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New (e,2e) studies of atomic and molecular targets

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Abstract. We report new coplanar (e,2e) measurements characterised by large energy transfer and close to minimum momentum transfer from the projectile to the target. Ionisation of the two-electron targets He and H_2 is investigated under these particular kinematics. The experimental data are compared with the predictions of the most elaborate theoretical models. The obtained good agreement motivated us to extend our research to the case of more complex targets such as Ar. Comparison with the most elaborate models in the case of multi-electron targets is excellent. Destructive and constructive interference effects in the case of H_2 are observed and discussed.

1. Introduction

Investigation of single ionisation (SI) processes by the so-called (e,2e) electron-impact coincidence technique yields important information on both the interaction dynamics and the electronic structure of the target [1,2]. An abundant literature exists reporting such studies under a large variety of kinematical conditions. We address here particular kinematics which has rarely been considered, characterised by large energy transfer and close to minimum momentum transfer from the projectile to the target [3]. The remaining ion carries then a large momentum and hence contributes significantly in the collision process which can therefore not anymore be treated as a binary electron-electron interaction as it is often assumed. The goal of these measurements is first to provide detailed data for electron-impact ionisation of simple targets such as helium and molecular hydrogen which will constitute a severe test for the state-of-the-art models in these particular kinematics. Another interest is to contribute to the development of powerful theoretical approaches for the description of more complex multi-electron atomic and molecular targets in order to elucidate the role of the different interactions in the ionisation process.

The difficulty of these experiments arises from the choice of the kinematical parameters, which do satisfy neither Bethe ridge nor dipolar conditions, resulting in very small cross sections. Incidentally,

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this is the reason why this particular kinematical regime has remained rather unexplored to date. The recently improved sensitivity of our multi-parameter experimental set-up [4] opens up the possibility to perform such difficult experiments.
The detailed description of the spectrometer and experimental procedure are given elsewhere [4,5] and will not be repeated here. A coplanar geometry is used where both electrons are observed in the collision plane defined by the directions of the incident and scattered beam. The energy of the slow ejected electrons (named b) is varied from 205 to 74 and to 37 eV, whereas the scattered electron (named a) is detected in coincidence at fixed energy E_a = 500 eV and fixed scattering angle θ_a = –6° with respect to the incident direction. The incident energy is consequently adjusted to fulfil the energy conservation. Due to the small values of the cross sections, we choose to work with a reduced coincidence energy resolution ΔE_{coinc} = ± 2,5 eV in order to obtain a reasonable signal.
In all this work, no attempt was made to determine the absolute scale for the measured triply differential cross section (TDCS). Hence, these were normalised to the theoretical results.

2. Single ionisation of atoms: He and Ar

We report here (e,2e) TDCS for single ionisation of He and Ar under the chosen kinematics described above. The aim of this part is twofold: first, to validate the experimental procedure (described in [4,5]) using the He data as a standard, and second, to investigate the pertinence of the theoretical models in the case of the "heavier" Ar atom.

2.1. Triple differential cross sections for He

The experimental TDCS distributions are shown in figures 1(a) - (c) for the three investigated ejected electron energies 205, 74 and 37 eV. It is nowadays commonly accepted that the convergent close coupling (CCC) predictions describe very well the TDCS distribution for ionisation of simple targets like He. We compare our experimental data with the results of CCC calculations [6]. Details about the ingredients of this model can be found in [7].
Our measurements are obtained on a relative scale. We use the CCC results to normalise our data to the absolute theoretical scale at the maximum of the binary lobe. We find excellent agreement between experiment and theory, both in the shape of the distribution and in the position of the binary and recoil lobes. The CCC results show a shift of the binary lobe of some 10° from the momentum transfer direction, and so do our data, though at the highest energy the CCC theory yields a slightly smaller shift than experiments. Such peak shifts are consistent with known trends for He [1,2], indicating that the first Born approximation (FBA) is not sufficiently accurate. Such agreement between theory and experiment in He proves that measured TDCS are not affected by systematic experimental artefacts. The small experimental peak observed at about 300° might probably be attributed to spurious backscattering on metal surfaces, but its presence does not alter the conclusions therein made. Therefore, the same experimental procedure can be applied with good confidence for the study of other more complex targets.

