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Coupled channel treatment of the L-shell ionization in ion–atom collisions

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Abstract. The deviations of the experimental L-subshell ionization probabilities relative to the semiclassical approximation are discussed in terms of couplings between the target atomic states during the collision by means of a relativistic one-centre coupled channel formalism with hydrogenic wavefunctions. Numerical results are compared with experimental data for systems He+Pt, Ne+Pt, Ne+Yb at 0.9 MeV amu^{-1} and S+Au at 1 MeV amu^{-1}. A simple inclusion of the screening effect presented in this work yields a major improvement in the description of the L-shell ionization probabilities. The intra-L-shell couplings modify slightly the ionization probabilities for light projectiles but they have a marked effect for heavier projectiles. The couplings between the L and M shells were also included, but they were found to have only a small influence as compared with intra-L-shell couplings

1. Introduction

Involving the minimum number of subshells with different binding energies, angular momenta, spatial symmetries and degree of relativistic electron motion, the L shell of heavy elements provides a suitable environment for a detailed testing of the theories aimed to describe the collisionally induced inner-shell ionization. As for the K-shell ionization, the semiclassical approximation (SCA) (Kocbach 1980, Hansteen 1990) turns out to provide a satisfactory description of the L-subshell ionization cross sections for protons in a wide range of collision velocities (Jitschin 1984) and target elements, particularly when the calculations are done with Rutherford trajectories, relativistic electron wavefunctions and the binding correction is taken into account. For heavier projectiles, deviations occur between experimental and first-order theoretical cross section values, which increase with increasing projectile nuclear charge and decreasing velocity. These deviations can exceed one order of magnitude even for collision systems asymmetric enough that direct (Coulomb) excitation to the continuum is the only mechanism of inner-shell ionization and its perturbative treatment is expected to be a tenable approximation.

In principle, before making a comprehensive comparison between theory and the inclusive experiments currently performed, one has to consider the contribution to the vacancy production of the electron capture to the projectile (McDaniel *et al* 1979, Berinde *et al* 1985) and the effect of multiple ionization (Uchai *et al* 1985, Piticu *et* *al* 1987) on the L-vacancy decay (Berinde *et al* 1987a). It has been shown (Berinde *et al* 1987b) that the disagreement persists both after subtraction of the electron capture contribution from the measured L-subshell vacancy production cross sections and after the correction for the influence of simultaneous multiple ionization of the outer (M, N, O) shells on the atomic parameters used to transform the L x-ray yields into subshell vacancy production cross sections. Similar discrepancies relative to SCA are present in many other experimental data on the L-subshell cross sections using heavier projectiles (e.g. Li *et al* 1976, Palinkas *et al* 1983, Jitschin 1984, Berinde *et al* 1988a, b).

Similar to the K-shell ionization (Andersen *et al* 1982), the impact parameter dependence of the ionization probabilities is expected to be more useful to test the theories aimed at describing the L-shell ionization, and in particular to understand the dynamic behaviour of the electron wavefunction during the collision (Schmidt-Böcking *et al* 1987a). Unfortunately, such data are still scarce in the literature (Laegsgaard *et al* 1974, Stiebing *et al* 1976, Andriamonje *et al* 1981, Konrad *et al* 1984, Nolte *et al* 1984, Berinde *et al* 1984, Ullrich *et al* 1986, Dexheimer *et al* 1986, Zehendner *et al* 1987, Schmidt-Böcking *et al* 1987b). Nevertheless, the existing differential data clearly show disagreement with the first-order ionization theories, similar or even greater than that shown by the integral cross sections. A mechanism of vacancy sharing between the L subshells during the collision has been proposed by Sarkadi and Mukoyama (1981) to explain these discrepancies. This vacancy sharing effect was first studied qualitatively in a simple two-step model, ionization and subshell couplings being treated as two independent, successive processes. The model has been refined by Finck *et al* (1983), Sarkadi and Mukoyama (1984), Sarkadi (1986a), Berinde *et al* (1988b) and Legrand *et al* (1989).

