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2003 J. Phys. B: At. Mol. Opt. Phys. 36 L211

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LETTER TO THE EDITOR

Double shake-off model for the triple photoionization of beryllium

A S Kheifets¹ and Igor Bray²

¹ Research School of Physical Sciences and Engineering, The Australian National University, Canberra ACT 0200, Australia

² Centre for Atomic, Molecular, and Surface Physics, School of Mathematical and Physical Sciences, Murdoch University, Perth, 6150, Australia

E-mail: A.Kheifets@anu.edu.au and I.Bray@murdoch.edu.au

Received 2 April 2003

Published 16 June 2003

Online at stacks.iop.org/JPhysB/36/L211

Abstract

We propose a model for the triple photoionization of Be in which a core $1s$ electron absorbs the photon $\gamma + 1s^2 2s^2 \text{Be} \rightarrow \epsilon_1 p + 1s 2s^2 \text{Be}^+$ and the valence $2s^2$ electrons are shaken off into continuum due to the sudden change of the core potential. We decompose the double shake-off amplitude into a single shake-off $2s^2 \rightarrow ns\epsilon s$ and a subsequent electron impact ionization of the doubly charged Be^{2+} ion $\epsilon s + 1sns \text{Be}^{2+} \rightarrow \epsilon_2 l + \epsilon_3 l + 1s \text{Be}^{3+}$. The latter process is described by the T -matrix of inelastic electron scattering on the ‘semi-hollow’ $1sns \text{Be}^{2+}$ ion in the monopole singlet channel. The convergent close-coupling method is used to evaluate the T -matrix.

Over recent years, multiple photoionization processes have been under intensive scrutiny. The interest in these processes is motivated by the fact that a photon, outside the domain of superstrong laser fields, can only couple to a single atomic electron. The simultaneous ejection of two or more atomic electrons after the absorption of a single photon is driven entirely by many-electron correlations. The many-body problem, especially with several electrons in the continuum, can only be solved in various approximations and extensive experimental studies are needed to find the limitations of these approximate solutions.

The simplest direct multiple photoionization process is the double photoionization (DPI) of He. As a result of massive theoretical and experimental efforts over the past ten years, a good understanding of this fundamental process now emerges. The mechanisms of He DPI are well understood. Cross sections are established reliably in a wide photon energy range with good agreement between theory and experimentation. Some important aspects of He DPI have been reviewed recently by Briggs and Schmidt (2000).

As the He DPI problem is now close to being solved, researchers are diverting their attention to other atomic targets. The Be atom is a very attractive choice. The core $1s^2$ and

valence $2s^2$ shells of Be are well separated, both in energy and coordinate space. From the double ionization threshold at 27.5 eV up to the first autoionization resonance at about 115 eV, the core electrons remain spectators and take no direct part in photoionization. This allows the application of various frozen-core models and the treatment of Be DPI in a similar way to that of He (Kheifets and Bray 2002, Colgan and Pindzola 2002, Citrini *et al* 2003). Experimental data on direct Be DPI recently became available (Wehlitz and Whitfield 2001). The total DPI cross section in the range of photon energies between 32 and 80 eV was found to be in fair agreement with theoretical predictions (Wehlitz and Whitfield 2002). Hasegawa *et al* (2002) reported the total DPI cross section of Be in the photon energy range above 115 eV. This measurement fell into the region of autoionizing resonances and could not be compared with the theories of direct DPI. In general, for atoms with more than one electron shell, multiple photoionization can proceed sequentially via Auger transitions or the excitation of autoionizing resonances. These processes, interesting in themselves, are not strong markers of electron correlations and will be excluded from our further consideration.

The cross section of direct Be DPI was found to be smaller than that of He. This was interpreted by Wehlitz and Whitfield (2001) to be the result of a larger separation and weaker correlation of the two valence $2s$ electrons. However, Kheifets and Bray (2002) observed a much stronger angular correlation between the photoelectrons in Be than that in He. The Gaussian width parameter, which governs the angular correlation at equal energy sharing between the photoelectrons, was found to be 68° and 90° for Be and He, respectively, at the same excess energy of 20 eV above the threshold. We remind the reader that the Gaussian width parameter converges to zero at the threshold which corresponds to the rigidly correlated back-to-back Wannier escape.

