

LETTER TO THE EDITOR

Fully differential cross sections for transfer ionization—a sensitive probe of high level correlation effects in atoms

A L Godunov¹, Colm T Whelan¹ and H R J Walters²

¹ Department of Physics, Old Dominion University, Norfolk, VA 23529-0116, USA

² Department of Applied Mathematics and Theoretical Physics, The Queen's University of Belfast, Belfast BT7 1NN, UK

E-mail: godunov@physics.odu.edu

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Abstract

The transfer ionization process offers a unique opportunity to study radial and angular electron correlations in the helium atom. We report calculations for the multiple differential cross sections of the transfer ionization process $p + \text{He} \rightarrow \text{H} + \text{He}^{++} + e^-$. The results of these calculations demonstrate the strong sensitivity of the fully differential cross sections to fine details of electron correlation in the target atom. Specifically, angular electron correlation in the ground state of helium results in a broad peak in the electron emission spectra in the backward direction, relative to the incoming beam. Our model explains some of the key effects observed in measurements of multiple differential cross sections using the COLTRIMS technique.

In this letter, the fine details of atomic electron correlation are investigated through transfer-ionization collisions. Information on correlation of target electrons from differential cross sections of single ionization, such as photo-ionization, Compton scattering or $(e, 2e)$ (Åberg 1976, Bergstrom *et al* 1993, Whelan 1999, Stefani 1999) is readily available. In recent times it has become clear that much detailed information on correlated target states can be found by studying double electron transitions. Examples of such processes are $(\gamma, 2e)$, $(e, 3e)$ and excitation-ionization (Huetz *et al* 1991, Lahmam-Bennani 1991, Marchalant *et al* 1998, Rash *et al* 2002). Unfortunately, such processes concerning charged projectiles, although sensitive to target effects, are largely dependent on post collision interaction between ejected electrons and the projectile. Transfer ionization is another example of ionization in double electron transitions. For the proton impact, the projectile in the final state is neutral (H^0) and therefore the post-collision Coulomb interaction has a small effect on differential cross sections of ionization (Barrachina 1990).

Total cross sections for transfer ionization have been studied both experimentally and theoretically (Shah and Gilbody 1985, Dunseath and Crothers 1991, Belkic *et al* 1997, Cohen

1996). However, few measurements have been reported for single and double differential cross sections (Palinkas *et al* 1989, Bross *et al* 1994). The fully differential cross section from the transfer ionization process $p + \text{He} \rightarrow \text{H} + \text{He}^{++} + e^-$ has recently been measured using the COLTRIMS technique (Mergel *et al* 2001, Schmidt-Böcking *et al* 2001a, 2003a, 2003b). These experiments reveal that (a) the ejected electron is predominantly emitted into the backward direction, (b) the direction of maximum ejection is insensitive to the impact energy but shows some dependence on the momentum transfer and (c) the captured electron, recoil He^{2+} ion and the ejected electron always share comparable momenta. Schmidt-Böcking suggested that a contribution from non- s^2 terms to the initial state wavefunction of helium might explain these effects (Schmidt-Böcking *et al* 2001b, 2003a). Kheifets (see Schmidt-Böcking *et al* 2003a) evaluated a double overlap integral between the correlated helium ground state and two electron continua. However, no data for multiply differential cross sections were presented.

Transfer ionization at intermediate collision velocities may proceed via few channels (McGuire 1997). Most of the mechanisms are sensitive to collision energy. Since the experimentally observed features lacked sensitivity to collision energy, one may assume that the target correlation plays the leading role. If this is the case, then one may use fully differential cross sections of transfer ionization as a sensitive probe of target correlation because such a process is free from the effect of final state Coulomb interaction between the projectile and the ejected electron. In this letter we adopt the following picture; transfer ionization can be viewed as a double ionization process where one electron travels in the direction of the scatter projectile, and there is no post collision interaction between the captured and ejected electrons. We employ a first-order model where the transfer ionization is the result of single interaction between the projectile and target. Therefore, the only mechanism for double electron transition in our model is target correlation. Further, when we draw a parallel between transfer ionization and double ionization, the design of the transfer ionization experiment corresponds to a very particular double ionization kinematics, i.e. one electron of the target being ionized and the second ejected due to correlation. Thus, there exist two possibilities: either the electron which is finally captured is the one first ionized and the ejected one comes out because of target correlation, or vice versa. In this letter our goal is not studying collision mechanisms for transfer ionization but the understanding of experimentally observed effects in fully differential cross sections, and how much can be explained by electron correlation through a simple collision model.

