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# Positronium formation in positron–simple metal clusters collisions

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## Abstract

Positronium formation through electron capture from ground-state closed-shell sodium clusters is studied theoretically at intermediate impact energies. The charge transfer process is described in the independent electron model by a continuum distorted-wave approximation. In this approximation, distortions in the final state are introduced by using two Coulomb wavefunctions associated with the positron- and electron-residual target interactions. The cluster is described within the framework of the spherical background jellium model and the Kohn–Sham formalism with a local-density approximation that includes exchange, correlation, and a self-interaction correction. Using a partial-wave technique, differential and total cross sections for the isoelectronic neutral and charged closed-shell sodium clusters  $\text{Na}_{20}$ ,  $\text{Na}_{19}^-$  and  $\text{Na}_{21}^+$  are computed and compared with the predictions of the Coulomb–Born type approximation which neglects the distortions in the final state.

## 1. Introduction

Collisions of positrons impacting on atoms (ions) have been the subject of active research over the last few years, partly motivated by the measurements obtained with the presently available high-intensity positron beams. There are interesting reviews on the issue (see, for instance, [1, 2]).

A continuum distorted-wave final-state (CDW-FS) approximation has been developed to study positron–hydrogenic atom (ion) [3–5] and positron–alkali atom collisions [6]. A four-body CDW-FS (CDW-FS-4b) model has been developed recently to study the collision positron–alkali-earth metal [7] and positron–metastable helium [8]. In CDW-FS, the final state of the collision is distorted by two Coulomb wavefunctions associated with the positron- and electron residual target interactions. In this way, higher-order terms are included to describe the collision process.

Contrary to the inelastic electron–metallic clusters interactions which have been intensively studied over the last decade [9, 10], to the best of our knowledge, no theoretical work has been devoted to the study of positron–metallic cluster collisions with positronium formation.

Since closed-shell metal clusters are practically spherical, the problem has the same global symmetry as in positron–atom collisions. Thus one can use the same approximations and techniques used in atomic collision physics. As a matter of fact, collision of slow protons and  $\text{Na}_{20}$  cluster targets have been studied in this way [11].

The positronium formation reaction in collision of positron with closed-shell metal clusters is a problem of many active electron. As for proton projectiles [11], an independent electron model (IEM) is used for the resolution of this problem.

Our goal is to obtain theoretical results for total and differential cross section (TCS and DCS, respectively) in order to simulate future experiments. In addition, positronium formation may be used to test the validity of models used to describe the cluster structure.

Our theoretical approach is outlined in the next section. Section 2.1 is devoted to the cluster description. Concerning the description of the collision process, the basic formalism of the CDW-FS formalism is presented in section 2.2. Our results are presented and discussed in section 3. Finally, some perspectives as well as a conclusion are given in section 4. Atomic units are used unless specified otherwise.

## 2. Theoretical method

### 2.1. Cluster description

The cluster of  $N$  valence electrons is described in the spherical background jellium model (SJM) which has been successfully applied to the study of ground-state properties of small closed-shell simple metal clusters [12]. This model consists in replacing the real ionic core potential by a constant positive background of spherical symmetry corresponding to a uniformly distributed charge density and is expected to work better for a temperature above the melting temperature of the cluster. Indeed, on average, the spherical symmetry of the ionic background imposed in the SJM simulates well the liquid structure of the ions within the cluster. According to a quantal tight-binding Hamiltonian method combined with Monte Carlo thermodynamical simulations, Poteau *et al* [13] have shown that  $\text{Na}_{20}$  melts around 290 K. One may expect a small deviation of the value of this temperature for charged species [14]. As a typical experiment involving free positively charged simple metal clusters (e.g.  $\text{Na}_{21}^+$ ), the experiment of the Orsay group is carried out in the temperature range  $T = 250\text{--}600$  K [15]. Thus, most of the clusters present in the beam have a liquid structure. Furthermore, in usual experimental conditions, the clusters are not oriented in space and the comparison between measurements and theoretical predictions must be done by averaging the latter over all possible cluster orientations, i.e. over all possible ion positions.

For a metal cluster having  $A$  singly charged ionic cores, this potential is given by

$$V_{\text{jel}}(r) = \begin{cases} -\frac{A}{2R_C} \left[ 3 - \left( \frac{r}{R_C} \right)^2 \right] & \text{for } r \leq R_C \\ -\frac{A}{r} & \text{for } r > R_C \end{cases} \quad (1)$$

where  $R_C = A^{1/3}r_s$  and  $r_s$  is the Wigner–Seitz radius (for example, in the case of  $\text{Na}_{20}$   $r_s = 4$  and  $R_C = 10.86$ ). In the Kohn–Sham formulation of the density functional theory, the ground-

state electronic density  $\rho_C$  of an  $N$ -electron system is written in terms of single-particle orbitals  $\varphi_i$ :

$$\rho_C(\mathbf{r}) = \sum_{i=1}^N \rho_i(\mathbf{r}) = \sum_i |\varphi_i(\mathbf{r})|^2. \quad (2)$$

