INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS B: ATOMIC, MOLECULAR AND OPTICAL PHYSICS

J. Phys. B: At. Mol. Opt. Phys. **39** (2006) 4435–4442 doi:10.1088/0953-4075/39/21/008

Total polarization of the 185 nm emission line of mercury excited by electron impact

G Außendorf[1], F Jüttemann[1], K Muktavat[2], L Sharma[2], R Srivastava[2], A D Stauffer[3], K Bartschat[4], D V Fursa[5], I Bray[5] and G F Hanne[1]

[1] Physikalisches Institut, Universität Münster, Wilhelm-Klemm-Straße 10, D-48149 Münster, Germany

[2] Department of Physics, Indian Institute of Technology Roorkee, Roorkee 247667, India

[3] Department of Physics and Astronomy, York University, Toronto M3J 1P3, Canada

[4] Department of Physics and Astronomy, Drake University, Des Moines, IA 50311, USA

[5] The ARC Centre for Antimatter-Matter Studies, Murdoch University, 90 South Street, Murdoch, Western Australia, 6150, Australia

Received 2 August 2006, in final form 4 September 2006
Published 20 October 2006
Online at stacks.iop.org/JPhysB/39/4435

Abstract

Results for the three Stokes parameters (polarization components) P_1, P_2 and P_3 of the VUV Hg transition $6s6p\,^1P_1 \rightarrow 6s^2\,^1S_0$ (185 nm) obtained from electron–photon (e, eγ) coincidence measurements at electron impact energies of 15 eV, 50 eV and 100 eV (Aussendorf *et al* 2006 *J. Phys. B: At. Mol. Opt. Phys.* **39** 2403) were combined to obtain the total degree of polarization $P_{\rm tot} \equiv \sqrt{P_1^2 + P_2^2 + P_3^2}$. In addition, the Stokes parameter P_4 was measured at an electron impact energy of 15 eV. The measured data are compared with predictions from a fullrelativistic distorted-wave calculation, a five-state semirelativistic Breit–Pauli R-matrix (close-coupling) model, and a convergent close-coupling ansatz, where relativistic effects are accounted for by recoupling of nonrelativistic results. In agreement with the theoretical predictions, no influence of the electron spin was observed for scattering angles $\theta \leqslant 30^\circ$. At 15 eV excitation energy and scattering angles $\theta \geqslant 40^\circ$, however, an increasing importance of spin effects is predicted.

1. Introduction

The Stokes parameters P_1, P_2 and P_3 of the VUV Hg transition $6s6p\,^1P_1 \rightarrow 6s^2\,^1S_0$ (185 nm) have recently been measured in electron–photon (e, eγ) coincidence studies using spin-polarized incident electrons [1]. The above Stokes parameters describe the polarization of photons emitted perpendicular to the scattering plane. The role played by spin effects in the electron impact excitation process can be revealed in such experiments. If spin effects (and hyperfine coupling) are negligible, the total degree of light polarization is $P_{\rm tot} \equiv \sqrt{P_1^2 + P_2^2 + P_3^2} = 1$.

Former measurements at this transition were carried out by Masters *et al* [2] at an incident electron energy of 100 eV and by Murray *et al* [3] at 16 eV. In these experiments, laser-induced fluorescence from a stepwise electron–photon excitation process was detected. The above experimental results for unpolarized electrons showed a significant 'loss of coherence' of the excited state at scattering angles $\theta \leqslant 30^\circ$. Specifically, Masters *et al* [2] observed $P_{tot} \leqslant 0.44$ at 100 eV for the Hg isotope with nuclear spin $I = 0$, i.e., no depolarization due to hyperfine coupling, while Murray *et al* [3] found $P_{tot} \leqslant 0.6$ at 16 eV. This is in strong disagreement with theoretical predictions suggesting that spin effects are negligible at such high energies and small scattering angles. This results in $P_{tot} = 1$ for the Hg ($I = 0$) isotope, independent of the models and approximations used.

It was the purpose of the present investigation to shed light on this long-standing discrepancy. Furthermore, the above results were complemented by measuring the Stokes parameter P_4 for photons emitted in the scattering plane, perpendicular to the incident beam direction.

