J. Phys. B: At. Mol. Opt. Phys. **27** (1994) 3967–3974. Printed in the UK

L x-ray production in La, Nd, Gd, Er and Lu by 1–5 MeV protons

Y C Yu, E K Lin, C W Wang, P J Tsai and C H Chang
Institute of Physics, Academia Sinica, Taipei, Taiwan, Republic of China

Received 7 March 1994, in final form 13 June 1994

Abstract. Cross sections for production of L_α, L_β, L_γ and L_ℓ x-rays in lanthanum, neodymium, gadolinium, erbium and lutetium were measured with proton bombardment in the energy range 1–5 MeV. These cross sections and their ratios are found to be, in general, in good agreement with the ECPSSR (energy loss and Coulomb deflection effects, perturbed stationary state approximation with relativistic correction) theory.

1. Introduction

In the last few decades the inner shell ionization of atom by charged particles has been measured extensively. These studies have proven to be very important in understanding the complex mechanism that are involved in the ion–atom collisions. The literature for L x-ray studies with protons as reviewed a decade ago (Sokhi and Crumpton 1984) indicated a limited number of experimental data especially for rare-earth elements. Most of the published data were scattered through different groups and reported only in graphical form. This makes the comparison of experimental and theoretical L x-ray production cross sections more difficult. Subsequently, Braziewicz *et al* (1991) measured L x-ray production by light ion bombardment, in which the production cross sections in rare-earth elements by 0.15–4.0 MeV protons were reported. The elements Gd and Lu were not included in their investigation. The data of the last decade on L-shell cross section for proton impact were summarized recently in tabular form by Orlic *et al* (1994). Their compilation adds new data for rare-earth elements ionized by protons; for La and Nd at energies lower than 4.6 MeV; for Gd, lower than 2.0 MeV; for Er, lower than 3.4 MeV. No L x-ray production data for Lu by protons were reported.

The ECPSSR theory by Brandt and Lapicki (1981) of inner-shell ionization, which accounts for energy loss (E) and Coulomb deflection (C) of the ion with perturbed stationary state (PSS) wavefunction and relativistic mass (R) of the electron, has often been compared to the data. A test of theories and their possible modification clearly requires more data with various projectiles at different velocities for a wide range of elements. In this paper, we present the L x-ray production cross sections for five targets, La, Nd, Gd, Er and Lu, by 1–5 MeV protons, extending the investigated energy range to higher energy at 5.0 MeV. The individual L_α, L_β, L_γ and L_ℓ x-ray production cross sections with L_β/L_α, L_γ/L_α and L_ℓ/L_α intensity ratios were extracted from experiment and compared with predictions of the ECPSSR theory. For the cases investigated the electron capture contribution to the L x-ray production cross sections was estimated in the ECPSSR calculation (Lapicki and McDaniel 1980, 1981) to be less than 0.1%.

0953-4075/94/173967+08$19.50

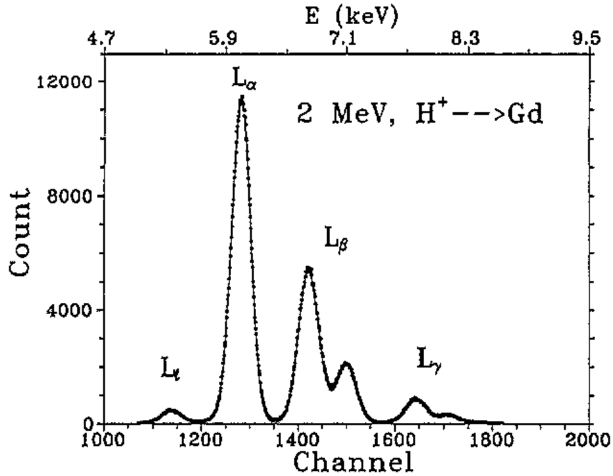

Figure 1. Typical spectrum for L x-ray yield in Gd bombarded by 2 MeV protons. The smooth curve shows the fitted spectrum.

