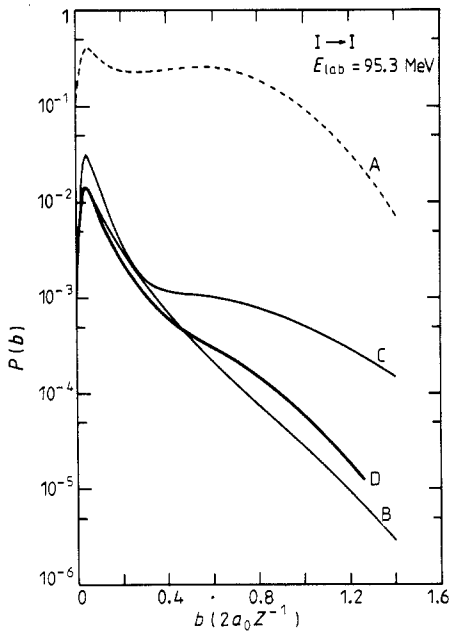

Figure 2. Probability for K vacancy production in (I, I) collisions as a function of impact parameter at a collision velocity of $v/c = 0.04$ (otherwise as in figure 1.).

the present model for the collision energies of 28.8 and 95.3 MeV ($v/c = 0.022$ and 0.04), respectively. In these cases the inequality (2.3) is well fulfilled ($v/v_L \simeq 0.1$ for the higher energy). Also shown are the corresponding probabilities for a pure radial and rotational coupling, $P_{RC}(b)$, and the UA $2p_{1/2}$ and $2p_{3/2}$ ionisation probabilities ($P^{ION}_{2p_{1/2}}$ and $P^{ION}_{2p_{3/2}}$). It is seen that the necessity to create a vacancy during the collision not only reduces the vacancy production probability dramatically, but also changes $P(b)$ significantly, because of the strong impact parameter dependence of the ionisation process.

For the higher collision energy it is seen that P_{RC} has the well known two-peak structure of the rotational coupling model, with a kinematic peak at small b and an adiabatic peak at larger b. The strong fall-off of the ionisation amplitude at large b reduces the adiabatic peak to a narrow shoulder in P^{IC}, while the kinematic peak has been reinforced by the peaked structure of the 2p ionisation probability around 90° scattering angle at low collision energies ($v/v_L \leq 0.1$) due to quadrupole transitions to the continuum (Aashamar *et al* 1978). For the lower energy there is no kinematic peak, since small enough internuclear distances are not reached during the collision. The structure in $P^{IC}(b)$ is then almost entirely determined by the ionisation process.

If one considers transitions to a specific continuum state (with given j_f, l_f, m_f and ε_f), large oscillations are seen in the calculated b distributions. However, when the sum over all final states is performed, they are completely averaged out. Incidentally, this averaging out of details makes us confident that our results are relatively stable against changes in the details of our model. It is seen from the figures that at large b, $P^{IC}(b)$ is above the $2p_{1/2}$ ionisation probability, indicating that a considerable fraction of the $2p\pi$ vacancies is coupled down. At small b on the other hand, more $2p\sigma$ vacancies are transferred to the $2p\pi$ state than vice versa. These features may be explained by noting that at large b the coupling is too weak to allow for more than just one coupling, in which case a net transfer of a vacancy from the state with the higher vacancy number will result, while for small b several couplings back and forth are likely.

For the (I, I) case, no experimental data seem to be available. However, for the very similar (I, Ag) system the impact parameter dependences of both K- and L-shell vacancy production have been measured. In order to evaluate P_{MC} from the equation (3.4), we have taken the measured $P_L(b)$ values of Morenzoni *et al* (1982a) and fitted an exponential tail, $\exp(-\kappa b)$, at large b where no data were available, so that the experimental L vacancy production cross section ($\sigma_L = 7.88 \times 10^4$ b; Meyerhof *et al* 1977) was reproduced. The dependence of $P_{MC}(b)$ on the details of this fitting was found to be negligible for the values of b where K-shell ionisation has been measured.