2.2. Triple differential cross sections for Ar

The experimental data and theoretical results for Ar ($3p^6$) case are displayed in figure 2 (a)-(c), for the same energies of the ejected electrons 37, 74 and 205 eV as for He. Our results are compared with calculations performed within the distorted wave Born approximation (DWBA), without and with a correction by the Gamow factor (DWBA-G) to account for the post-collision interaction (PCI) [8].
The dashed blue line denotes the DWBA calculations and the solid red line represents the DWBA-G results. Both experiments and the DWBA-G results are normalised to the maximum of the TDCS predicted by DWBA.
One of the most important features of this model is to consider the distorting effect of the target potential on the electrons in the continuum state. For an energetic projectile as in our conditions, we can also consider only single knout-out collisions to eject an electron into the continuum. When the

scattered and ejected electrons are of comparable energies one should also take into account the PCI in the continuum final state. This is done here *via* the so-called Gamow factor.

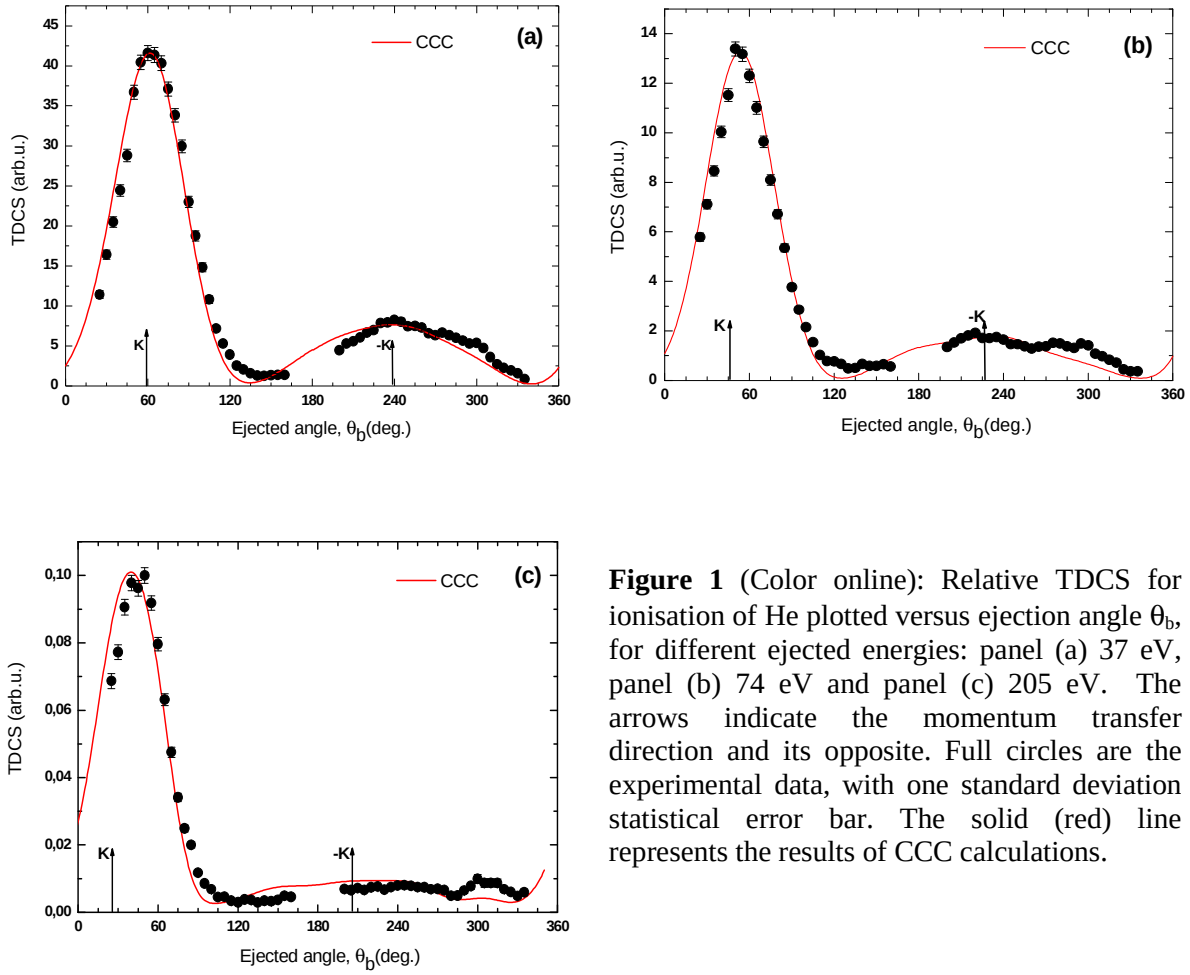


Figure 1 (Color online): Relative TDCS for ionisation of He plotted versus ejection angle θ_b, for different ejected energies: panel (a) 37 eV, panel (b) 74 eV and panel (c) 205 eV. The arrows indicate the momentum transfer direction and its opposite. Full circles are the experimental data, with one standard deviation statistical error bar. The solid (red) line represents the results of CCC calculations.

This factor, also known as the Coulomb density of states, represents the normalisation constant between the two-body Coulomb function describing the interaction of the two continuum electrons in the continuum state [9].

A detailed comparison with our experimental data has already been reported in [8], hence it will not be repeated here. We only want to emphasize the contribution of the Gamow factor in the DWBA model.

In all cases presented here we observe in the experimental data an important intensity of the recoil peak which increases with increasing ejected electron energy relative to the binary peak. The important size of the recoil peak can be attributed to a reflection of the ejected wave from the target potential. The Gamow factor enhances the recoil peak intensity and shifts the binary peak away from the direction of the momentum transfer.

The agreement between theory and experiment is good already for the DWBA model, at least for the two lowest energies, but it further improves when taking into account the Gamow factor. As we see from figure 2, the DWBA predicts an important recoil peak relative to the binary one, especially for 205 eV. The Gamow correction modifies the intensity of the recoil peak, which becomes relatively more important with respect to the binary peak, in agreement with the experiments.