Another level of complexity is presented by triple photoionization (TPI) in which the absorption of a single photon results in the simultaneous ejection of three atomic electrons. The first TPI measurement was performed on Li by Wehlitz *et al* (1998). These authors suggested that for sufficient excess energy, Li TPI is reasonably well described by the double ionization of the inner electrons followed by a shake-off of the outer electron. This decomposition of the three-electron break-up process into a two-electron emission plus shake-off requires sufficiently large excess energy above the triple ionization threshold which should exceed the binding energy of the outer $2s$ electron: $\Delta E = \omega - IP^{3+} > |\epsilon_{2s}|$. The DPI of the $1s^2$ core of the Li^+ ion can be described accurately by non-perturbative models (Kornberg and Miraglia 1993, Kheifets and Bray 1998). As to the shake-off probability of the outer $2s$ electron, Wehlitz *et al* (1998) assumed that both inner-shell electrons are removed instantaneously which would be the case in the infinite photon energy limit. Various calculations (Wehlitz *et al* 1998, van der Hart and Greene 1998, Cooper 1999, Santos *et al* 2001) predicted different shake-off rates depending on the way they described the Li ground state. The multi-configuration Dirac-Fock calculation of Santos *et al* (2001) seems to be in best agreement with the experiments of Wehlitz *et al* (1998). A more realistic approach to Li TPI was taken by Pattard and Burgdorfer (2001). These authors suggested a so-called half-collision model (HCM) in which the two primary electrons ejected from the $1s^2$ shell collide with the $2s$ electron on their way out of the atom.

Li TPI is an analogue of He DPI as both processes result in the complete fragmentation of the target atom. A similar, but somewhat more complex process is Be TPI. The Be atom has already been earmarked as a good candidate for studying TPI (Wehlitz *et al* 2002). From the triple ionization threshold at 181.4 eV, there is a wide photon energy range of more than 100 eV where the TPI cross section is not affected by sequential or autoionization processes and where straightforward comparison between theoretical predications and experimentation is possible.

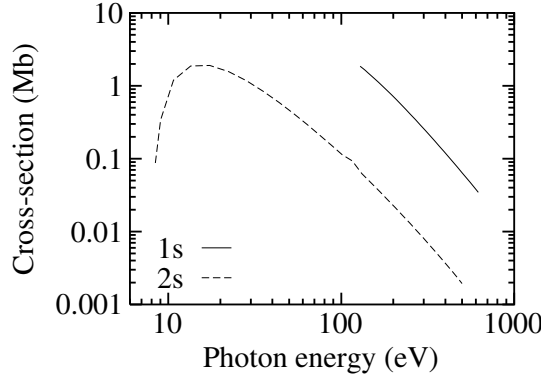


Figure 1. Single photoionization cross sections of the $1s^2$ and $2s^2$ shells of Be.

In anticipation of forthcoming experiments, we propose a theoretical model of Be TPI in which we decompose this process into a single photoionization of the inner $1s^2$ shell $\gamma + 1s^2 2s^2 \text{ Be} \rightarrow \epsilon_1 p + 1s 2s^2 \text{ Be}^+$ and a double shake-off (DSO) of the outer $2s^2$ shell. We do not assume an instantaneous departure of the two $2s^2$ electrons. Rather, we allow them to interact with each other and the nucleus on their way out of the atom. In this sense, our approach is similar to the HCM of Pattard and Burgdorfer (2001). However, instead of the time-dependent perturbation theory in coordinate space employed by these authors, we use the non-perturbative convergent close-coupling (CCC) formalism. In our model, we further decompose the DSO from single shake-off to a virtual intermediate state $2s^2 \rightarrow ns\epsilon s$ followed by electron-impact ionization of the doubly charged Be^{2+} ion leading to the triply ionized final state $\epsilon s + 1sns \text{ Be}^{2+} \rightarrow \epsilon_2 l + \epsilon_3 l + 1s \text{ Be}^{3+}$. The latter process is described by the T -matrix of inelastic electron scattering on the ‘semi-hollow’ $1sns \text{ Be}^{2+}$ ion in the monopole singlet channel.

We write the single photoionization cross section of a closed ns^2 shell in atomic units, as defined by Amusia (1990)

$$\sigma_{ns^2}^+(\omega) = \frac{8\pi^2\omega}{c} \int d^3k |\langle kns|D|ns^2\rangle|^2 \delta(\omega - E + \epsilon_{1s}). \quad (1)$$

This expression corresponds to the momentum space normalization of the continuous wavefunction: $\langle \mathbf{k}|\mathbf{k}'\rangle = \delta(\mathbf{k} - \mathbf{k}')$. The dipole operator D is evaluated in the random phase approximation with exchange (RPAE) using the computer code of Chernysheva *et al* (1974). The photoionization cross sections of the inner $1s^2$ shell as well as the outer $2s^2$ shell are presented in figure 1. We see that at the photon energy range of our interest $\sigma_{1s^2}^+ \gg \sigma_{2s^2}^+$. It is well known that the photoabsorption process takes place preferentially near the nucleus to accommodate the recoil momentum. At the same photon energy the probability of this process is much higher for inner-shell electrons than for outer-shell electrons. It is for this reason that we choose the $1s^2$ single photoionization as the most probable precursor of the TPI.