Atomic units are used throughout the letter unless otherwise stated.

The triple differential cross section for transfer ionization as a function of the scattered angle Ω_f , the energy E_e and the angle Ω_e of the ionized electron is defined as

$$\frac{d^3\sigma}{dE_e d\Omega_e d\Omega_f} = \frac{1}{(2\pi)^3} \frac{K_f k_e}{K_i} |f|^2, \quad (1)$$

where K_i and K_f are the momenta of the incoming projectile and the scattered particle, k_e is the momentum of the ejected electron, Ω_f and Ω_e are the solid angle elements about the direction of the scattered particle and the ejected electron. The transition amplitude f is given by

$$f = -\frac{\mu}{2\pi} \langle \Psi_f^{(-)}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{R}) | V_i | \Psi_i(\mathbf{r}_1, \mathbf{r}_2, \mathbf{R}) \rangle, \quad (2)$$

with

$$\Psi_i(\mathbf{r}_1, \mathbf{r}_2, \mathbf{R}) = \frac{1}{(2\pi)^{3/2}} \exp(i\mathbf{K}_i \mathbf{R}_i) \Phi_i(\mathbf{r}_1, \mathbf{r}_2) \quad (3)$$

the asymptotic wavefunction for the unperturbed initial state and V_i is the interaction potential between the projectile of charge Z_p and the two-electron atom with nuclear charge Z_t , namely

$$V_i = -\frac{Z_p}{|\mathbf{r}_1 - \mathbf{R}|} - \frac{Z_p}{|\mathbf{r}_2 - \mathbf{R}|} + \frac{Z_p Z_t}{R}. \quad (4)$$

Here, \mathbf{r}_1 , \mathbf{r}_2 and \mathbf{R} are the position vectors for the two helium electron and the projectile, $\Phi_i(\mathbf{r}_1, \mathbf{r}_2)$ is the wavefunction for the ground state of the helium atom, μ is the reduced mass of the projectile and the target, and $\mathbf{R}_i = \mathbf{R} - (\mathbf{r}_1 + \mathbf{r}_2)/(M_t + 2)$ is the relative coordinate of the projectile and the target in the incoming channel, where M_t is the He^{2+} mass. $\Psi_f^{(-)}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{R})$ is the exact wavefunction for the three-particle system $\text{H}^0 + \text{He}^{2+} + e$.

In our model, the wavefunction for the final state is

$$\Psi_f^{(-)} \approx \frac{1}{(2\pi)^{3/2}} \exp(i\mathbf{K}_f \mathbf{R}_f) \varphi_{nl}(\mathbf{r}_1 - \mathbf{R}) \psi_{\tilde{k}_2}(\mathbf{r}_2), \quad (5)$$

where $\varphi_{nl}(\mathbf{r}_1 - \mathbf{R})$ is the hydrogenic wavefunction for the captured electron, $\psi_{\tilde{k}_2}(\mathbf{r}_2)$ is the Coulomb wavefunction for the ionized electron in the field of the He^{2+} recoil ion and $\mathbf{R}_f = (M_p \mathbf{R} + \mathbf{r}_1)/(M_p + 1) - \mathbf{r}_2/(M_t + 1)$, where M_p is the projectile mass. The wavefunction (5) corresponds to a first-order model. Within this model we study the sensitivity of the fully differential cross section for transfer ionization to angular and radial correlations in the target wavefunction. $\Phi_i(\mathbf{r}_1, \mathbf{r}_2)$. We remark that in our actual calculations we properly include symmetrization over space coordinates; however, it is conceptually useful to present the theory as if the particles were distinguishable.

It is instructive to look at the transition amplitude in more detail. This amplitude contains three terms arising from the potential V_i . The first terms may be considered as one electron of the target being captured by interaction with the projectile and the second is ejected because of correlation. The second term describes the picture when the one electron is initially ionized and then the second is captured due to correlation. The last term takes into account the heavy particle interaction in ion-atom collision.