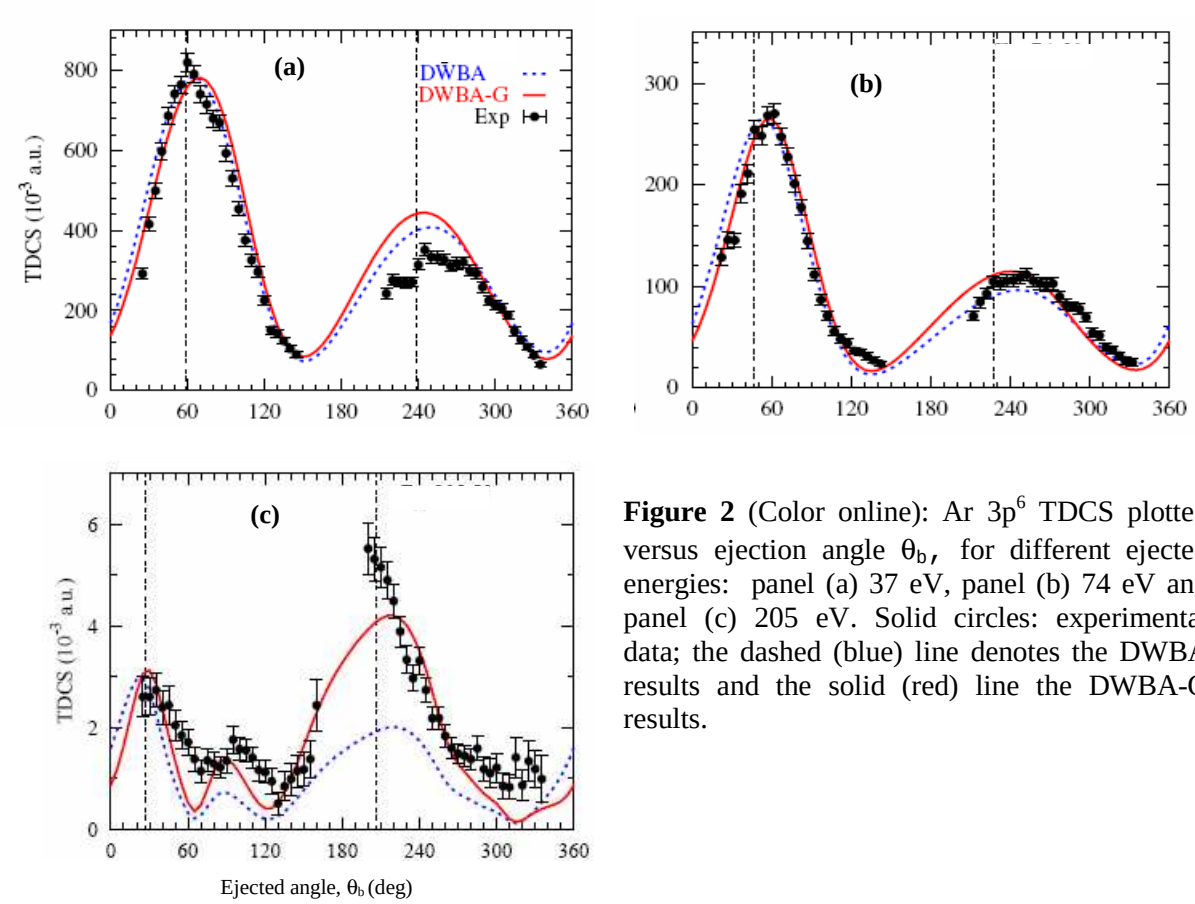


Figure 2 (Color online): Ar $3p^6$ TDCS plotted versus ejection angle θ_b, for different ejected energies: panel (a) 37 eV, panel (b) 74 eV and panel (c) 205 eV. Solid circles: experimental data; the dashed (blue) line denotes the DWBA results and the solid (red) line the DWBA-G results.

Although not shown here, a similar agreement between the DWBA-G and experiment was found for other atomic targets as Ne [8]. The Gamow contribution, accounting for the most important part of the PCI interaction, plays an important role especially for the recoil part of the distributions at higher energies.

3. Single ionisation of molecules: H_2

The experimental results for the TDCS distribution for H_2 are presented in figure 3(a)-(c), for the same three energies of the ejected electrons. The data are compared with theoretical results obtained using two models: the first one is based on the FBA in which a two-centre continuum (TCC) approximation with correct boundary conditions in the entrance and exit channels is applied. The fast incident and the scattered electrons are described by plane waves [10]. This model was previously successfully applied to describe high energy (~ 4.1 keV) (e,2e) processes on H_2 [11]. The second model is the molecular three-body distorted wave (M3DW) coupled with an orientation-averaged molecular orbital (OAMO) approximation [12,13]. The M3DW is a two-centre approach in which the three continuum electrons are described by distorted waves. The electron-electron Coulomb factor is included in the final state wave functions, which means that the final state post collision interactions are included to all orders of perturbation theory. In the OAMO approximation an averaging over all molecular orientations is used for the initial state. This approximation is known to be accurate for a small momentum transferred to the remaining ion.

Both FBA-TCC and M3DW-OAMO models were found to perform well in the intermediate impact energy region and low ejected electron energies, for the ionisation of diatomic (H_2 and N_2) or triatomic molecules. In the present work we investigate a different kinematical regime where a large momentum

transfer to the recoiling ion is involved, implying an active participation of the ion in the collision process.