These orbitals obey the Schrödinger equation

$$\left[-\frac{1}{2}\nabla^2 + V_{KS}(\mathbf{r})\right] \varphi_i(\mathbf{r}) = \epsilon_i \varphi_i(\mathbf{r}) \quad (3)$$

where  $V_{KS}(\mathbf{r})$  is an effective single-particle potential given by

$$V_{KS}(\mathbf{r}) = V_{jel}(\mathbf{r}) + V_H(\rho_C(\mathbf{r})) + V_{xc}(\rho_C(\mathbf{r})) \quad (4)$$

where  $V_H(\rho_C(\mathbf{r}))$  is the Hartree potential and  $V_{xc}(\rho_C(\mathbf{r}))$  is the exchange-correlation potential. Since the form of  $V_{xc}$  is not known in general, several approximations have been proposed in the literature. In this work, we have used the form obtained by Gunnarsson and Lundqvist [16] within the framework of the local-density approximation (LDA):

$$V_{xc}(\rho_C(\mathbf{r})) = -\left(\frac{3\rho_C(\mathbf{r})}{\pi}\right)^{1/3} - 0.0333 \log\left(1 + \frac{11.4}{r_s(\mathbf{r})}\right) \quad (5)$$

where  $r_s(\mathbf{r}) = [3/4\pi\rho_C(\mathbf{r})]^{1/3}$  is the local Wigner–Seitz radius. For a neutral cluster, the asymptotic behaviour of  $V_{KS}$  is given by the exchange contribution to  $V_{xc}$ , which behaves at large distance as  $\rho_C(\mathbf{r})^{1/3}$ . Consequently, the Kohn–Sham potential  $V_{KS}$  decreases exponentially to zero, i.e. it does not reproduce the correct  $1/r$  asymptotic behaviour. This problem does not appear in Hartree–Fock (HF) theory, because the HF exchange potential exactly compensates the self-interaction term contained in the Hartree potential. Following Perdew and Zunger [17], we have added a self-interaction correction (SIC) that restores the correct asymptotic behaviour of the potential (we will call this method LDA-SIC).

## 2.2. CDW-FS formalism

Let us consider the electron capture process with positronium formation in positron–simple metal cluster collisions of  $N$  valence electrons at intermediate impact energies. Within the framework of the IEM model, the *prior* form of the CDW-FS matrix element for one valence initial cluster orbital may be written as

$$T_{\alpha\beta}^- = \langle \chi_\beta | V_\alpha | \Phi_\alpha \rangle \quad (6)$$

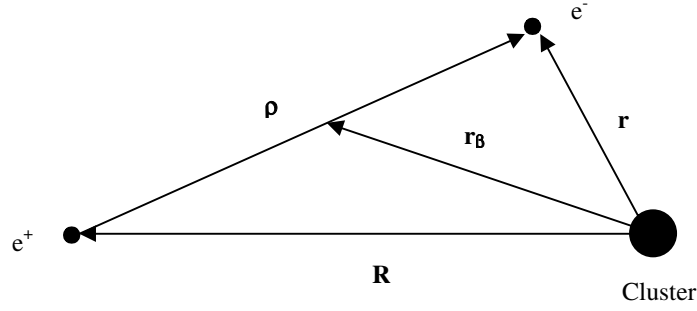
with

$$\Phi_\alpha = N_{v_\alpha}^+ \varphi_i(\mathbf{r}) \exp(i\mathbf{k}_\alpha \cdot \mathbf{R}) {}_1F_1(-iv_\alpha'; 1; -i\mathbf{k}_\alpha \cdot \mathbf{R} + i\mathbf{k}_\alpha R) \quad (7)$$

and

$$\begin{aligned} \chi_\beta = N_{\beta_+}^- N_{\beta_-}^- \varphi_f(\rho) \exp(-i\mathbf{k}_\beta \cdot \mathbf{r}_\beta) \\ \times {}_1F_1\left(i\beta_+; 1; -\frac{i}{2}\mathbf{k}_\beta \cdot \mathbf{R} - \frac{i}{2}k_\beta R\right) {}_1F_1\left(-i\beta_-; 1; -\frac{i}{2}\mathbf{k}_\beta \cdot \mathbf{r} - \frac{i}{2}k_\beta r\right). \end{aligned} \quad (8)$$

Coordinates are sketched in figure 1.  $\varphi_i$  is the initial orbital bound wavefunction of the captured valence electron of the cluster and is given by equation (3).  $\varphi_f$  is the final bound wavefunction of the positronium atom (only the ground-state is considered in this paper).  $\mathbf{k}_\alpha$  and  $\mathbf{k}_\beta$  are the



**Figure 1.** Coordinates used in the text.

wavevectors for the reduced positron in the entry channel and for the reduced positronium atom in the final channel, respectively. Moreover, the following quantities have been defined [4]:

$$\beta_+ \simeq \beta_- = \frac{(Z+1)\mu_\beta}{k_\beta} = \frac{(Z+1)\mu_\alpha}{k_\pm} \quad (9)$$

$$v'_\alpha = \frac{Z\mu_\alpha}{k_\alpha} \quad (10)$$

where  $Z$  is the net charge of the target and the reduced masses are  $\mu_\alpha \simeq 1$  and  $\mu_\beta \simeq 2$ .