In figure 3 we show the calculated single-collision and double-collision K-shell ionisation probabilities for 63 MeV collision energy, together with experimental results of Morenzoni *et al* (1982b) and Anholt *et al* (1980) (measured at 60 MeV). The contribution from the $1s\sigma$ ionisation is negligible ($P_{1s\sigma}(b) \leq 2 \times 10^{-7}$, according to the Briggs model). We also compare with the simplified double-collision formula of Meyerhof *et al* (1977)

$$P_M(b) = c_0 \sigma_L P_{RC}(b) \tag{4.2}$$

where σ_L is the total cross section for L vacancy production in the first collision and c_0 is defined in (3.2). It is seen that this formula reproduces the double-collision results very well for impact parameters up to the L-shell scaling length a_L. Our

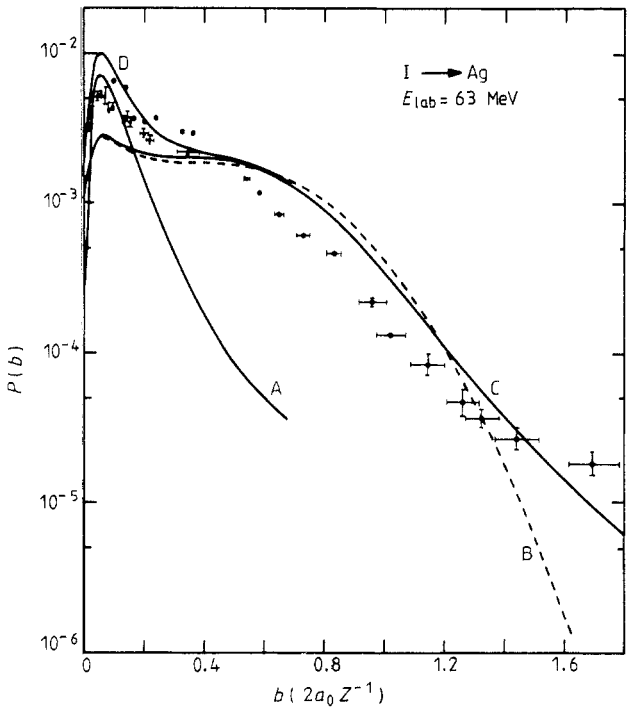

Figure 3. Probability for the projectile and target K vacancy production in 63 MeV (I, Ag) collisions as a function of impact parameter. Curve A denotes the probability P^{IC} from the post-ionisation coupling model, curves B and C are the double-collision probabilities P_M and P_{MC}, respectively, and curve D is the sum of A and C. The data (●) are from Morenzoni *et al* (1982). Also shown are the data from a 60 MeV collision (○) from Anholt *et al* (1980).

calculations are also in good agreement with the 'in-plane' double-collision formula of Anholt (1982) at $b \lesssim a_L$, but as noted by him, his formula also breaks down at large b.

From figure 3 it is clearly seen that the K-vacancy production is dominated by different processes for different impact parameter ranges. At large impact parameters the dominating process is indeed the double-collision process, which is the conventional explanation of K vacancy production in near-symmetric systems. However, at very small impact parameters corresponding to the kinematic peak, the single-collision process strongly dominates. When these two processes are added, they describe the qualitative features of the experimental $P(b)$ quite well over the whole impact parameter region. As for the quantitative agreement, it should be kept in mind that our perturbative calculation of the molecular orbitals gives an energy separation between the $2p\sigma(1/2)$ and $2p\pi(1/2)$ levels which is too small at the avoided crossing compared with more elaborate calculations (Morović *et al* 1982). This may lead to an overestimate of the coupling strength and be a reason why our theory lies above the data. Also at b larger than the L-shell radius, the united-atom perturbation theory is no longer reliable. Another uncertainty is introduced by the statistical factor N and by the fact that the experimental $P_L(b)$ is usually an average over the ionisation probabilities of the $2p_{3/2}$ and $2p_{1/2}$ states.

In figure 4 the K vacancy production probability for 100 MeV (I, Ag) collisions is shown. As, to our knowledge, no data for $P_L(b)$ are available, we have calculated the double-collision process only by means of (4.2) which should be reliable for $b \leqslant a_L$. When compared with the experimental data from Guillaume *et al* (1978) the sum of the single-collision and double-collision probabilities again shows a similar b dependence. For small impact parameters, however, the data are more overestimated than for the lower collision velocity.