2. Experimental procedure

Proton beams were produced by the 3 MV tandem accelerator of our institute. The details of the experimental set-up are described elsewhere (Lin *et al* 1992). Therefore, only a brief description shall be presented here. Thin targets (around 15 μg cm^{-2}) were prepared by vacuum evaporation and deposition of the elements of interest onto 10 μg cm^{-2} carbon foils. A Canberra HpGe x-ray detector with a resolution of 185 eV at 5.9 keV was mounted at 60° to the incident beam direction. Two 50 mm^2 silicon surface barrier detectors were mounted at 120° and 170° to the incident beam direction for counting ions elastically scattered from the target. A Faraday cup was located at the end of the beam line to measure current transmitted through the target.

The areas of x-ray peaks were evaluated using the 1993 version of the GUPIX program (Maxwell *et al* 1989). This program used a digital filter to eliminate background, and then employed a non-linear least squares fitting procedure to give a complete analysis of each L x-ray spectrum. The efficiency of the x-ray detector was obtained from K x-rays emitted from thin targets ($Z = 22$–31) bombarded by 2–3 MeV protons and normalized to the cross sections of the ECPSSR theory. Additionally, the efficiency determined with radioactive ^{57}Co, ^{65}Zn, ^{75}Se, ^{88}Y and ^{241}Am calibrated sources allowed for absolute normalization of the efficiency curve. Taking into account the uncertainties in the efficiency of the x-ray detector, the particle detector solid angle, counting statistics and peak fitting procedure, the overall uncertainties in this experiment are estimated to be about 12%. Figure 1 shows a typical spectrum for L x-ray yield in Gd bombarded by 2 MeV protons. It is seen that the spectrum is very clean and a good fit to the data for L_α, L_β, L_γ and L_ℓ is obtained.

3. Results and discussion

In order to compare the experiment to the theory, the theoretical ionization cross sections were converted to x-ray production cross sections by the following relations (Close *et al* 1973):

$$\sigma_{L\alpha} = [\sigma_{L1}(f_{13} + f_{12}f_{23}) + \sigma_{L2}f_{23} + \sigma_{L3}]\omega_3 F_{3\alpha}$$

$$\sigma_{L\beta} = \sigma_{L1}\omega_1 F_{1\beta} + (\sigma_{L1} f_{12} + \sigma_{L2})\omega_2 F_{2\beta} + [\sigma_{L1}(f_{13} + f_{12} f_{23}) + \sigma_{L2} f_{23} + \sigma_{L3}]\omega_3 F_{3\beta}$$

$$\sigma_{L\gamma} = \sigma_{L1}\omega_1 F_{1\gamma} + (\sigma_{L1} f_{12} + \sigma_{L2})\omega_2 F_{2\gamma}$$

$$\sigma_{L\ell} = [\sigma_{L1}(f_{13} + f_{12} f_{23}) + \sigma_{L2} f_{23} + \sigma_{L3}]\omega_3 F_{3\ell}$$

where σ_{L1}, σ_{L2} and σ_{L3} are the subshell ionization cross sections for the L_1, L_2 and L_3 subshells, respectively; ω_i the fluorescence yields for the three subshells; f_{ij} is the Coster–Kronig radiationless yields for subshells i to j; $F_{i\alpha}$, $F_{i\beta}$, $F_{i\gamma}$, $F_{i\ell}$ are the fractional radiative widths involving the L_i subshell. In the present calculations, the values of ω_i and f_{ij} are taken from Krause (1979), while the fractional radiative widths are from the tables of Campbell and Wang (1989) which were based on the code of Scofield (1974).