2. Theoretical considerations

2.1. Scattering geometry and stokes parameters

The electron impact excitation and the optical decay are described in the natural coordinate frame [4, 5]. The scattering plane is spanned by the wavevector $\boldsymbol{k}_0$ of the incident electron (which also defines the x axis) and the wavevector $\boldsymbol{k}_1$ of the scattered electron. The z axis is chosen perpendicular to the scattering plane and serves as the quantization axis of the projections of electron spin (including the polarization vector $\boldsymbol{P}_e$ of the incident electron beam) and the magnetic sublevels of the target. Photons emitted along the $z(P_1, P_2, P_3)$ and $-y(P_4)$ directions are detected in coincidence with the scattered electrons.

The total degree of light polarization is defined as

$$P_{tot} \equiv \sqrt{P_1^2 + P_2^2 + P_3^2}. \tag{1}$$

Here the two linear polarizations $P_{1,2}$ and the circular polarization P_3 were defined in [1] for photons emitted perpendicular to the scattering plane. This parameter is an adequate means to determine whether or not the system after the collision is in a pure quantum state [4–6]. Furthermore, the Stokes parameter P_4, defined as

$$P_4 = \frac{I(0^\circ) - I(90^\circ)}{I(0^\circ) + (90^\circ)}, \tag{2}$$

is measured in the $-y$ direction. Here $I(\beta)$ denotes the light intensity transmitted by a linear polarizer aligned at an angle β with respect to the wavevector $\boldsymbol{k}_0$ of the incident electrons.

2.2. Scattering state and light polarization

Due to the invariance of the interaction with respect to reflection in the scattering plane, the scattering process of a $J = 0 \rightarrow J = 1$ parity changing transition can be described by six independent scattering amplitudes $f(M, m, m_0)$ [5, 7]. Here $m_0 = \pm 1/2$ and $m = \pm 1/2$ denote the spin projections of the incident and the scattered electron, respectively, while M represents the magnetic substate $|J, M\rangle$ of the excited mercury atom. Specifically, the non-vanishing amplitudes in the natural frame are

$$\begin{aligned} &f(+1, +1/2, +1/2), \qquad &f(+1, -1/2, -1/2), \\ &f(-1, +1/2, +1/2), \qquad &f(-1, -1/2, -1/2), \\ &f(0, +1/2, -1/2), \qquad &f(0, -1/2, +1/2). \end{aligned} \tag{3}$$

Here the fixed quantum numbers $J_0 = M_0 = 0$ and $J = 1$ have been omitted. Note that the substates with $M = \pm 1$ can only be excited by non-spin-flip processes ($m = m_0$) while the substate with $M = 0$ can only be excited by spin-flip processes ($m = -m_0$).

If the incident electrons are in a pure spin state $|m_0\rangle$, the asymptotic scattering state can be expressed as

$$\Psi_{\mathrm{scatt},r\to\infty} \to \{f(1, m_0, m_0)|1, +1\rangle|m_0\rangle + f(0, -m_0, m_0)|1, 0\rangle| - m_0\rangle + f(-1, m_0, m_0)|1, -1\rangle|m_0\rangle\} \frac{\mathrm{e}^{\mathrm{i}k_1 r}}{r}, \quad (4)$$

where $\mathrm{e}^{\mathrm{i}k_1 r}/r$ is the radial part of the observed scattered wave.

Equation (4) describes a pure state of the entangled collision system consisting of the excited atom and the scattered electron. Therefore, it can generally not be written as a single product of an atomic state and a state for the scattered electron. This is seen from the appearance of amplitudes for spin-up and spin-down continuum electrons after the collision. As a result, the emitted photons will not be in a single pure state and, when averaged over the final spin projections, $P_{\mathrm{tot}} < 1$ is generally to be expected. However, if spin-flip processes ($m = -m_0$) are negligible, then the scattered wave (4) can be written as a product of an excited atomic and a scattered electron state:

$$\Psi_{\mathrm{scatt},r\to\infty} \to \{f(1, m_0, m_0)|1, +1\rangle + f(-1, m_0, m_0)|1, -1\rangle\}\, |m_0\rangle \frac{\mathrm{e}^{\mathrm{i}k_1 r}}{r}. \quad (5)$$

In this case the atoms will emit completely polarized light, i.e., $P_{\mathrm{tot}} = 1$, in all directions. This situation is sometimes referred to as 'coherent excitation'.