Moreover,

$$N_{\beta_\pm}^- = \Gamma(1 \mp i\beta_\pm) \exp\left(\mp \frac{\pi}{2}\beta_\pm\right) \quad (11)$$

$$N_{v'_\alpha}^+ = \Gamma(1 + iv'_\alpha) \exp\left(-\frac{\pi}{2}v'_\alpha\right). \quad (12)$$

The perturbation potential is given by

$$V_\alpha = \frac{1}{R} - \frac{1}{\rho} + V_P(R) \quad (13)$$

where  $V_P$  is the short-range part of the positron–residual target interaction. In this work, the short-range potential  $V_P^j(R)$  associated with the cluster orbital labelled  $j$  is taken as

$$V_P^j(R) = -V_{\text{jel}}(R) - \sum_{i=1; i \neq j}^N V_H(\rho_i(r)) - \frac{(Z+1)}{R}. \quad (14)$$

One may greatly simplified the scattering problem by using, instead of the above state-dependent potentials, an average potential given by

$$V_P(R) = \frac{1}{N} \sum_{j=1}^N V_P^j(R). \quad (15)$$

This procedure has been used in [11] for the description of the electronic ground state of  $\text{Na}_{20}$ . The authors of [11] have shown that the eigenenergies of the occupied orbitals of  $\text{Na}_{20}$  obtained by using this averaging procedure are practically identical to those calculated with the state-dependent effective potentials. Very recently, on the basis of *ab initio* calculations, Ullrich *et al* [18] have confirmed the validity of such an approach for the description of static and dynamical properties of simple metal clusters. This approximation will be justified in section 3 on a particular example.

Then, the *prior* version of the CDW-FS matrix element reads [4, 5]

$$\begin{aligned}
T_{\alpha\beta}^- &= N_{\nu_\alpha}^+ N_{\beta_+}^{-*} N_{\beta_-}^{-*} \int d\mathbf{R} d\mathbf{r} \exp\{i\mathbf{k}_\alpha \cdot \mathbf{R} + i\mathbf{k}_\beta \cdot \mathbf{r}\} \varphi_f^*(\rho) \\
&\quad \times \left( \frac{1}{R} - \frac{1}{\rho} + V_P(R) \right) {}_1F_1\left(-i\beta_+; 1; \frac{i}{2}\mathbf{k}_\beta \cdot \mathbf{R} + \frac{i}{2}k_\beta R\right) \\
&\quad \times {}_1F_1\left(i\beta_-; 1; \frac{i}{2}\mathbf{k}_\beta \cdot \mathbf{r} + \frac{i}{2}k_\beta r\right) {}_1F_1(-i\nu'_\alpha; 1; -i\mathbf{k}_\alpha \cdot \mathbf{R} + i k_\alpha R) \varphi_i(\mathbf{r}). \quad (16)
\end{aligned}$$

In order to evaluate the CDW-FS matrix element, a partial-wave expansion technique introduced in [4] has been used.

The initial valence electronic orbital that is a solution of equation (3) is written as

$$\varphi_i(\mathbf{r}) = R_{n_t, l_t}(r) Y_{l_t, m_t}(\hat{\mathbf{r}}) \quad (17)$$

where  $n_t$ ,  $l_t$  and  $m_t$  are the orbital quantum numbers. The final wavefunction is given by

$$\varphi_f(\rho) = \frac{1}{\sqrt{2}} \exp(-\rho/2) \quad Y_{00}(\hat{\rho}) = \frac{1}{\sqrt{8\pi}} \exp(-\rho/2). \quad (18)$$

The DCS for a captured electron of quantum numbers  $(n_t, l_t, m_t)$  reads

$$\left[ \frac{d\sigma}{d\Omega} \right]_{n_t, l_t, m_t} = \frac{1}{4\pi^2} \frac{k_\beta}{k_\alpha} \mu_\alpha \mu_\beta \left| T_{\alpha\beta}^- \right|^2 \quad (19)$$

with

$$\left| T_{\alpha\beta}^- \right|^2 = \frac{(4\pi)^3}{(k_\alpha k_+ k_-)^2} \hat{l}_t \left| \sum_{l_i L} i^{l_i} e^{i\delta_{l_i}} \hat{l}_i \hat{L}^{1/2} (-1)^L S_{l_i L} Y_{L, m_t}(\hat{k}_\beta) \right|^2 \quad (20)$$

where the notation  $\hat{l} = 2l + 1$  has been used. Moreover, we have defined

$$S_{l_i L} = \sum_{l' l_f} i^{-l-l_f} e^{i(\delta_{l'} + \delta_{l_f})} (-1)^{l'} \mathcal{A}_{l_i L}^{l' l_f} \mathcal{R}_{l_i l_f}^{l'} \quad (21)$$

with

$$\begin{aligned}
\mathcal{A}_{l_i L}^{l' l_f} &= \hat{l} \hat{l}' \hat{l}_f \begin{pmatrix} l_t & l & l' \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l_i & l' & l_f \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l & l_f & L \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l_i & L & l_t \\ 0 & -m_t & m_t \end{pmatrix} \\
&\quad \times \left\{ \begin{matrix} l_i & L & l_t \\ l & l' & l_f \end{matrix} \right\}
\end{aligned}$$

$$\mathcal{R}_{l_i l_f}^{l'} = \int_0^\infty F_{l_i}(k_\alpha R) \mathcal{V}_{l' l_f}(R) F_{l_f}(k_+ R) dR \quad (22)$$