The vacancy sharing mechanism can be properly described within a coupled-channel formalism. Coupled-state calculations of the L-shell impact ionization have been made by Martir *et al* (1982), using a target-centred expansion and pseudostates for the continuum. Since only global L-shell ionization cross sections are presented, no information on the intrashell couplings can be extracted from their results. It is worth mentioning, however, that in order to reproduce the magnitude of the cross sections for the collisions investigated, Martir *et al* modified the target atom potential to bring the L-shell removal energies closer to the experimental ionization potentials, whilst leaving the wavefunctions unchanged. Another full coupled-channel calculation is due to Mehler *et al* (1987), using wavepackets for the continuum states. They have shown that for very asymmetric collisions the magnitude of the L-subshell vacancy production is not very sensitive to the couplings in the continuum. Thus one may try to calculate the coupling to the continuum perturbatively, while treating the couplings between the L subshells in a close coupling approach. This scheme was called the 'coupled subshell approximation' (CSA) (Amundsen and Jakubassa-Amundsen 1988). Simplified CSA calculations have been reported by Spies *et al* (1984), Kocbach (1984), Berinde *et al* (1985), Sarkadi (1986b) and Sarkadi and Mukoyama (1987, 1990). These calculations, like the two-step model, provided strong arguments for the existence of intra-L-shell couplings during the collision. However, in most cases this conclusion was achieved by comparing with the experimental data only relative subshell ionization cross sections or probabilities. Even the absolute cross sections of Sarkadi and Mukoyama were obtained by multiplying the SCA cross sections with a correction factor which is essentially also a ratio of cross sections.

Absolute CSA calculations have been recently published by Amundsen and Jakubassa-Amundsen (1988). These calculations reproduce the impact parameter dependence of the L-subshell ionization probabilities much better than SCA, but for heavier projectiles CSA seriously underestimates the magnitude of the experimental data. The same is true also for the more complete coupled-channel calculations of Mehler (1987). The aim of the present work is to improve the CSA model for the L-shell ionization with vacancy sharing in order to achieve a better description of the absolute value of the subshell ionization probabilities. More specifically, we accounted in a simple way for the screening effect of the spectator electrons upon the couplings strength. We also performed coupled-channel calculations including both the L and M shells, to test the importance of the intershell couplings.

2. Theory

The physical assumptions in what follows are similar to those used by Amundsen and Jakubassa-Amundsen (1988). Accordingly, the collision systems investigated were chosen asymmetric enough so that no quasimolecular orbital correlating to the projectile intereacts with those correlating to the target L shell. The reference frame used is centred on the target nucleus and the incoming beam direction is chosen for the quantization axis. The internuclear motion is described by a classical Rutherford trajectory. Concerning the electronic problem, it will be convenient to consider the vacancy instead of the electron as the active 'particle' of the collision process. The one-particle problem is described by the two-centre Dirac Hamiltonian:

$$H = -\mathrm{i}c\hbar\boldsymbol{\alpha}\cdot\boldsymbol{\nabla} + \beta mc^2 - \frac{Z_{\mathrm{T}}e^2}{r} - \frac{Z_{\mathrm{P}}e^2}{|\boldsymbol{r}-\boldsymbol{R}|} \tag{1}$$

where $\boldsymbol{R}$ is the internuclear vector, while Z_{T} and Z_{P} are the target and projectile atomic numbers, respectively. The recoil potential arising because the coordinate system centred on the target is not inertial, has been neglected, being unimportant for L-shell ionization. The scattering wavefunction in the time-dependent Dirac equation ($e = \hbar = c = 1$):

$$\mathrm{i}\frac{\partial\Psi(\boldsymbol{r}, t)}{\partial t} = H\Psi(\boldsymbol{r}, t) \tag{2}$$

for a vacancy initially in a given continuum state f, can be expanded as

$$\Psi(\boldsymbol{r}, t) = \sum_m a_{mf}(t, E)\psi_m(\boldsymbol{r})\,\mathrm{e}^{-\mathrm{i}t\varepsilon_m} + \psi_f(\boldsymbol{r}, E)\,\mathrm{e}^{-\mathrm{i}tE} \tag{3}$$