2.3. Geometry and light polarization

Due to the angular characteristics of electric dipole radiation, the $M = 0$ magnetic sublevel will not contribute to the radiation emitted along the quantization axis, i.e., perpendicular to the scattering plane. Therefore, it is impossible to optically detect any population of the $M = 0$ state if we observe photons emitted perpendicular to the scattering plane. Since such an observation can only detect non-spin-flip processes and the electrons are assumed to be initially in a pure state $|m_0\rangle$, equation (5) applies again in that case. Hence, we should always measure $P_{\mathrm{tot}} = 1$ regardless of the population of the $M = 0$ state [5].

As explained above, completely polarized electron impact leads always to $P_{\mathrm{tot}} = 1$. However, the individual Stokes parameters in equation (1) for a given scattering angle can be different for the two spin projections of the incident electrons. Consequently, a mixture of incident spin-up and spin-down electrons can result in a total degree of polarization $P_{\mathrm{tot}} < 1$. If, for example, the Stokes parameters for primary spin-up and spin-down electrons are of the same magnitude but differ in sign, the total polarization for unpolarized incident electrons would yield $P_{\mathrm{tot}} = 0$.

2.4. Hyperfine depolarization

In the present experiment, a natural isotope mixture of mercury atoms with non-vanishing nuclear spin was used as target. Consequently, the measurement averages over a mixture of different nuclear spins. This requires a correction due to the depolarization caused by the hyperfine interaction (see [8]). Instead of $P_{\mathrm{tot}} = 1$, the condition for 'coherent excitation' becomes

$$0.84 \leqslant P_{\mathrm{tot}} \leqslant 0.95. \quad (6)$$

2.5. Relativistic effects and exchange

The scattering process may be spin dependent due to spin–orbit coupling or exchange of the colliding electrons. The 6s6p $^1P_1$ mercury state can be described by an intermediate coupling scheme

$$\Psi(^1P_1) = \alpha\Psi^0(^1P_1) + \beta\Psi^0(^3P_1). \tag{7}$$

The mixing coefficients $\alpha = 0.987$ and $\beta = 0.171$ were given by Lurio [9], with Ψ^0 denoting the pure LS-coupled states. The excitation of the 6s6p $^1P_1$ mercury state can thus be spin dependent not only by spin–orbit interaction in the continuum but also by exchange excitation via the triplet part of the wavefunction.

At small scattering angles, $\theta < 30°$, and an impact energy of 15 eV and higher, however, spin-flips due to either the spin–orbit interaction or electron exchange are expected to be very small. Hence, a nearly coherent population is predicted, i.e., $0.84 \leqslant P_{tot} \leqslant 0.95$ after accounting for hyperfine depolarization effects.

2.6. Meaning of the Stokes parameter P_4

Similar considerations as those outlined in subsection 2.2 lead to the conclusion that $P_4 = 1$ for excitation of the Hg ($I = 0$) isotope if spin effects (electron exchange and spin–orbit interaction for the continuum electron) are negligible. Equation (5) applies again in this case and P_4 is the only Stokes parameter that can be different from zero due to symmetry arguments [10]. However, the natural isotope mixture leads to a much larger range for P_4, namely $0.00 \leqslant P_4 \leqslant 0.85$, even for 'coherent excitation'. Therefore, a 'loss of coherence' in the excitation process can only be shown by direct observation of a spin dependence in the measured Stokes parameter P_4.

3. Results

Details of the experimental setup and of the theories were outlined in [1] and will not be repeated here. Briefly, the measured data are compared with predictions from a fullrelativistic distorted-wave calculation, (RDWBA), a five-state semirelativistic Breit–Pauli R-matrix (close-coupling) model (BPRM-5), and a convergent close-coupling (CCC) ansatz, where relativistic effects originating from target spin–orbit mixing are accounted for by transforming the nonrelativistic results to the intermediate coupling scheme. In the latter calculation channel coupling effects have been taken to convergence.