$$\mathcal{V}_{l' l_f}(R) = \int_0^\infty r R_{n_t, l_t}(r) J_{l'}(r; R) F_{l_f}(k_- r) dr \quad (23)$$

$$J_{l'}(r; R) = \frac{1}{2} \int_{-1}^{+1} \tilde{R}_{1s}(\rho) \left( \frac{1}{R} - \frac{1}{\rho} + V_P(R) \right) P_{l'}(u) du \quad (24)$$

and

$$\rho = (r^2 + R^2 - 2rRu)^{1/2}. \quad (25)$$

The functions  $F_l(k_{\pm}r)$  and  $F_l(k_{\alpha}r)$  are the Coulomb radial wavefunctions with the Sommerfeld parameters  $\eta = \beta_{\pm}$  and  $\eta = \nu'_{\alpha}$ , respectively. The phase shifts,  $\delta_l$  are the usual Coulomb phase shifts  $\delta_l = \arg \Gamma(l + 1 + i\eta)$ .  $P_l$  indicates the Legendre polynomial of degree  $l$ . The matrix element in the so-called Coulomb–Born-approximation (CBA) is obtained by setting  $\beta_+ = \beta_- = 0$ . In this approximation, the distortions in the final state are ignored.

For a closed shell of quantum numbers  $(n_i l_i)$ , the DCS is given by

$$\left[ \frac{d\sigma}{d\Omega} \right]_{n_i l_i} = 2 \sum_{m_i} \left[ \frac{d\sigma}{d\Omega} \right]_{n_i l_i m_i} \quad (26)$$

and the TCS is

$$[\sigma]_{n_i l_i} = 2 \frac{16\pi \mu_{\alpha} \mu_{\beta}}{k_{\alpha}^3 (k_+ k_-)^2} \hat{l}_i \sum_{l_i L} \hat{l}_i \hat{L} \tilde{S}_{l_i L} \tilde{S}_{l_i L}^* \quad (27)$$

with

$$\tilde{S}_{l_i L} = \sum_{l' l'_f} i^{-l-l'_f} e^{i(\delta_l + \delta_{l'_f})} (-1)^{l'} \tilde{\mathcal{A}}_{l_i L}^{l' l'_f} \mathcal{R}_{l_i l'_f}^{l' l'_f} \quad (28)$$

and

$$\tilde{\mathcal{A}}_{l_i L}^{l' l'_f} = \hat{l}_i \hat{l}_f \begin{pmatrix} l_i & l & l' \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l_i & l' & l_f \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l & l_f & L \\ 0 & 0 & 0 \end{pmatrix} \begin{Bmatrix} l_i & L & l_i \\ l & l' & l_f \end{Bmatrix}.$$

The factor of two in the expressions (26) and (27) takes into account of the spin degeneracy. Finally, total cross sections are obtained from  $\sigma_{n_i l_i}$  and  $[d\sigma/d\Omega]_{n_i l_i}$  by summing over all the initial states

$$\frac{d\sigma}{d\Omega} = \sum_{n_i l_i} \left[ \frac{d\sigma}{d\Omega} \right]_{n_i l_i} \quad (29)$$

$$\sigma = \sum_{n_i l_i} [\sigma]_{n_i l_i}. \quad (30)$$

### 3. Results

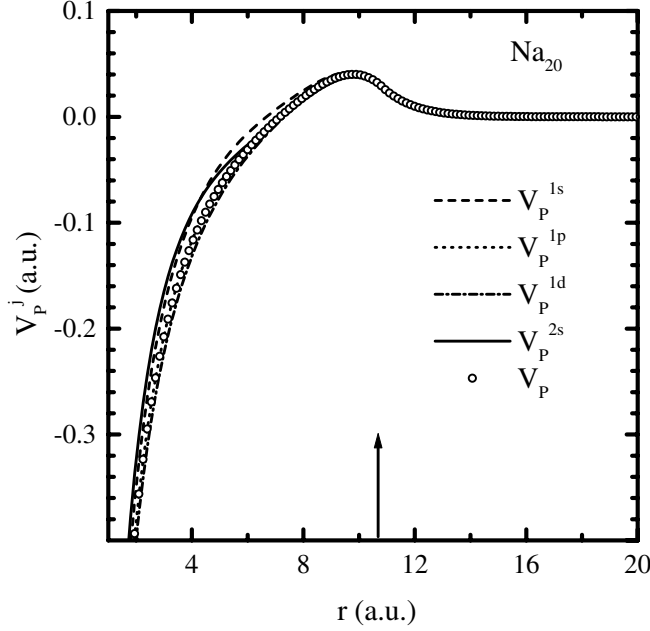
CDW-FS and CBA differential and total cross sections for positronium formation from ground-state closed-shell sodium clusters as a function of the positron impact energy are given. We have studied the isoelectronic series  $\text{Na}_{19}^-$ ,  $\text{Na}_{20}$ ,  $\text{Na}_{21}^+$  in which all clusters have 20 valence electrons. Distorted-wave models are usually reliable at intermediate and high impact energies. In the cluster series studied here, the average velocity of the valence electrons lies in the interval [0.2, 0.4] au. Then, impact energies of the positron greater than 1 eV correspond to the intermediate and high-energy regime. However, we have considered only impact energies lower than the threshold energy for positronium formation from the outer shell ( $2p^6$ ) of the cluster ionic cores ( $\text{Na}^+$ ) which is 42 eV [19]. Thus only the capture of the valence electron has been considered in this work.