In order to evaluate the scattering amplitude (2) it is useful to introduce the Fourier transform of the hydrogen wavefunction $\varphi_{nl}(\mathbf{r})$,

$$\varphi_{nl}^F(\mathbf{s}) = \int \exp(+i\mathbf{s}(\mathbf{r}_1 - \mathbf{R})) \varphi_{nl}(\mathbf{r}_1 - \mathbf{R}) d(\mathbf{r}_1 - \mathbf{R}) \quad (6)$$

$$\varphi_{nl}(\mathbf{r}_1 - \mathbf{R}) = \frac{1}{(2\pi)^3} \int \exp(-i\mathbf{s}(\mathbf{r}_1 - \mathbf{R})) \varphi_{nl}^F(\mathbf{s}) d\mathbf{s}. \quad (7)$$

Then, the first term for the transition amplitude that corresponds to ‘transfer first’ transition in our model, after using the Bethe integral, takes the form

$$t_{\text{tr}} = -\frac{\mu}{2\pi} \frac{1}{(2\pi)^6} \int \frac{(-4\pi Z_p)}{|\mathbf{s}_0 - \mathbf{s}|^2} \varphi_{1s}^F(\mathbf{s}) d\mathbf{s} \\ \times \int \psi_{\tilde{k}_2}^*(\mathbf{r}_2) \exp[i\mathbf{r}_1 \mathbf{Q} - i\mathbf{r}_2 \mathbf{Q}/(M_t + 1)] \Phi_i(\mathbf{r}_1, \mathbf{r}_2) d\mathbf{r}_1 d\mathbf{r}_2, \quad (8)$$

where the momentum transferred is $\mathbf{Q} = \mathbf{K}_i(M_t + 1)/(M_t + 2) - \mathbf{K}_f$ and $\mathbf{s}_0 = \mathbf{K}_i - \mathbf{K}_f(M_p)/(M_p + 1)$. One may see from equation (8) that transfer and ionization processes are separable. The angular dependence for the ejected electrons is determined by the second integral describing ionization and does not depend on states for the captured electron. We will show later that in this model the peak around $140^\circ \pm 15^\circ$ to the initial-state momentum vector of the captured electron comes from terms beyond the $(ns)^2$ terms in a multi-configuration Hartree-Fock description of the target. The contribution from charge transfer may be viewed

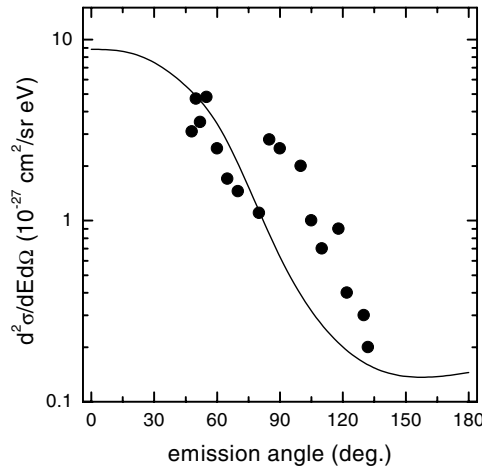


Figure 1. Double differential cross section of electron emission at 600 eV following transfer ionization of He by 1 MeV proton impact. Theory: solid line. Experimental data are from Pálincás *et al* (1989).

as a background for the peak in differential cross sections. It is clear that improving the description for the charge transfer transition may change the absolute value for the cross section but would not affect the peak.

The amplitude that describes the other mechanism, ‘ionization first’ transition in our model, may be written as

$$t_{\text{ion}} = -\frac{\mu}{2\pi} \frac{1}{(2\pi)^6} \int \frac{(-4\pi Z_p)}{|s_0 - s|^2} \varphi_{1s}^F(s) ds \int \psi_{k_2}^*(\mathbf{r}_2) \times \exp[i\mathbf{r}_1(\mathbf{s} - \mathbf{s}_0 + \mathbf{Q}) - i\mathbf{r}_2(\mathbf{s}_0 - \mathbf{s} - \mathbf{Q}/(M_t + 1))] \Phi_i(\mathbf{r}_1, \mathbf{r}_2) d\mathbf{r}_1 d\mathbf{r}_2. \quad (9)$$

In the amplitude (9) the angular distributions for the ejected electrons and transferred electrons are no longer separable. The amplitude corresponding to the last term in the interaction potential (4), i.e. interaction between heavy particles, may be written in a form similar to (9) using the Fourier transform (6) as well.