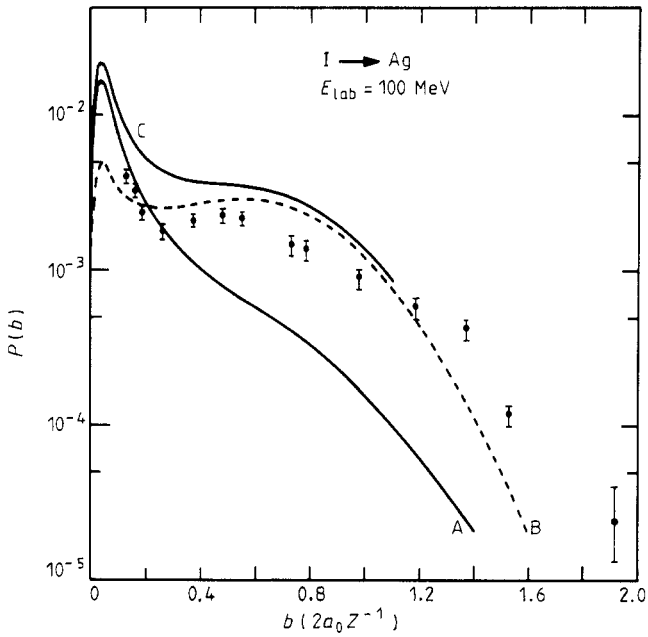

Figure 4. Probability for the projectile and target K vacancy production in 100 MeV (I, Ag) collisions as a function of impact parameter. Curve A denotes the probability P^{IC} from the post-ionisation coupling model, curve B is the double-collision probability P_M and curve C is their sum. The data are from Guillaume *et al* (1978).

The numerical results presented here are for two systems only, but the model is applicable for all systems where $2p\pi$–$2p\sigma$ couplings contribute to the K vacancy production rate, provided that the combined system is heavy enough for the UA $2p_{1/2}$–$2p_{3/2}$ spin–orbit splitting to be large ($Z \geqslant 70$), and the collision energy low enough to keep the $2p\sigma(1/2)$–$2p\pi(1/2)$ avoided crossing well outside the adiabatic distance q^{-1}. Electronic relativistic effects in the ionisation amplitudes will to some extent favour the single-collision process for very heavy systems, but these effects are not nearly as large for the UA L shell as they are for the K shell. Since the single-collision contribution for the systems presently considered is about three times the double-collision probability at small b, we conjecture that the single-collision process will also be important at small b for much lighter systems, probably even for systems far too light for our present model to be valid. On the other hand for large b, and consequently for the total K vacancy production cross sections, we can confirm that the double collision process is the dominant one for collisions in solids.

5. Conclusion

We have calculated the impact-parameter-dependent K vacancy production probability in a single-collision process assuming a temporal separation between the ionisation process and the $2p\pi$–$2p\sigma$ couplings, which is approximately true for relativistic systems. Our results show that the ionisation process considerably influences the impact parameter dependence, as compared with the pure coupling theory. Interference effects resulting from the coupling of the $2p\sigma$ and $2p\pi$ levels with different vacancy occupation numbers are not seen in the total K vacancy production probabilities due to the summation over all final states, but might conceivably be observable in multiply differential cross sections. The single-collision process is found to give a larger contribution than the double-collision process at small impact parameters for (I, Ag) collisions in solids, while the latter process dominates at large impact parameters and thus provides the total cross section. The combination of the two processes gives a considerably improved agreement between theory and experiment for impact parameter distributions. It is conjectured that the qualitative features of the model are rather generally valid, and further experimental results to test this would be very welcome.