For the sake of clarity and for the discussion of the results, the ionization potentials calculated in the approximation LDA-SIC for the occupied orbitals 1s, 1p, 1d and 2s of the three clusters are given in table 1.

In figure 2 we compare the orbital-dependent short-range potentials of the positron–residual target interaction of  $\text{Na}_{20}$  with the average potential defined by equation (15). As can be clearly seen from the figure, over the entire range of distances, the potentials are very

**Table 1.** Ionization potential (in eV) of the occupied single-particle states of  $\text{Na}_{19}^-$ ,  $\text{Na}_{20}$  and  $\text{Na}_{21}^+$  calculated in the approximation LDA-SIC.

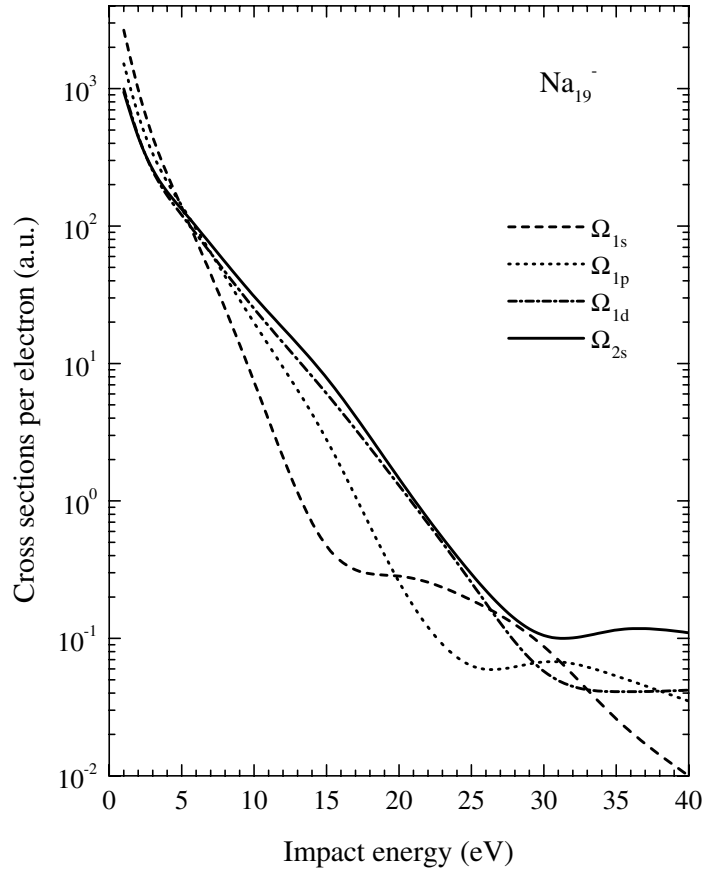
Orbital	$\text{Na}_{19}^-$	$\text{Na}_{20}$	$\text{Na}_{21}^+$
1s	4.120	6.336	8.605
1p	3.166	5.408	7.693
1d	2.073	4.322	6.612
2s	1.615	3.853	6.132

**Figure 2.** Short-range part of the positron–residual target interaction. Comparison between the  $V_P^j$  potentials (see equation (14)) (1s, broken curve; 1p, dotted curve; 1d, chain curve; 2s, full curve) and the average potential  $V_P$  (see equation (15)) (open circles) of  $\text{Na}_{20}$ . The arrow at the bottom of the figure indicates the position of the cluster radius.

close together. Thus, it is completely justified the use a common average potential for all of the active electrons. We have checked that the same conclusions hold for  $\text{Na}_{19}^-$  and  $\text{Na}_{21}^+$ .

Before discussing our results, some comments and explanations have to be made concerning collective effects and fragmentation which are not included in our present model.