We evaluated the multiple differential transfer-ionization $p + \text{He} \rightarrow \text{H}^0 + \text{He}^{2+} + e$ cross sections for electrons emitted into the scattering plane in 400 keV–1.2 MeV collisions. Two sets of wavefunctions $\Phi_i(\mathbf{r}_1, \mathbf{r}_2)$ for the ground state of helium were calculated numerically within the multi-configuration Hartree–Fock (MCHF) method (Froese Fisher 1996), with one set allowing for both radial and angular correlations, including $(ns)^2$, $(ps)^2$ and $(nd)^2$ terms with $n \leq 5$, and with the second set including $(ns)^2$ terms only. The triple differential cross sections presented below were calculated by numerical integration of the square of a coherent sum of amplitudes (8) and (9).

Since absolute triple differential transfer-ionization cross sections for $p + \text{He}$ are not available, we tested our model for double differential cross sections compared with the absolute data of Pálincás *et al* (1989) for 1 MeV proton impact (see figure 1). Our calculations largely agreed with the experimental data, however, the peak around 90° was not present. This peak corresponds to the p – e – e scattering (Ishihara and McGuire 1988). However, that is a second-order mechanism and is not included in our first-order model.

In figure 2 we present the triple differential cross section of transfer ionization as a function of the electron emission angle for 1 MeV proton impact and three different scattering angles. The calculations employ a coherent sum of both (8) and (9) amplitudes, and with

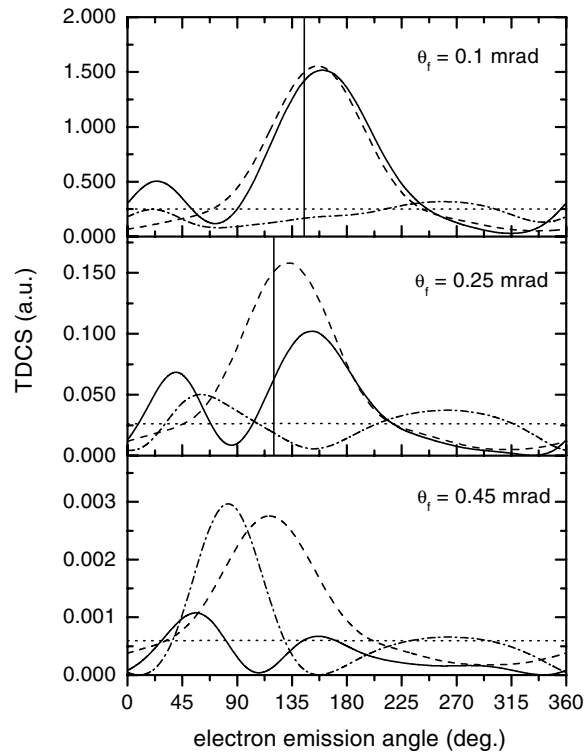


Figure 2. Triple differential cross section for transfer first ionization in a proton–helium collision at $E_i = 1$ MeV, and electron emission energy $E_e = 50$ eV. The scattering angles are 0.1 mrad, 0.25 mrad and 0.45 mrad. Theory: solid line, calculations include both (8) and (9) amplitudes with radial and angular correlations in the initial state wavefunction; chain line, the initial state includes radial correlations only; dashed line, calculations with amplitude (8) only but with radial and angular correlations; dotted line, calculations with amplitude (8) and radial correlation only. Experiment, vertical lines represent positions of the peak in the differential cross sections derived from COLTRIMS measurements (Schmidt-Böcking *et al* 2001a, 2003a).

amplitude (8) alone. It is clearly seen that ‘transfer first’ amplitude with radial correlation only in the wavefunction of the helium initial state produces angle independent electron distribution. Allowing for non- s^2 terms results in a broad peak in the backward direction for the ejected electrons. The peak position corresponds well to the experimentally observed one. However, calculations with both ‘transfer first’ and ‘ionization first’ amplitudes bring new aspects into cross section. For small scattering angles (around 0.1 mrad) amplitude (8) dominates. Inclusion of ‘ionization-first’ amplitude has little effect on the non- s^2 peak in the backward direction, but produces a second small peak at small emission angles. As the scattering angle increases, the contribution from both amplitudes becomes comparable. And finally strong interference between the two amplitudes becomes apparent. The magnitude for the non- s^2 peak in this case is comparable with the magnitude for the peak at small emission angles. For a detailed comparison with experimental data we would need multiple differential cross sections in the form we have given here. However, such data are not yet available. In COLTRIMS experiments (Mergel *et al* 2001, Schmidt-Böcking *et al* 2001a, 2003a, 2003b), five final state momentum components were measured in coincidence, three of the He^{2+} recoil ion and the two transverse momentum components of the H^0 . Then, by applying momentum