We write the TPI cross section as

$$\begin{aligned} \sigma^{3+}(\omega) = & \frac{8\pi^2\omega}{c} \sum_{\epsilon_f > 0} \int d^3k_1 d^3k_2 |\langle k_1 1s|D|1s^2\rangle|^2 |\langle k_2 f|\mathcal{O}|2s^2\rangle|^2 \\ & \times \delta(\omega - IP^{3+} - E_1 - E_2 - \epsilon_f). \end{aligned} \quad (2)$$

Here $E_i = k_i^2/2$, $i = 1, 2$. Following the general philosophy of the CCC method, we introduce a complete set of pseudostates $|f\rangle$ diagonalizing the ‘semi-hollow’ Be^{2+} Hamiltonian. Positive

energy pseudostates, $\epsilon_f > 0$, contribute to TPI. The DSO amplitude from the outer $2s^2$ shell is defined in equation (2) as

$$\langle \mathbf{k}_2 f | \mathcal{O} | 2s^2 \rangle = \langle \mathbf{k}_2 f | 2s^2 \rangle + \sum_j \int d^3 \mathbf{k} \frac{\langle \mathbf{k} j | 2s^2 \rangle \langle \mathbf{k}_2 f | T | j \mathbf{k} \rangle}{k_2^2/2 + \epsilon_f - k^2/2 - \epsilon_j + i\delta}. \quad (3)$$

The first term in the right-hand side of equation (3) corresponds to the direct (or ‘bare’) shake-off whereas the second integral term describes the shake-off assisted by the inelastic electron scattering on the ‘semi-hollow’ Be^{2+} ion. This process is evaluated via the half on-shell T -matrix which is found by solving the Lippmann–Schwinger equation in the monopole singlet channel. Equation (3) is analogous to the CCC expression of the amplitude of He DPI given by Kheifets and Bray (1996). However, in the latter case, the T -matrix is evaluated in the dipole, rather than the monopole, singlet channel.

The bare shake-off amplitude $\langle \mathbf{k} j | 2s^2 \rangle$ is calculated as an overlap between the valence $2s^2$ shell of the neutral Be atom and the two-electron state $|\mathbf{k} j\rangle$ which consists of a Coulomb wave and a pseudostate of the semi-hollow Be^{2+} ion. The Coulomb wave $|\mathbf{k}\rangle$ is calculated in the asymptotic charge $Z_{\text{asym}} = 2$ of the nucleus $Z = 4$ screened by the core and valence electrons. This bare shake-off amplitude is sensitive to the ground-state correlation as the valence electrons can be found, with finite probability, in virtual excited states nl^2 , $l > 0$, $n \geq 2$. In the present calculation we account for all virtual excitations with $l \leq 4$ and $n \leq 5$ by constructing the multiconfiguration Hartree–Fock ground state (Dyall *et al* 1989). Convergence with respect to the final state was tested by gradually increasing the number of pseudostates, the maximum angular momentum of the Coulomb waves and the size of the discrete \mathbf{k} -grid representing the integration over intermediate continuum states in the integral term of equation (3). Convergence was achieved in the calculation comprising $20 - l$ pseudostates for $0 \leq l \leq 4$ and 75 \mathbf{k} -grid points.

To simplify the notation in equation (2) we introduce the DSO cross section

$$\sum_{\epsilon_f > 0} \int d\mathbf{k}_2 |\langle \mathbf{k}_2 f | \mathcal{O} | 2s \rangle|^2 \delta(\omega - IP^{3+} - E_1 - E_2 - \epsilon_f) \equiv \sigma_{2s^2}^{2+}(\Delta E - E_1) \quad (4)$$

where $\Delta E = \omega - IP^{3+}$ is the excess energy above the TPI threshold. Using expressions (1) and (4) we rewrite equation (2) as

$$\sigma^{3+}(\omega) = \int_0^{\Delta E} dE_1 \frac{\omega}{\omega'} \sigma_{1s^2}^{+}(\omega') \sigma_{2s^2}^{2+}(\Delta E - E_1). \quad (5)$$

Here $\omega' = E_1 - \epsilon_{1s}$ is an equivalent photon energy corresponding to the energy of the primary photoelectron E_1 .