in terms of the bound (ψ_m) and continuum (ψ_f) hydrogen-like wavefunctions, with energies ε_m and E, respectively. Thus, the time-dependent Dirac equation (2) reduces to a system of coupled equations for the vacancy amplitudes in the bound states:

$$\begin{aligned}\frac{\mathrm{d}a_{nf}(t, E)}{\mathrm{d}t} &= -\mathrm{i}\sum_m a_{mf}(t, E)\langle\psi_n(\boldsymbol{r})|\,V(\boldsymbol{r}, \boldsymbol{R})|\psi_m(\boldsymbol{r})\rangle\,\mathrm{e}^{-\mathrm{i}t(\varepsilon_m-\varepsilon_n)}\\ &\quad + \langle\psi_n(\boldsymbol{r})|\,V(\boldsymbol{r}, \boldsymbol{R})|\psi_f(\boldsymbol{r}, E)\rangle\,\mathrm{e}^{-\mathrm{i}t(E-\varepsilon_n)}\end{aligned} \tag{4}$$

with the boundary condition $a_{nf}(t = -\infty) = 0$. Here $V(\boldsymbol{r}, \boldsymbol{R})$ denotes the perturbing potential due to the projectile:

$$V(\boldsymbol{r}, \boldsymbol{R}) = -\frac{Z_{\mathrm{P}} e^2}{|\boldsymbol{r} - \boldsymbol{R}|}. \tag{5}$$

When writing equations (3) and (4) we assumed that the continuum states are all filled with vacancies during the collision (the amplitude for the continuum state in equation (3) is 1 all over the collision), i.e. $a_{nf} \ll 1$, and we neglected the back coupling and the continuum-continuum couplings. This is equivalent to treating the continuum in the first order of the perturbation theory. The approximation seems to be justified for the asymmetric collisions under study, where the total L-shell ionization probabilities are not too large and are relatively well reproduced by the first-order perturbation theory (Dexheimer *et al* 1986). Accordingly, in the system of coupled equations (4), the continuum-bound matrix elements play the role of a source term for the close coupling among the bound states.

In the radial part of the matrix elements of equation (4) the perturbing potential (5) has been expanded in multipoles (Kocbach *et al* 1980), the number of terms in the expansion being limited only by the angular momentum selection rules. The continuum-bound matrix elements have been computed using continuum Dirac hydrogenic wavefunctions generated numerically (Barnett 1981). The bound-bound matrix elements have been expressed as a convergent power series. The angular part in all matrix elements has been calculated according to the well known angular momentum algebra.

If the bound-bound couplings are neglected in the equations (4), one obtains the SCA amplitudes for ionization (Kocbach 1980). In order to see the effect of the vacancy transfer between the bound states, we also performed SCA calculations in the united atom (UA) approximation (Briggs 1975). To approximately include the screening, the binding energies ε_n are taken as the experimental binding energies of the UA and the source terms in equation (4) are calculated with hydrogenic wavefunctions seeing the central charge $Z_{\mathrm{T}} + Z_{\mathrm{P}} - 4.15$. Consistently, in the coupled channel calculations we used the same source terms, while the bound-bound matrix elements are calculated with target wavefunctions, using for ε_n the experimental target binding energies.

The diagonal matrix element

$$V_{nn} = \langle \psi_n(\boldsymbol{r}) | V(\boldsymbol{r}, \boldsymbol{R}) | \psi_n(\boldsymbol{r}) \rangle \tag{6}$$

represents, in the first order of the perturbation theory, the 'binding correction'. For $R = 0$ one is expecting

$$V_{nn}(R = 0) = \varepsilon_n^{\mathrm{UA}} - \varepsilon_n^{\mathrm{SA}} \tag{7}$$

where $\varepsilon_n^{\mathrm{UA}}$ is the binding energy of the 'united atom' (having the atomic number $Z_{\mathrm{T}} + Z_{\mathrm{P}}$) and $\varepsilon_n^{\mathrm{SA}}$ is the binding energy of the target (Z_{T}). However, in the hydrogenic approximation underlying the above treatment, we have found that $V_{nn}(R = 0)$ is 20-40% larger than the difference of the experimental binding energies for the systems under investigation. This would lead to a significant reduction of the calculated ionization probabilities and is very likely to be the reason for the severe underestimation by Mehler (1987) and Amundsen and Jakubassa-Amundsen (1988) of the experimental values measured with heavier ions. In order to approximately account for the screening effect of the spectator electrons we renormalized the matrix elements in such a way that condition (7) with experimental values for the binding energies is fulfilled for all bound states.