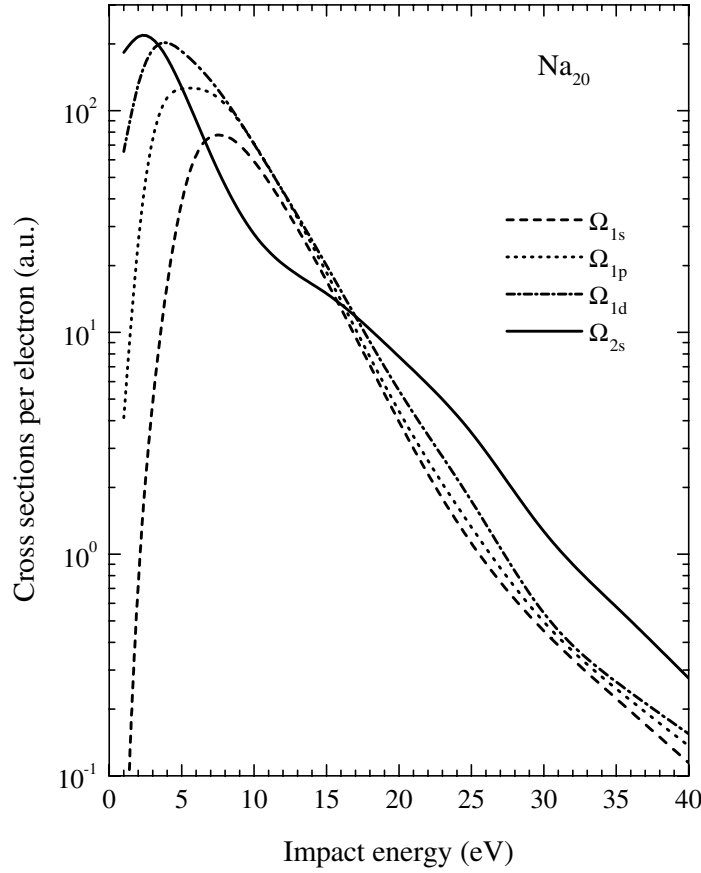
Firstly, concerning a possible surface plasmon excitation during the collision process, the relevant quantity to be discussed is the energy difference between the electronic bound and continuum states, namely  $\Delta E = -\varepsilon_i + T^{\text{Ps}}/2$ , where  $T^{\text{Ps}}$  is the kinetic energy of the positronium atom in the final channel. From the energy conservation law we have  $\Delta E = \frac{1}{2}(E - \varepsilon_i - \varepsilon_{1s}^{\text{Ps}})$  where  $\varepsilon_i$  ( $< 0$ ) is the bound-state energy of the orbital  $i$ ,  $E$  and  $\varepsilon_{1s}^{\text{Ps}} \approx -6.8$  eV are the positronium impact energy and the binding energy of the positronium atom in its ground state, respectively. With the values of  $(-\varepsilon_i)$  given in table 1 and for the lowest impact energy considered in this work,  $E = 1$  eV,  $\Delta E$  ranges from 4.7 eV (2s state of  $\text{Na}_{19}^-$ ) and 8.2 eV (1s state of  $\text{Na}_{21}^+$ ). For sodium clusters having 20 electrons, the surface plasmon is a resonant process and has an energy around 2.8 eV. Since  $\Delta E$  is always greater than 2.8 eV, even in the most unfavourable energetic case, we do not expect any collective excitation during the collision.



**Figure 3.** Total cross sections per electron (see text) for  $e^+Na_{19}^-$  collisions. CDW-FS results:  $\Omega_{1s}$ , broken curve;  $\Omega_{1p}$ , dotted curve;  $\Omega_{1d}$ , chain curve;  $\Omega_{2s}$ , full curve.

Concerning fragmentation, it is only an indirect process by energy deposition. Indeed, the ratio of the masses of the particles involved in the collision is very small  $m_{e^+}/m_{Na} \approx 1/(23 \times 1840)$  and therefore it would be legitimate to think that frontal collisions between the incoming positron and one of the cluster atoms cannot directly induce fragmentation. After the capture of an electron from a cluster orbital, the remaining cluster (i.e.  $Na_{19}$ ,  $Na_{20}^+$  and  $Na_{21}^{++}$ ) is electronically excited. One may estimate the higher bound to the excitation energy as being the difference between the electronic ground-state energies of the cluster before and after the collision. In the LDA-SIC approximation this quantity is roughly given by  $1/R_C \approx 2.5$  eV (in a macroscopic picture, this is the electrostatic energy required to remove a charge unity from the surface of a metallic sphere of radius  $R_C$  to infinity). As this latter quantity is larger than the lowest dissociation energy, which is around 1 eV for sodium clusters of these sizes [15], fragmentation may occur (mainly sequential evaporation of a monomer or a dimer). The fragmentation process is not sudden (at least for  $Na_{19}^-$  and  $Na_{20}$ ) and the system takes some time to dissociate. Depending on the impact energy, if the Ps atom (formed during the collision process) is far enough from the collision region when the evaporation occurs one may neglect fragmentation in the description of the collision process. Let us suppose that the incoming cluster is initially produced at  $T = 0$  K then the evaporation time is estimated using