The DSO cross section (4) as well as the cross sections of the single shake-off processes leaving the Be ion in various doubly charged states ($1snl$, $n = 2, \dots, 6$) are shown in figure 2. In the same figure we also show the cross sections of electron scattering on the ‘semi-hollow’ $\text{Be}^{2+} 1s2s$ ion leading to the same final states. These cross sections are obtained from the monopole T -matrix $\langle \mathbf{k}_2 nl | T | 2s \mathbf{k} \rangle$ taken on the energy shell. We see that at small electron energies the shake-off cross sections to higher excited states $n \geq 4$ and the DSO scale with the same constant to the corresponding electron scattering and electron impact ionization cross sections in the monopole channel. This situation is very similar to photoionization–excitation and DPI processes which scale, near the threshold, to corresponding electron scattering and electron impact ionization cross sections in the dipole channel (Samson 1990, Samson *et al* 1992). This scaling can be interpreted as the dominance of the two-step double ionization, assisted by inelastic electron scattering, over the direct shake-off mechanism (Kheifets 2001).

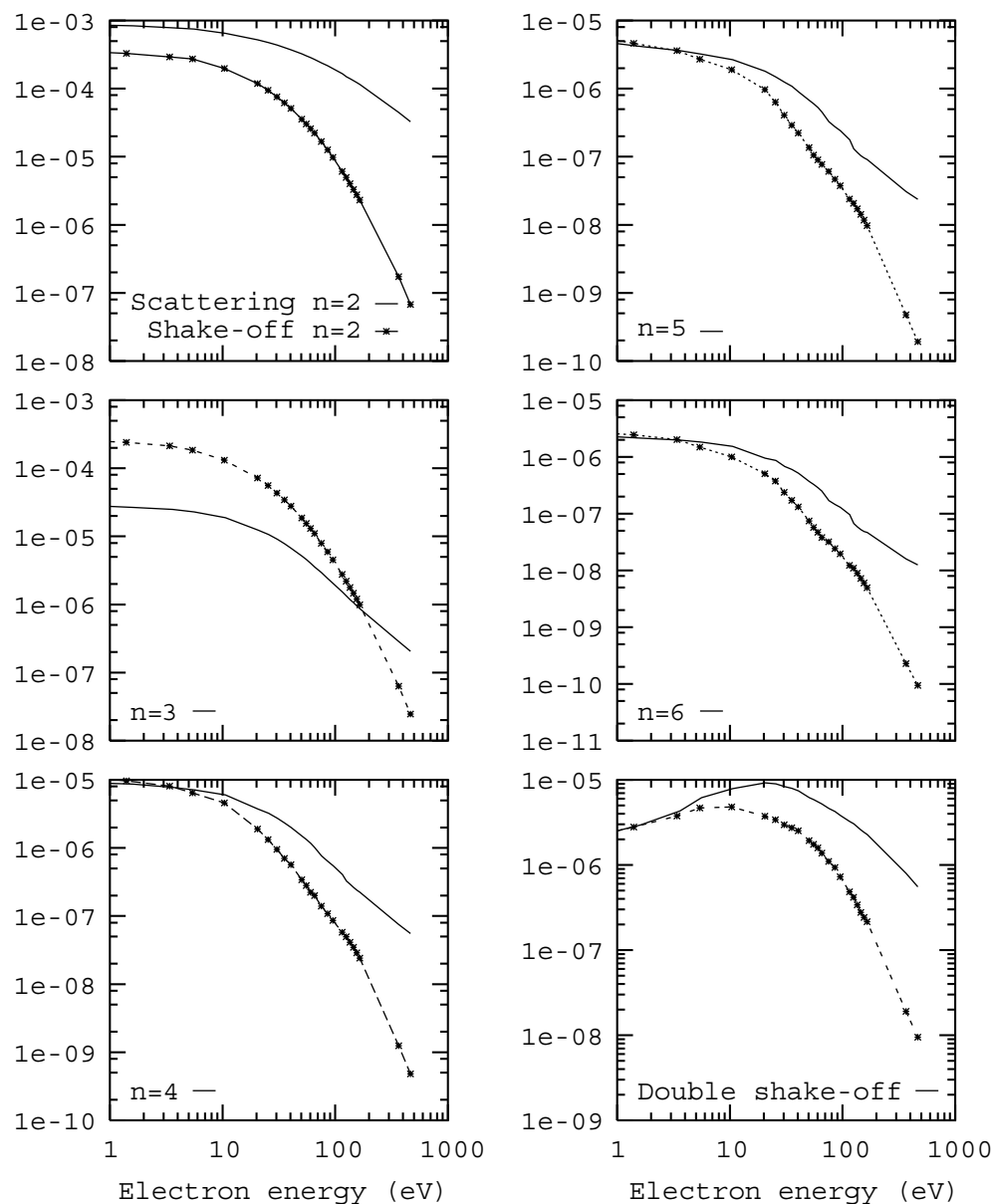


Figure 2. Shake-off cross sections of the Be^{2+} ion leading to various doubly charged states ($1snl$, $n = 2 \dots 6$) and a triply ionized state are shown by the dotted curve. Cross sections of the inelastic electron scattering on the semi-hollow $\text{Be}^{2+} 1s2s$ ion leading to the same final states are shown by the solid curve. The same scaling constant is applied to all the graphs.

The TPI cross section of Be is shown in figure 3. In part (a) we show the ratio of triple-to-single photoionization cross sections in comparison with the experimental (Wehlitz *et al* 1998) and calculated (Kheifets and Bray 1998) ratios for Li. The calculated triple-to-single ratio is fitted to the experiment by multiplying an *ab initio* double-to-single ratio of Li^+ by an empirical shake-off probability of the outer $2s$ electron. This fitted value of 0.0035 is to be compared with the calculated values of 0.001 74 (Wehlitz *et al* 1998), 0.004 65 (Cooper 1999)

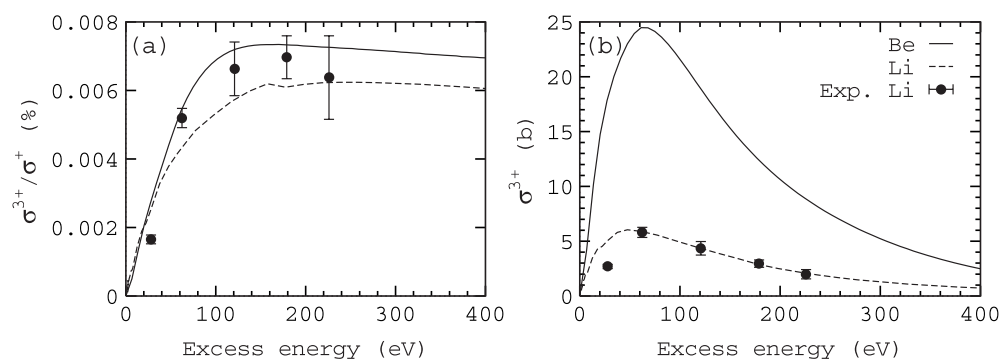