For the present results, the comparison between experiments and theories shows an overall satisfactory agreement, in particular concerning the shape of the binary peak. The M3DW-OAMO model yields a better description of the binary peak at higher energy whereas the FBA-TCC model seems to be more accurate for the lowest energies. We note a shoulder in the M3DW-OAMO binary peak at 205 eV, which might also be present in the experiments. This shoulder can probably be attributed to final state elastic scattering of the projectile by the target.

The FBA-TCC model does not yield the shift of the binary peak from the momentum transfer direction observed in the experimental data. This is expected from the first Born approximation, which is thus found to be insufficiently accurate under the kinematics investigated here.

Both models fail to describe the intensities of the recoil part of the distributions, except for the FBA-TCC at 74 eV. We even note the vanishing recoil peak for the M3DW model, particularly at 205 eV. This can be explained by the breakdown of the OAMO approximation in the description of the ion contribution at our kinematics.

3.1. Interference effects

A closer inspection of the size of the recoil peak relative to the binary lobe can be done by comparing with the He measurements performed in the same kinematical conditions. We observe for the energies 37 and 74 eV, that the relative intensity of the recoil peak in H_2 is smaller than in the case of He whereas for the highest energy the comparison goes in the opposite way.

This recoil suppression or enhancement in H_2 can be attributed to interference effects [6]. This effect was predicted by Stia *et al* [14] who showed that the effect of the coherent emission from the two molecular centres can be seen in the angular distribution of the ejected electrons. They also predicted that the TDCS angular distribution for molecular hydrogen can be expressed as twice that for atomic hydrogen modulated by an interference factor I:

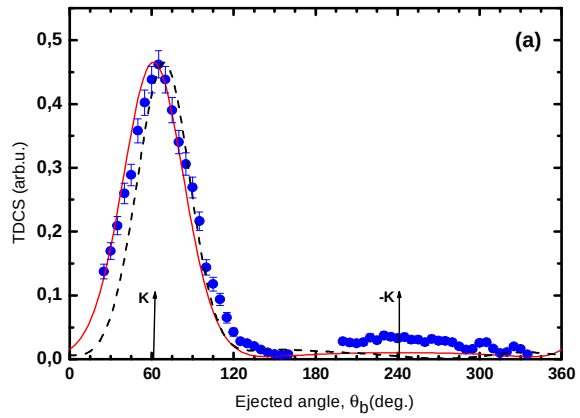
$$\sigma_{e2e}(H_2) = 2 * \sigma_{e2e}(H) * I$$