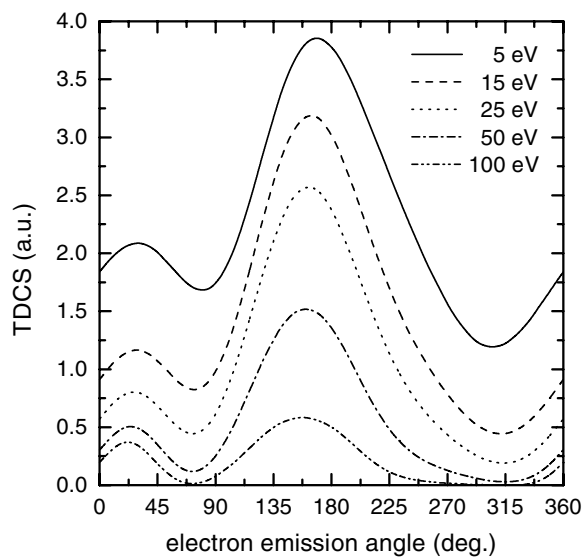


Figure 3. Triple differential cross section for transfer ionization in a proton–helium collision at $E_i = 1000$ keV, and the scattering angle of 0.1 mrad. The ejected electron energies are in the interval of 5 eV–100 eV.

and energy conservation, the positions of the peak in momentum distribution of the ionized electron were derived. However, the experiment does not indicate the strength of the peak relative to the background. The experimental results supporting our calculations and the peak position in the backward direction allow us to reproduce the key aspects reported for the first time. Specifically, the calculations confirm that the only reason for the peak in the backward direction is the effect of angular (non- s^2) correlation in the initial state wavefunction.

A triple differential cross section for different electron emission energies is presented in figure 3. We note that both the shape and position for the non- s^2 maximum in the electron emission spectra lacks sensitivity to the energy of the ejected electron. As the energy of the ejected electron increases the transfer ionization cross section decreases. These results do not agree with experimental observations (Schmidt-Böcking *et al* 2003a, 2003b) where the captured electron, recoil He^{2+} ion and the ejected electron always share comparable momenta.

Calculations for different projectile energies are presented in figure 4, which shows that the position for the maximum is quite insensitive to the collision energy although the magnitude of the cross section decreases quickly with the increasing energy. The same effect was observed in the experiment of Schmidt-Böcking and his collaborators (2003a).

The results of our calculations within the first-order collision model with wavefunctions allowing for angular electron correlation for the initial state reproduce the effects observed experimentally in multiply differential cross sections. Particularly, (a) there is a propensity for the ejected electron to be detected in the backward direction to the incident protons and (b) the direction of maximum ejection is insensitive to the impact energy but shows dependence on the momentum transfer.

While our simple model explains qualitatively some of the observed effects, a more sophisticated treatment of the problem is necessary to resolve the question about comparable momenta for the captured electron, recoil He^{2+} ion and the ejected electron in the final state. It also appears that additional experimental data from COLTRIMS measurements in a form

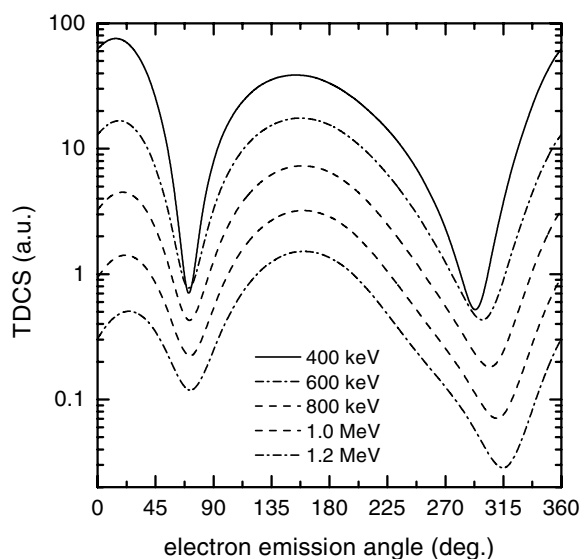


Figure 4. Triple differential cross section for transfer ionization for projectile energies 400 keV–1.2 MeV. The scattering angle is 0.1 mrad; electron emission energy is 50 eV.

allowing direct comparison with theoretical calculations would help to better understand the correlation effects in differential cross sections of transfer ionization.