For a given impact parameter b, the equations (4) are integrated along the trajectory $R(t)$, separately for every accessible continuum state labelled by the quantum number κ_f, j_f, m_f and kinetic energy E. The resulting probabilities are summed incoherently to give the observed L-subshell ionization probability for the subshell n:

$$P_n(b)=\sum_f \int_{E=0}^{E_{\max}} |a_{nf}(t=+\infty, E)|^2 \, dE. \tag{8}$$

In the present calculations (both SCA and CSA) the integral over kinetic energy has been extended up to $E_{\max}=150$ keV and the sum over the continuum states up to $f_{7/2}$.

3. Results and discussions

The above theoretical approach has been applied to the description of the L-subshell ionization probabilities for the systems He + Pt, Ne + Pt and Ne + Yb at 0.9 MeV amu^{-1} bombarding energy. The results are compared with the detailed experimental data of Dexheimer *et al* (1986) in figures 1-3. In order to see the vacancy sharing effects, we also performed SCA-UA-like calculations, corresponding to the system of equations (4) when the bound-bound couplings are neglected. As can be seen in figure 1, in the case of very asymmetric systems the intrashell coupling is not very important for the L-subshell ionization, but nevertheless it has the effect of improving the agreement between theory and experiment.

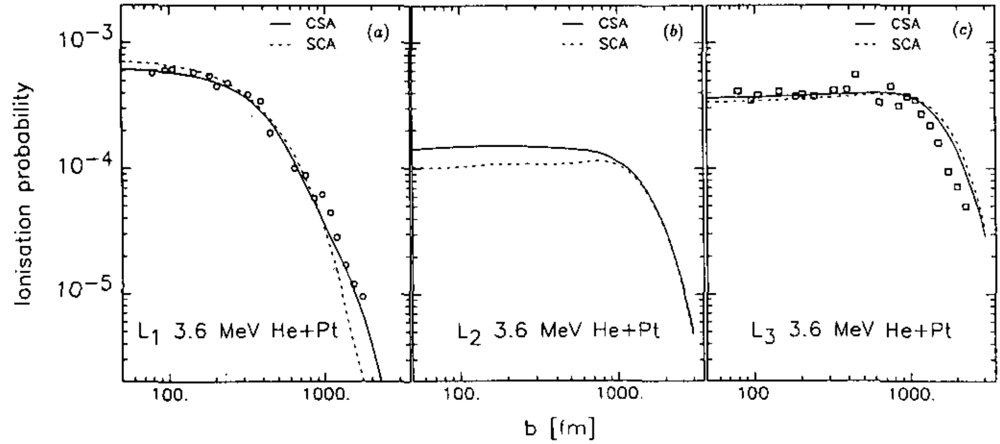

Figure 1. L-subshell ionization probabilities as a function of the impact parameter for 0.9 MeV amu^{-1} He + Pt: (*a*) L_1; (*b*) L_2; (*c*) L_3. Experimental data (L_1, circles; L_3, squares) from Dexheimer *et al* (1986). Theory: ——, CSA results; ····, SCA-UA results.