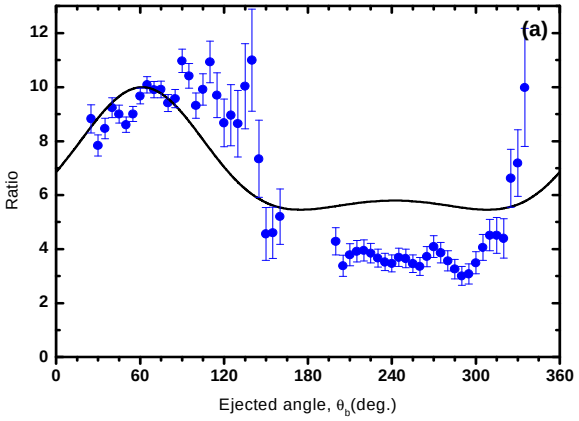
where

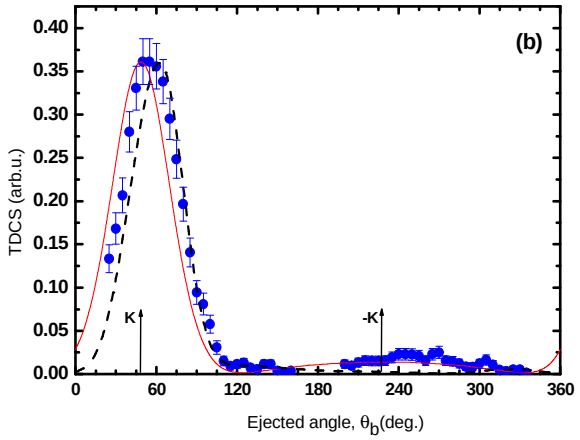
$$I = 1 + \frac{\sin(q\rho)}{q\rho}$$

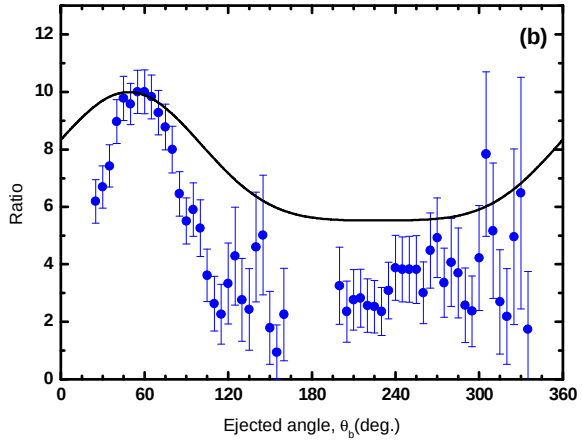
Here q is the momentum of the recoiling ion and ρ the equilibrium internuclear distance in the H_2 molecule. For the experimental data the comparison with atomic hydrogen was not possible, hence we used the distribution for the He atom, which can be considered as an equivalent two-electron single centre atom. The ratio $R = \sigma_{e2e}(H_2) / \sigma_{e2e}(He)$ of the relative TDCS measured for both targets was thus compared with the predicted I factor, and the results are presented in figure 4 (a) – (c). Both R and I values are normalised to an arbitrary value in the region of the binary peak.

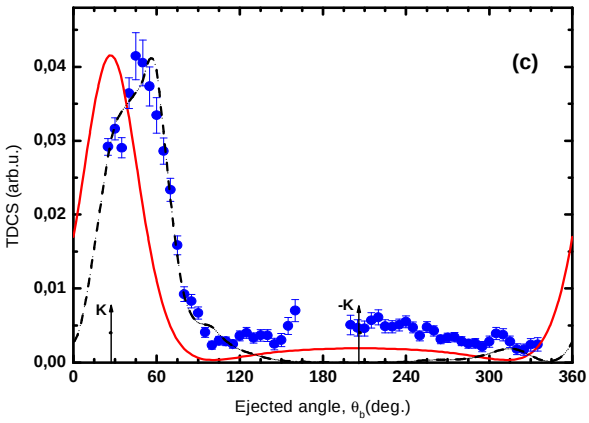
The comparison shows a relatively good agreement taking into account the large error bars in the recoil peak intensities due to the fact that we are taking the ratio of two small quantities in this angular range. The expected oscillatory behaviour can be observed in both the theoretical and experimental interference factor. Even more, we observe a reduction of the recoil peak intensity at lower energies relative to the height of the binary peak and an enhancement at 205 eV. Similar interference effects were previously seen by Milne-Brownlie *et al* [15], but to our best knowledge, this is the first time that both the destructive and constructive characters of the interference process are simultaneously observed in the same (e,2e) experiments.











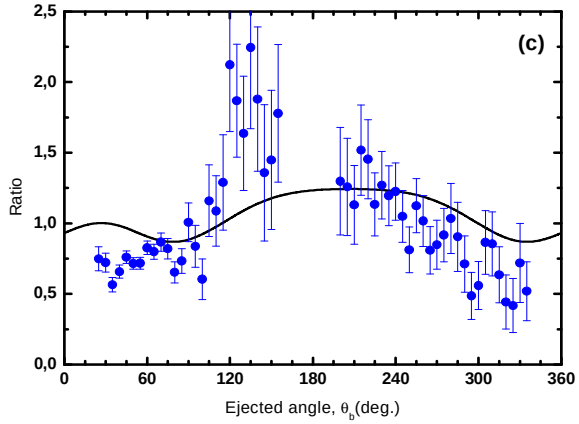


Figure 3 (Color online): TDCS for H_2 represented versus ejected angle θ_b for the ejected energies of 37 eV (panel (a)), 74 eV (panel (b)) and 205 eV (panel (c)). The dotted (black) and full (red) lines represent the theoretical results from M3DW-OAMO and FBA-TCC models. Solid circles: experimental data.