3.1. Total polarization P_{tot} at 100 eV, 50 eV and 15 eV electron energy

Experimental results of the total degree of polarization P_{tot} for unpolarized electrons are shown in figure 1 and compared with theoretical predictions and the previous measurements. For 100 eV incident energy, only RDWBA and CCC results are available, and both lead to similar results (see figure 1, top row). No significant differences between the two sets of theoretical data are found at scattering angles $5° \leqslant \theta \leqslant 20°$. The present experimental values are significantly closer to the theoretical predictions than the results of Masters *et al* [2], and they do not indicate the presence of significant spin effects. Except for the RDWBA results around $\theta = 70°$, the theoretical numbers lie entirely within the range $0.84 \leqslant P_{tot} \leqslant 0.95$, as required for negligible spin effects. We attribute the trend to slightly lower experimental values, which is also seen for the other energies, to some currently unknown systematic error, rather than to a physical effect. We emphasize that the efficiency and the analysing power

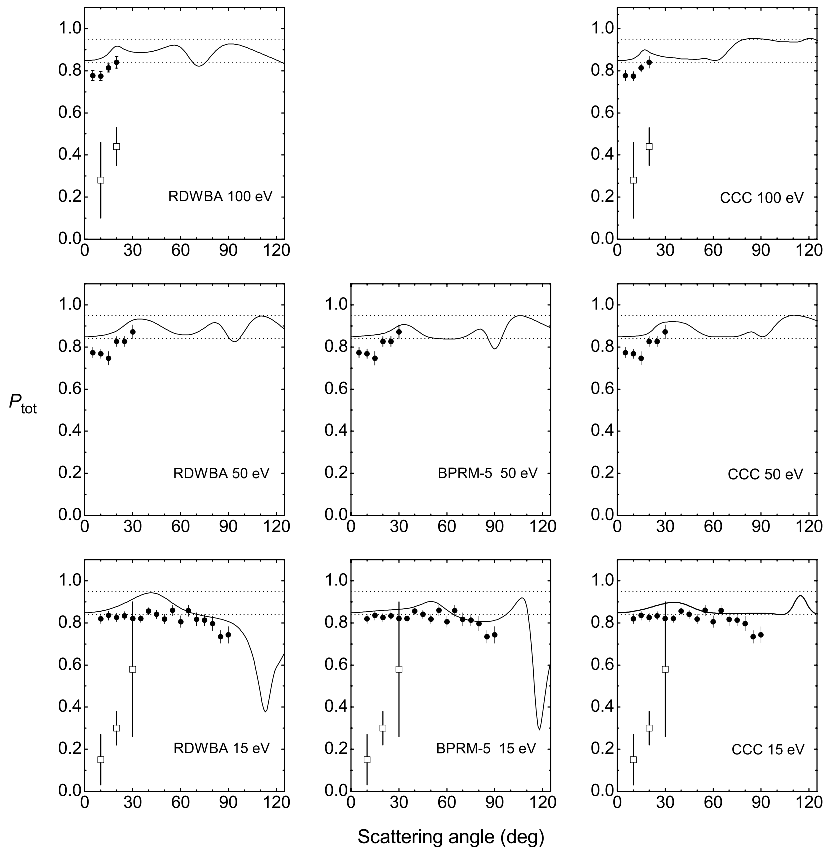

Figure 1. Total degree of polarization P_{tot} of light emitted perpendicular to the scattering plane from the VUV Hg transition $6s6p^1P_1 \rightarrow 6s^2\,{}^1S_0$ (185 nm) after unpolarized electron impact excitation at 100 eV (top row), 50 eV (centre row) and 15 eV (bottom row). The narrow dotted pair of lines (· · · · · ·) represent the theoretical interval $0.84 \leqslant P_{tot} \leqslant 0.95$ (6), while the bullets (•) are the present experimental results. Previous experimental results using laser-induced fluorescence from a stepwise electron–photon excitation process for 100 eV [2] and for 16 eV (bottom row) [3] are shown as open squares. The lines represent the RDWBA, BPRM-5 and CCC results.

of our optical analyser were calibrated carefully. A convolution of the theoretical results with our instrumental energy and angular resolution could not explain this deviation from the expectation either.

Similar conclusions can be drawn from the results for 50 eV (centre row of figure 1). For the measured range of scattering angles $5^\circ \leqslant \theta \leqslant 30^\circ$, the three theoretical approaches give similar results. Significant spin effects are not predicted for $\theta \leqslant 80^\circ$.

The experimental and theoretical results at 15 eV are shown in the bottom row of panels in figure 1. At this energy the theoretical predictions differ significantly from each other at scattering angles $\theta \geqslant 80^\circ$. Within the experimental uncertainty, no deviation from $0.84 \leqslant P_{tot} \leqslant 0.95$ is observed in the angular range $\theta \leqslant 90^\circ$. In the RDWBA and the Breit–Pauli R-matrix calculations, however, deviations between the spin-up and spin-down results start to appear around 40°. This is shown in figure 2, which also includes the experimental results of the spin-resolved P_{tot}. After accounting for the uncertainty of the experimental results, however, no significant spin effect is observed for $\theta \leqslant 90^\circ$. Unfortunately, no experimental data could be obtained for $\theta > 90^\circ$.