For the description of heavier projectile data the inclusion of the intrashell couplings is important. In figure 2 are shown the results for Ne + Pt. The data are almost quantitatively described by our coupled subshell calculations. In particular, the calculated $P_{L_1}(b)$ and $P_{L_3}(b)$ do not cross at small b, as they do for the He + Pt system, in agreement with experiment. Also, the slope of $P_{L_1}(b)$ at large b is much better reproduced. A few conspicuous features of the intrashell couplings can be deduced from an inspection of figure 2. Thus, the evolution of the vacancy flux inside the L

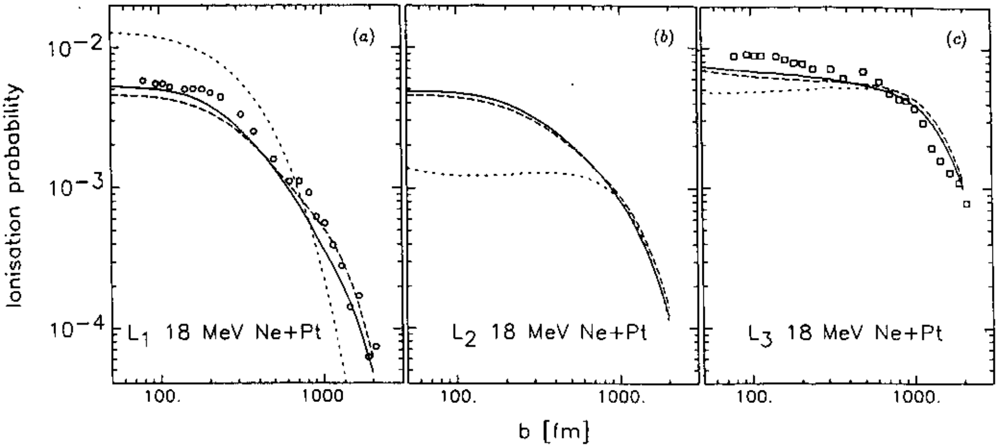

Figure 2. L-subshell ionization probabilities as a function of the impact parameter for 0.9 MeV amu^{-1} Ne+Pt: (*a*) L_1; (*b*) L_2; (*c*) L_3. Experimental data (L_1, circles; L_3, squares) from Dexheimer *et al* (1986). Theory: ——, CSA results; ····, SCA-UA results; ----, coupled channel calculations using L and M shells.

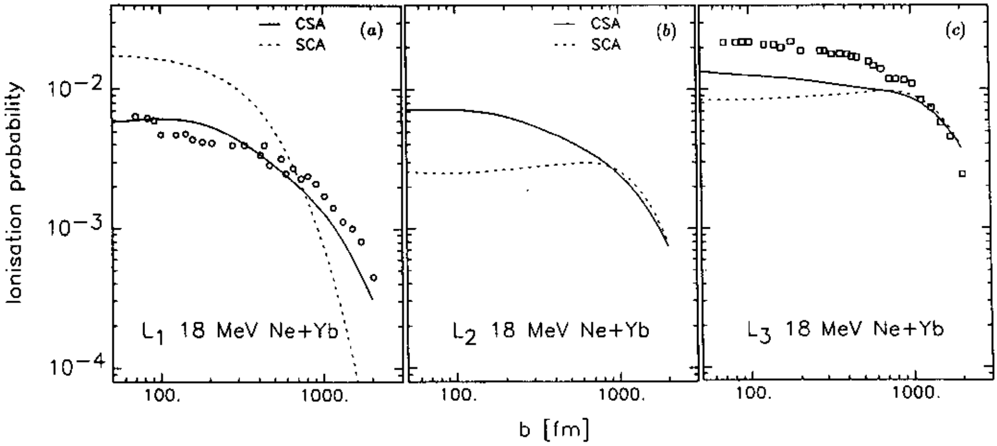

Figure 3. Same as figure 1, but for 0.9 MeV amu^{-1} Ne+Yb.

shell is largely governed by its gradient, similar to a diffusion process. According to SCA, at small impact parameters the L_1 subshell ionization dominates due to the relativistic enhancement. The couplings, however, share the vacancies almost statistically between the L_1 and L_2 subshells, and to a less extent with L_3 (presumably because of the latter being more distant in energy). The opposite occurs at large impact parameters, where the fall-off of the L_1 primary ionization is compensated by the transfer of vacancies from the higher subshells, yielding a much better agreement with the experiment. In the processes occurring at small impact parameters, the L_2-subshell ionization probability is strongly enhanced due to the coupling with the L_1 subshell.