Figure 4 (Color online): Solid circles: the experimental interference factor plotted versus the ejected angle θ_b for the ejected energies of 37 eV (panel (a)), 74 eV (panel (b)) and 205 eV (panel (c)). The full line represents the predicted I factor.

4. Conclusion

(e,2e) TDCS for ionisation of He and Ar atoms and for molecular hydrogen are reported. The data obtained for the ionisation of He show an excellent agreement with the CCC calculations. The Ar case validates the use of the Gamow factor correction into the DWBA model to account for the PCI in the particular kinematics used here.

The H_2 results are compared with the most elaborate available molecular calculations. Reasonable agreement is found for the regions of the angular distributions corresponding to the binary peak. By comparing the He and H_2 results, the molecular effect observed can be a signature of the interferences due to the two molecular centres. A destructive and constructive interference contribution can be seen from the experimental data. However, the discrepancies between the predictions and our data in the recoil region call for a better modelling of the ionisation process.

More experimental data with better resolution and varied kinematics and targets are desirable, and an effort should be made towards determining the absolute scale for the cross section.

5. References

[1] Ehrhardt H, Jung K, Knoth G and Schlemmer P 1986 Z. *Phys. D: At. Mol.Clust.* **1** 3-32
[2] Lahmam-Bennani A 1991 *J. Phys. B: At. Mol. Opt. Phys.* **24** 2401-2442
[3] Catoire F, Staicu Casagrande E M, Nekkab M, Dal Cappello C, Bartschat K and Lahmam-Bennani A 2006 *J. Phys. B: At. Mol. Opt. Phys.* **39** 2827-2838
[4] Catoire F, Staicu Casagrande E M, Lahmam-Bennani A, Duguet A, Naja A, Ren X G, Lohmann B and Avaldi L 2007 *Rev. Sci. Instrum*. **78** 013108
[5] Naja A, Staicu Casagrande E M, Ren X G, Catoire F, Lahmam-Bennani A, Dal Cappello C and Whelan C T 2007 *J. Phys. B: At. Mol. Opt. Phys.* **40** 2871-2884
[6] Staicu Casagrande E M, Naja A, Mezdari F, Lahmam-Bennani A, Bolognesi P, Joulakian B, Chuluunbaatar O, Al-Hagan O, Madison D H, Fursa D V and Bray I 2008 *J.Phys. B: At. Mol. Opt. Phys.* **41** 025204
[7] Bray I, Fursa D V, Kheifets A and Stelbovics A T 2002 *J. Phys. B: At. Mol. Opt. Phys.* **35** R117
[8] A S Kheifets, A Naja, E M Staicu Casagrande and A Lahmam-Bennani 2008 *J. Phys. B: At. Mol. Opt. Phys*. **41** 145201
[9] Brauner B, Briggs J S, Klar H, Broad J T, Rösel T, Jung K and Ehrhardt H 1991 *J. Phys. B: At. Mol. Opt. Phys.* **24** 657-73
[10] Weck P, Fojon O A, Hanssen J, Joulakian B, and Rivarola R D 2001 *Phys. Rev. A* **63** 042709
[11] Stia C R, Fojon O A, Weck P F, Hanssen J, Joulakian B and Rivarola R D 2002 *Phys. Rew. A* **66** 052709
[12] Gao J F, Madison D H and Peacher J L 2005 *Phys. Rev. A* **72** 020701 (R)
[13] Gao J F, Madison D H and Peacher J L 2005 *J. Chem. Phys.* **123** 204314
[14] Stia C R, Fojon O A, Weck P F, Hanssen J and Rivarola R D 2003 *J.Phys. B: At. Mol. Opt. Phys.* **36** L257
[15] Milne-Brownlie D S, Foster M, Gao J, Lohmann B and Madison D H 2006 *Phys. Rev. Lett.* **96** 233201

Acknowledgments

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