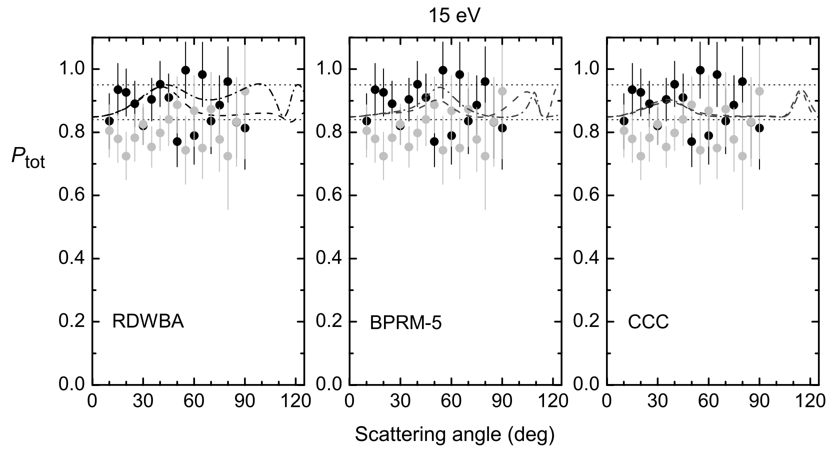

Figure 2. Total degree of polarization P_{tot} of light emitted perpendicular to the scattering plane from the VUV Hg transition 6s6p $^1P_1 \rightarrow 6s^2\ ^1S_0$ (185 nm) after polarized electron impact excitation at 15 eV. The narrow dotted pair of lines (······) represent the theoretical interval $0.84 \leqslant P_{tot} \leqslant 0.95$ (6), while the black and gray bullets are experimental results for spin-up (•) and spin-down (•) incident electrons. The lines represent the RDWBA, BPRM-5 and CCC calculations for spin-up (- - - -) and spin-down (— · — ·) incident electrons.

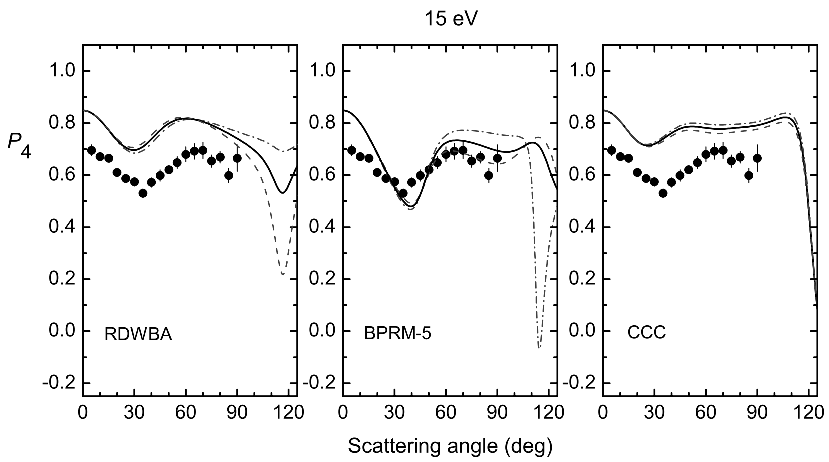

Figure 3. Stokes parameter P_4 of light emitted in the scattering plane perpendicular to the incident beam from the VUV Hg transition 6s6p $^1P_1 \rightarrow 6s^2\ ^1S_0$ (185 nm) after polarized electron impact excitation at 15 eV. (•) present experimental results for unpolarized electrons. The lines represent RDWBA, BPRM-5 and CCC results for unpolarized (——), spin-up (- - - -), and spin-down (— · — ·) electrons.

In summary, a significant 'loss of coherence' of the excited state for excitation with unpolarized electrons at scattering angles $\theta \leqslant 20^\circ$ for 100 eV incident energy, at angles $\theta \leqslant 30^\circ$ for 50 eV, and $\theta \leqslant 60^\circ$ for 15 eV was not found in the present study. These findings are contrary to previous observations at 100 eV and 16 eV by Masters *et al* [2] and Murray *et al* [3], respectively.