The experimental and theoretical results for the Ne + Yb are compared, as a function of impact parameter, in figure 3. Again, the CSA calculations represent a much better description relative to the SCA. However, the tendency to underestimate the L_3 subshell ionization probability at small impact parameters, already present for Ne + Pt, is more pronounced for Ne + Yb. One may ask whether part of the vacancies created in the M shell (with a much larger probability) are transferred in the same collision to the L shell, and in particular to the L_3 subshell. In order to check this effect, CSA calculations were done including the M shell in our basis set, the rest of the procedure remaining the same. The results for Ne + Pt are shown with a broken curve in figure 2; for Ne + Yb the effect of the coupling with the M shell is even smaller. Corroborating these results with the fact that CSA gives practically the same value as SCA for the ionization probability of the L shell as a whole, one may conclude that the coupling among the major shells is small compared with the intrashell coupling. This is a fortunate circumstance, allowing us to speak about the ionization of a specific shell instead of the entire atom.

The L_3- to L_1-subshell ionization probability ratios are shown in figure 4, where we added the data for S + Au at 1 MeV amu^{-1} (Berinde *et al* 1984). It is worth mentioning that the latter were corrected for outer-shell ionization effects. Again the CSA provides a much better description of the data relative to SCA. The improvement is similar to that provided by the CSA calculation of Amundsen and Jakubassa-Amundsen (1988). However, concerning the absolute values of the ionization probabilities, our calculations are much closer to the data. Most likely, this is due to a better account, in our approach, of the electron binding energy and coupling strength variation during the collision.

To summarize, the CSA with the simple inclusion of the screening effect presented in this work yields a major improvement of the description of the L-subshell ionization probabilities relative to SCA. As a consequence, the integral cross sections (Berinde *et al* 1988a) are also much better reproduced by the present approach (Legrand *et al* 1990). However, some discrepancies between the theory and experiment, such as the underestimation of the $P_{L_3}(b)$ at small impact parameters, still remain unexplained within this model of direct vacancy production and sharing. A possible origin of these discrepancies is the projectile induced rotation of the target electron distribution during the collision (Legrand *et al* 1991).

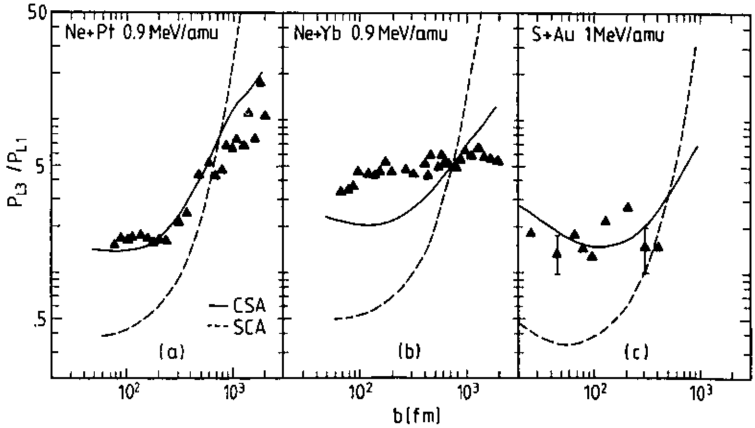

Figure 4. Impact parameter dependence of the ionization probabilities ratio $P_{L_3}(b)/P_{L_1}(b)$. Theory: ——, CSA; ----, SCA-UA. Experimental points: (*a*) Ne + Pt at 0.9 MeV amu^{-1} from Dexheimer *et al* (1986); (*b*) Ne + Yb at 0.9 MeV amu^{-1} from Dexheimer *et al* (1986); (*c*) S + Au at 1 MeV amu^{-1} from Berinde *et al* (1984).

In order to achieve a complete understanding of the L-shell ionization in moderately asymmetric collisions more differential data are needed, particularly for the L_2 subshell, as well as CSA calculations including the electronic wavefunction rotation.

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