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Appendix

In this appendix we show that the $2s\sigma$ state can be neglected for the transfer of a vacancy to the K shell in near-symmetric collisions. The potential coupling involving the diabatic $2s_{1/2}$ state,

$$V_\sigma \equiv V_{s_{1/2\pm1/2},p_{1/2\pm1/2}} = \mp f_1 R^{2\gamma+1}$$
$$V_\pi \equiv V_{s_{1/2\pm1/2},p_{3/2\pm1/2}} = f_2 R^{\gamma_1+\gamma} \tag{A.1}$$

where

$$f_1 = \left(\frac{2Z}{(2\gamma+2)^{1/2}}\right)^{2\gamma+2} \frac{(Z_1\alpha^{2\gamma+1} - Z_2\beta^{2\gamma+1})}{2\Gamma(2\gamma+1)(2\gamma+2)^{1/2}(2\gamma+1)^{1/2}(2\gamma+3)}$$

$$f_2 = \left(\frac{2Z}{(2\gamma+2)^{1/2}}\right)^{\gamma+1/2} Z^{\gamma_1+1/2} \left(\frac{(2\gamma+1)[1+\gamma+(2\gamma+2)^{1/2}](1+\gamma_1/2)}{4\Gamma(2\gamma+1)\Gamma(2\gamma_1+1)[1+(2\gamma+2)^{1/2}]}\right)^{1/2}$$
$$\times \frac{(3+\gamma_1-\gamma)}{(2+\gamma_1)} \frac{(Z_1\alpha^{\gamma_1+\gamma} - Z_2\beta^{\gamma_1+\gamma})}{(\gamma_1+\gamma+2)(\gamma_1+\gamma-1)}$$

with $\gamma = [1-(Z/c)^2]^{1/2}$ and $\gamma_1 = [4-(Z/c)^2]^{1/2}$, results from the dipole term of V_1 and thus vanishes for symmetric systems, i.e. for $\alpha = \beta$. For near-symmetric systems, $V_{\sigma,\pi}$ are very small, implying that the avoided crossings of the $2s\sigma$ state with the $2p\sigma$

state (at $R \sim 0.05\, a_L$ for I+Ag) and the $2p\pi(1/2)$ state (at $R \sim 0.2\, a_L$ for I+Ag) are very narrow. For an estimate of the transition probability to the $2p\sigma$ and $2p\pi(1/2)$ states the Landau–Zener formula may be applied (Zener 1932)

$$P_{LZ}^{\sigma(\pi)} = 1 - \exp(-2\pi\lambda_{\sigma(\pi)}) \tag{A.2}$$

$$\lambda_{\sigma(\pi)} = V_{\sigma(\pi)}^2 \left(\dot{R} \frac{\mathrm{d}}{\mathrm{d}R} \left| E_{p_{1/2(3/2),1/2}}(R) - E_{s_{1/2,1/2}}(R) \right| \right)^{-1} \Bigg|_{R=R_{cr}}$$

$$\dot{R} = \frac{vR_0}{2R} \left[\left(\frac{2R}{R_0} - 1 \right)^2 - 1 - \left(\frac{2b}{R_0} \right)^2 \right]^{1/2}$$

$$E_{s_{1/2,1/2}}(R) = E_{s_{1/2,1/2}}^{UA} + R^{2\gamma}(Z_1\alpha^{2\gamma} + Z_2\beta^{2\gamma}) \left(\frac{2Z}{(2\gamma+2)^{1/2}} \right)^{2\gamma+1}$$

$$\times \frac{[2+(2\gamma+2)^{1/2}]}{4\gamma\Gamma(2\gamma+1)[2+2\gamma+(2\gamma+2)^{1/2}]}$$

where $R_0 = 2Z_1Z_2/(\mu v^2)$ is the distance of closest approach in a head-on collision with μ the reduced mass, and R_{cr} is the crossing radius of the diabatic energy levels appearing in the denominator of λ. Any other quantities are defined in Jakubassa and Taulbjerg (1980). For a 63 MeV (I, Ag) collision ($v/c = 0.0325$), $P_{LZ}^{\pi} \simeq 8.4 \times 10^{-6}$ at $b = 0$ while the other crossing is not reached ($R_0 \simeq 0.1\, a_L$) such that the coupling to the p states can safely be neglected because the vacancy occupation number of the $2s\sigma$ and the $2p\pi$ states are of the same order of magnitude. This agrees with the result of Briggs and Taulbjerg (1975) for non-relativistic near-symmetric systems.

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