The measured L_α, L_β, L_γ, L_ℓ and the total L x-ray production cross sections of protons incident upon La, Nd, Gd, Er and Lu targets are listed in table 1. The measured cross sections for the total L-shell x-ray productions appear to be in very good agreement with the ECPSSR theory as seen in figure 2. The L_ℓ contribution over the investigated energy range was found to be about 2% of the total L x-ray production cross sections.

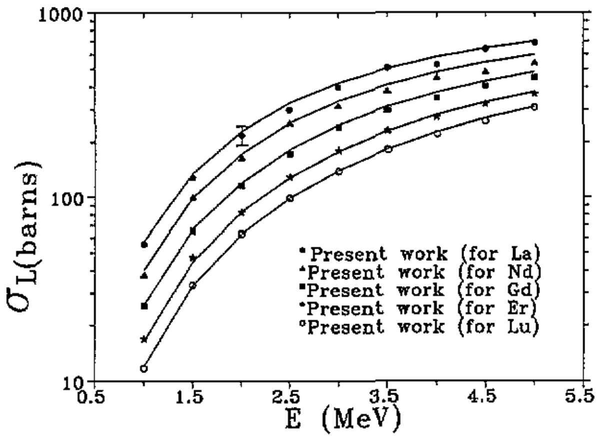

Figure 2. Measured cross sections for the total L-shell x-ray productions in La, Nd, Gd, Er and Lu. The curves represent the results for the ECPSSR theory.

Figures 3 and 4 represent the ratios L_α, L_β, L_γ and L_ℓ of the experimental to theoretical x-ray production cross sections as a function of ion beam energy for all the elements studied in this work. Data from other investigations (Khan *et al* 1978, Avaldi *et al* 1984, Jesus *et al* 1987, Kadom-Al-Neami *et al* 1990, Vigilante *et al* 1991, Braziewicz *et al* 1991) are also shown for comparison, except for the L_ℓ data of Braziewicz *et al* (1991) which were taken from the compilation of Orlic *et al* (1994). These data appear to be somewhat abnormal and were not considered here. Generally, the ECPSSR theory leads to good agreement for all targets and incident energies studied in this work. It is noted that theoretical predictions are subject to the uncertainties associated with the uncertainties of fluorescence yield, Coster–Kronig radiationless yield and fractional radiative width used in the translation from ionization cross sections to x-ray production cross sections.

For lanthanum, our result for L_α, L_β and L_γ x-ray production cross sections are consistent (within experimental uncertainties) with the data of Kandom-Al-Neami *et al*

Table 1. Measured L x-ray production cross sections (in b) by protons.

Z	E (MeV)	L_α	L_β	L_γ	L_ℓ	L
$_{57}$La	1.0	34.0	18.0	2.46	1.23	55.7
	1.5	77.1	41.4	6.43	3.01	128
	2.0	126	74.1	13.1	4.73	218
	2.5	171	104	19.0	6.48	300
	3.0	222	142	26.6	8.47	399
	3.5	279	183	31.9	10.6	505
	4.0	289	194	34.7	10.7	528
	4.5	344	241	40.3	12.7	638
	5.0	369	255	50.5	13.5	688
$_{60}$Nd	1.0	22.3	13.1	1.79	0.84	38.0
	1.5	55.9	37.3	5.16	2.09	100
	2.0	89.8	61.7	9.77	3.32	165
	2.5	137	96.7	15.2	5.00	254
	3.0	169	120	21.0	6.33	316
	3.5	194	154	25.7	7.26	381
	4.0	236	174	32.2	8.65	451
	4.5	251	188	36.6	9.23	485
	5.0	277	214	40.8	10.2	542
$_{64}$Gd	1.0	15.1	8.63	1.21	0.60	25.5
	1.5	37.2	23.4	3.51	1.48	65.6
	2.0	63.6	42.9	7.32	2.53	116
	2.5	92.4	65.9	11.3	3.71	173
	3.0	126	92.9	16.3	5.05	240
	3.5	154	117	21.0	6.15	298
	4.0	179	138	25.1	7.03	349
	4.5	208	160	29.4	7.68	405
	5.0	229	179	33.2	8.82	450
$_{68}$Er	1.0	10.0	5.70	0.83	0.39	16.9
	1.5	27.3	16.3	2.43	1.02	47.1
	2.0	45.0	30.2	5.16	1.81	82.2
	2.5	68.9	48.9	8.89	2.65	129
	3.0	94.1	68.8	13.2	3.63	180
	3.5	119	89.4	17.9	4.71	231
	4.0	139	108	21.1	6.06	274
	4.5	162	129	25.0	6.43	322
	5.0	182	148	29.1	7.14	366
$_{71}$Lu	1.0	6.78	3.99	0.59	0.31	11.7
	1.5	18.9	11.7	1.85	0.87	33.3
	2.0	34.6	23.0	3.95	1.60	63.2
	2.5	53.5	36.5	6.39	2.48	98.8
	3.0	74.5	51.4	9.39	3.46	139
	3.5	95.8	68.7	13.1	4.39	182
	4.0	114	85.0	16.9	5.21	221
	4.5	132	100	20.7	5.98	259
	5.0	158	120	24.5	6.86	309