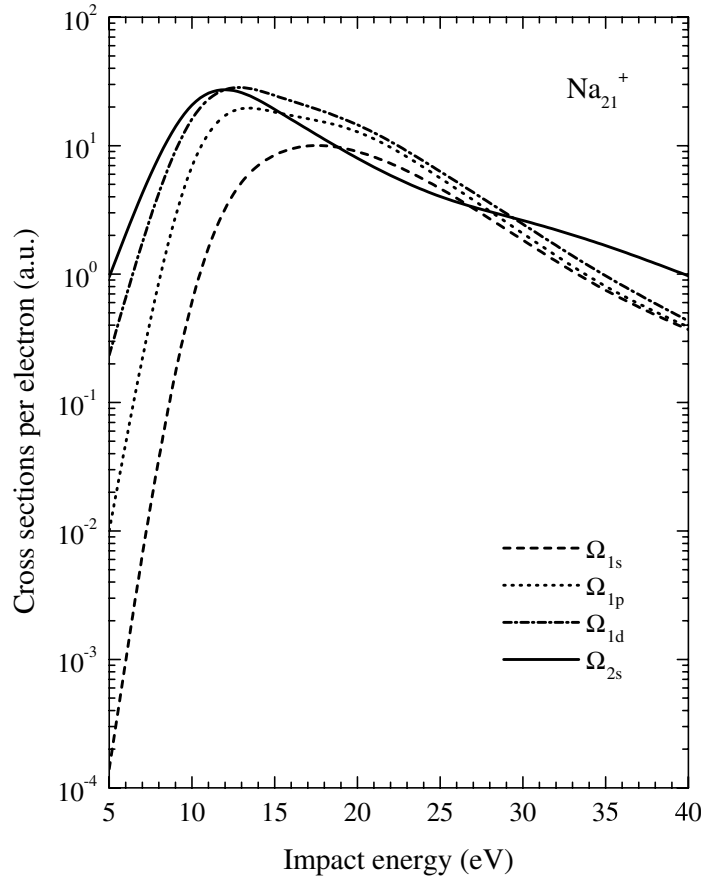




**Figure 4.** Same as in figure 2 but for  $\text{Na}_{20}$ .

a statistical model of Weisskopf [20] is  $\tau_{\text{ev}} \approx 2.5 \times 10^{-2}$  s. The velocity of the Ps atom is given by  $v^{\text{Ps}} = \sqrt{T^{\text{Ps}}} = \sqrt{E + \varepsilon_i - \varepsilon_{1s}^{\text{Ps}}}$ . For an impact energy  $E = 5$  eV,  $v^{\text{Ps}}$  ranges from 0.34 au (1s state of  $\text{Na}_{21}^+$ ) to 0.61 au (2s state of  $\text{Na}_{19}^-$ ). At  $t = \tau_{\text{ev}}$  the corresponding distance of the Ps atom from the cluster centre of mass,  $R^{\text{Ps}}$ , ranges from  $1.86 \times 10^4$  m to  $3.34 \times 10^4$  m. For  $T = 400$  K one finds  $\tau_{\text{ev}} \approx 1.5 \times 10^{-7}$  s and  $1.1 \times 10^{-1} < R^{\text{Ps}} < 2 \times 10^{-1}$  m. Thus, even at low impact energy and for not too high temperature it is not necessary to take into account the fragmentation in the calculation of the positronium formation cross section. Finally, beside all of the above arguments, one may expect a weak influence of the fragmentation on the positronium formation since this atomic system is neutral.

We show in figures 3–5 the CDW-FS total cross sections per electron (i.e.  $\Omega_{n,l_i} \equiv \sigma_{n,l_i}/2\hat{l}_t$ ) as a function of the positron impact energy for the three systems under study. It can be seen from the figures that as the energy increases, capture from the 2s orbital becomes dominant. This is in contrast to the behaviour of the TCS observed in collisions of positrons with atomic targets [6]. This difference may be attributed to the potential that bounds the electrons in atoms and clusters. In the latter, electrons are in the field of an effective potential (see figure 1 of [11]) that has small variations over a great distance ( $\sim R_C \cong 10$  with  $V_{\text{KS}} \cong -0.25 - Z/(R_C + \delta)$  and  $\delta \cong 1$  is the spill-out of the electronic distribution [12]) from the centre of mass of the



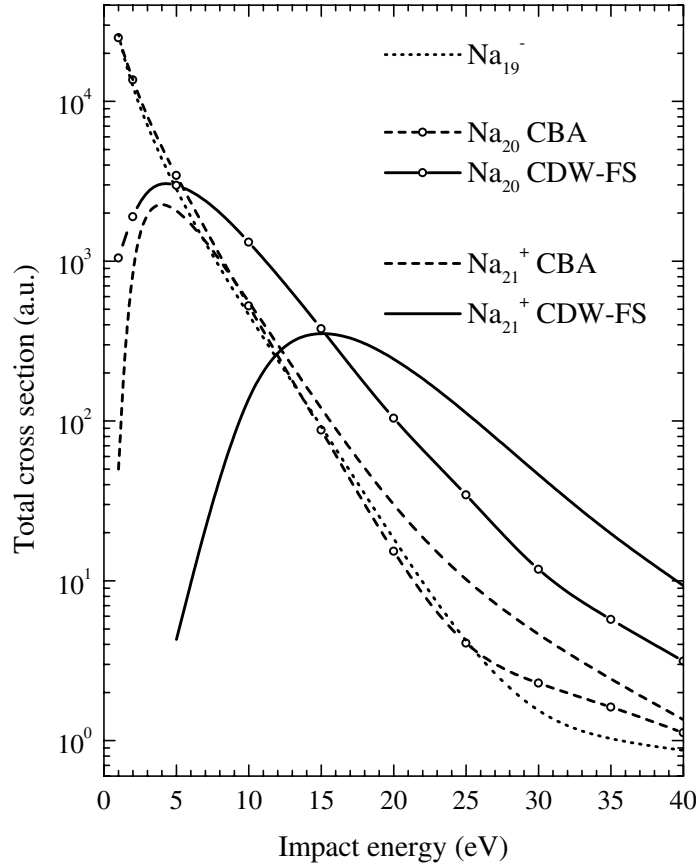
**Figure 5.** Same as in figure 2 but for  $\text{Na}_{21}^+$ .

cluster<sup>3</sup>. Moreover, the spatial extension of the wavefunction of each orbital is of the same order of magnitude. Consequently, electrons in the 2s state have an average velocity greater than those in the other states studied. Then, the fact that capture from 2s preferential as the incident energy increases may be explained from simple matching-velocity arguments.