3.2. Stokes parameter P_4 at 15 eV electron energy

Figure 3 shows the results for the parameter P_4. The overall agreement in the shape of the curves with the theoretical predictions is good. The observed minimum around $\theta = 35^\circ$ is also predicted by the calculations. The RDWBA and the CCC calculations show somewhat larger values than measured, whereas the BPRM-5 calculation is closer to the experimental data. However, the latter shows a more pronounced minimum than experimentally observed and predicted by the RDWBA and the CCC calculations. The BPRM-5 and RDWBA results suggest some spin dependence of P_4 for larger scattering angles, $\theta \geqslant 50^\circ$ and $\theta \geqslant 80^\circ$, respectively. This is not the case for the CCC calculation, which does not include the spin–orbit interaction explicitly. However, the uncertainty of the experimental data is too large to draw conclusions about possible spin effects. Consequently, no individual experimental data for spin-up and spin-down electrons are shown.

4. Conclusions

The present experimental results lie somewhat lower than the limit $0.84 \leqslant P_{\text{tot}}$ for negligible spin effects. We believe that these deviations should be attributed to a currently unknown systematic error, rather than to a physical effect. Clearly, our results do not support a significant 'loss of coherence' for electron impact excitation of the Hg 6s6p $^1P_1$ state at small scattering angles, which was reported in previous investigations at 100 eV [2] and 16 eV [3]. Within the experimental uncertainty, no significant influence of the electron spin was discovered for scattering angles $\theta \leqslant 30^\circ$ and incident energies of 100 eV, 50 eV and 15 eV. This finding is in good agreement with theoretical predictions using a relativistic distorted-wave Born approximation (RDWBA), a five-state semi-relativistic Breit–Pauli *R*-matrix model (BPRM-5), and a convergent close-coupling (CCC) approach with recoupling of non-relativistic scattering amplitudes. At 15 eV incident energy and scattering angles $\theta \geqslant 40^\circ$, an increasing importance of spin effects is predicted by the RDWBA and BPRM-5 calculations, both of which include the spin–orbit interaction explicitly. However, the experimental accuracy was not sufficient to either confirm or reject these predictions.

Similar conclusions can be drawn from the results of the parameter P_4. Here the agreement between experiment and theory is satisfactory in all cases.

Acknowledgments

The experimental work was supported by the Deutsche Forschungsgemeinschaft (GA, FJ, GFH). The RDWBA calculations were supported by grants from the Council of Scientific and Industrial Research of India (RS, KM) and the Natural Sciences and Engineering Research Council of Canada (ADS). DVF and IB would like to acknowledge support of the Australian Research Council. Support from the United States National Science Foundation under grant PHY-0244470 (KB) is also acknowledged.

References

[1] Außendorf G, Jüttemann F, Muktavat K, Sharma L, Srivastava R, Stauffer A D, Bartschat K, Fursa D V, Bray I and Hanne G F 2006 *J. Phys. B: At. Mol. Opt. Phys.* **39** 2403

[2] Masters A T, MacGillivray W R, Standage M C and Humphrey I 1997 *20th Int. Conf. on the Physics of Electronic and Atomic Collisions: Abstracts of Contributed Papers TH032* ed F Aumayr, G Betz and H P Winter Vienna

[3] Murray A J, Pascual R, MacGillivray W R and Standage M C 1992 *J. Phys. B: At. Mol. Opt. Phys.* **25** 1915

[4] Andersen N, Bartschat K, Broad J T and Hertel I V 1997 *Phys. Rep.* **279** 251
[5] Andersen N and Bartschat K 2001 *Polarization, Alignment, and Orientation in Atomic Collisions* (New York: Springer)
[6] Blum K 1996 *Density Matrix Theory and Applications* 2nd edn (New York: Plenum)
[7] Andersen N and Bartschat K 1994 *J. Phys. B: At. Mol. Opt. Phys.* **27** 3189
Andersen N and Bartschat K 1994 *J. Phys. B: At. Mol. Opt. Phys.* **29** 1149 (corrigendum)
[8] Wolcke A, Bartschat K, Blum K, Borgmann H, Hanne G F and Kessler J 1983 *J. Phys. B: At. Mol. Phys.* **16** 639
[9] Lurio A 1965 *Phys. Rev.* A **140** 1505
[10] Sohn M and Hanne G F 1992 *J. Phys. B: At. Mol. Opt. Phys.* **25** 4627