References

- Åberg T 1976 *Photoionization and Other Probes of Many Electron Interactions* ed F Wuilleumier (New York: Plenum)
- Barrachina R O 1990 *J. Phys. B: At. Mol. Opt. Phys.* **23** 2321–32
- Belkic D, Mancev I and Mergel V 1997 *Phys. Rev. A* **55** 378–95
- Bergstrom P M, Suric T, Pisk K and Pratt R H 1993 *Phys. Rev. A* **48** 1134–62
- Bross S W *et al* 1994 *Phys. Rev. A* **50** 337–42
- Cohen J S 1996 *Phys. Rev. A* **54** 573–86
- Dunseath K M and Crothers D S F 1991 *J. Phys. B: At. Mol. Opt. Phys.* **24** 5003–22
- Froese Fisher C 1996 *Atomic, Molecular and Optical Physics Reference Book* ed G W F Drake (New York: AIP) chapter 21
- Huetz A, Selles P, Waymel D and Mazeau J 1991 *J. Phys. B: At. Mol. Opt. Phys.* **24** 1917–33
- Ishihara T and McGuire J H 1988 *Phys. Rev. A* **38** 3310–18
- Lahmam-Bennani A 1991 *J. Phys. B: At. Mol. Opt. Phys.* **24** 2401–42
- Marchalant P J, Whelan C T and Walters H R J 1998 *J. Phys. B: At. Mol. Opt. Phys.* **31** 1141–78
- McGuire J H 1997 *Electron Correlation Dynamics in Atomic Collisions* (Cambridge: Cambridge University Press)
- Mergel V, Dörner R, Khayyat Kh, Achler M, Weber T, Jagutzki O, Lüdde H J, Cocke C L and Schmidt-Böcking H 2001 *Phys. Rev. Lett.* **86** 2257–60
- Pálincás J, Schuch R, Cederquist H and Gustafsson O 1989 *Phys. Rev. Lett.* **63** 2464–7
- Rash J, Walters H R J, Marchalant P J, Whelan C T and Madison D H 2002 *Photonic, Electronic and Atomic Collisions* ed J Burgdörfer *et al* (Princeton NJ: Rinton) pp 448–59
- Shah M B and Gilbody H B 1985 *J. Phys. B: At. Mol. Phys.* **18** 899–913
- Schmidt-Böcking H, Mergel V, Dörner R, Jagutzki O, Schmidt L, Weber T, Cocke C L, Lüdde H J, Weigold E, Popov Yu V, Cederquist H, Schmidt H T, Schuch R and Berakdar J 2001a *Correlations, Polarization, and Ionization in Atomic Systems (AIP Conf. Proc. vol 604)* pp 120–6
- Schmidt-Böcking H, Mergel H, Dörner R, Weber T, Jagutzki O, Lüdde H J, Schmidt L and Berakdar J 2001b *Proc. XXII Int. Conf. on Photonic, Electronic and Atomic Collisions (Santa Fe)* p 422

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- Schmidt-Böcking H, Mergel V, Dörner R, Cocke C L, Jagutzki O, Schmidt L, Weber T, Lüdde H J, Weigold E, Berakdar J, Ceredquist H, Schmidt H T, Schuch R and Kheifets A 2003a *Europhys. Lett.* **62** 477–83
- Schmidt-Böcking H, Mergel V, Schmidt L, Dörner R, Jagutzki O, Ullmann K, Weber T, Lüdde H J, Weigold E and Kheifets A 2003b *Radiat. Phys. Chem.* **68** 41–50
- Stefani G 1999 *New Directions in Atomic Physics* ed C T Whelan *et al* (New York: Kluwer) pp 17–32
- Whelan C T 1999 *New Directions in Atomic Physics* ed C T Whelan *et al* (New York: Kluwer) pp 87–104