In figure 6, CDW-FS and CBA TCS given by equation (30) are shown for the three studied clusters. For  $\text{Na}_{19}^-$ , the CDW-FS and CBA approximations give the same results as in this case the charge of the target is  $Z = -1$  and then  $\beta_+ = \beta_- = 0$  (see equation (9)). For all clusters, the capture from 1d orbitals (each of them containing 10 electrons) contributes at about 50% to the TCS.

Finally, we present in figure 7 the CDW-FS DCS as a function of the scattering angle for  $\text{Na}_{20}$ . The DCS summed over all the initial states is also shown (see equation (29)). As already mentioned, it can be seen that the contribution to the TCS of the 1d orbitals is always dominant.

<sup>3</sup> Table 1 indicates that the difference in energy between the ionization potential of the occupied states (calculated in the approximation LDA-SIC) of  $\text{Na}_{19}^-$  and  $\text{Na}_{20}$  or  $\text{Na}_{21}^+$  and  $\text{Na}_{20}$  is constant and is approximately equal to  $Z/(R_C + \delta)$ . Thus, since  $V_{\text{KS}} \approx -0.25 - Z/(R_C + \delta)$  it turns out that for a given orbital, the average velocity of the electrons are practically the same along an isoelectronic sequence.



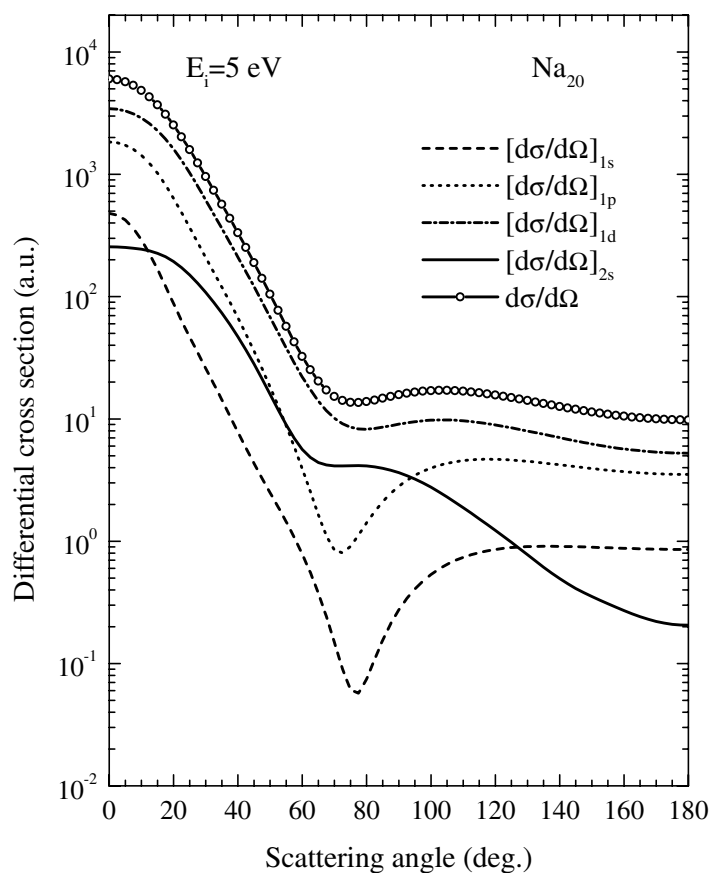
**Figure 6.** CDW-FS and CBA total cross section for positronium formation as a function of the positron energy.

#### 4. Conclusions and perspectives

In summary, positronium formation by positron impact on simple metal clusters has been studied by using the CDW-FS and CBA approximations within an IEM model. Differential and total cross sections have been computed for the isoelectronic series  $\text{Na}_{19}^-$ ,  $\text{Na}_{20}$  and  $\text{Na}_{21}^+$ .

This work is the beginning of a theoretical study of positron–cluster collisions. In the near future, we plan to study several points of interest in the dynamics of the collision process such as the evolution of the positronium formation cross section as a function of the cluster size. In this work, we have only studied ground-state positronium formation. Capture to the excited states of the positronium atom may play an important role in the charge transfer process studied. In the same way, many-electron correlations not included in our independent electron model are known to be very important in certain collision processes [10, 21]. The last two subjects will be a matter for future work. Finally, we will also evaluate capture from the cluster ionic core electrons at higher impact energies.

Although we have only presented results for a sodium cluster, other alkali clusters such as lithium or potassium clusters will also be of interest. The charge transfer process with positronium formation may become a useful tool to provide a considerable number of neutral clusters (for instance,  $\text{Na}_{19}$  from  $\text{Na}_{19}^-$ ) not easily obtained by other means [22].



**Figure 7.** Differential cross section as a function of the scattering angle for  $e^+ + \text{Na}_{20}$  at an impact energy of  $E_i = 5$  eV. CDW-FS results: 1s, broken curve; 1p, dotted curve; 1d, chain curve; 2s, full curve; summed over all orbitals, open circles.

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