Figure 3. (a) Triple-to-single photoionization cross sections ratio in Be (solid curve) and Li (dashed curve). The experimental ratio for Li is from Wehlitz *et al* (1998). The calculated ratio for Li is from Kheifets and Bray (1998). (b) As in (a) but for the absolute TPI cross section.

and 0.007 03 (Santos *et al* 2001). The triple-to-single ratio in Be, shown in figure 3(b), is about the same as in Li. However, the absolute TPI cross section is bigger by about a factor of 4 due to a large single photoionization cross section. The triple-to-single ratio in Be reaches its peak value at about 100 eV above the triple ionization threshold as compared to 200 eV in Li. This faster onset of TPI in Be is explained by a smaller energy scale of the DSO from the outer $2s^2$ shell, whereas in Li most of the energy dependence of TPI comes from the DPI of the inner $1s^2$ shell.

In conclusion, we proposed a model of Be TPI in which this process is decomposed into the single photoionization of the inner $1s^2$ shell followed by the DSO of the outer $2s^2$ shell. The latter is calculated using the CCC model of electron impact ionization of the ‘semi-hollow’ Be^{2+} ion. We predict a TPI cross section of the peak value above 20 barns, considerably bigger than that of Li. This significant cross section should render possible the experimental observation of TPI on Be. With respect to other atomic targets, Suzuki *et al* (2002) reported the first measurements of Mg TPI. The present theory can be easily modified to account for the DSO from the outer $3s^2$ shell of Mg. However, due to the proximity of the subvalent $2p^6$ shell, Mg TPI will be strongly affected by indirect processes not accounted for in our model.

One of the authors (ASK) expresses his gratitude to the Japanese Society for the Promotion of Science for supporting his visit to the Photon Factory (PF-KEK). ASK also wishes to thank Professor Y Azuma and members of his group at PF-KEK for many stimulating discussions. The authors thank Dr R Wehlitz for critical reading of the manuscript. The Australian Partnership for Advanced Computing is acknowledged for providing access to the Compaq AlphaServer SC National Facility.

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