(1990). The data of Vigilante *et al* (1991) for L_β lie approximately 30% above our data at 1.8 MeV. The values reported by Avaldi *et al* (1984) for L_γ are 25% below this work in the energy range 1–2 MeV. As compared to the work by Braziewicz *et al* (1991), their data

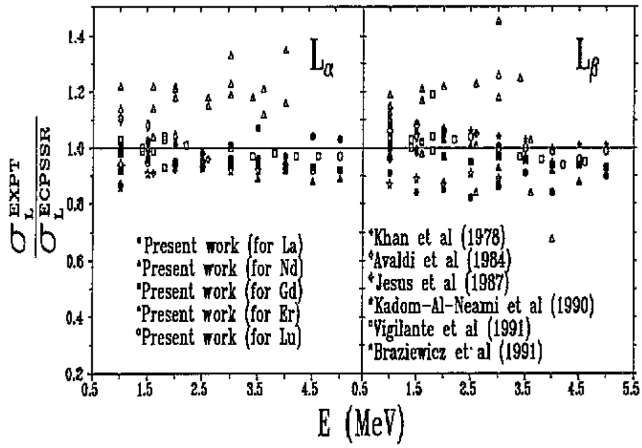

Figure 3. Ratios of L_α and L_β of the experimental to the theoretical production cross sections in La, Nd, Gd, Er and Lu as a function of incident proton energy.

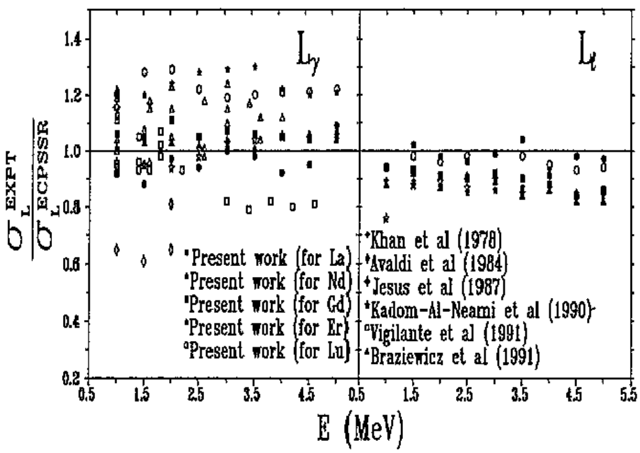

Figure 4. Ratios of L_γ and L_ℓ of the experimental to the theoretical production cross sections in La, Nd, Gd, Er and Lu as a function of incident proton energy.

for L_α, L_β and L_γ lie above our data by approximately 20%, 15% and 10%, respectively.

For neodymium, the present results for L_α and L_β are in good agreement with previous results of Khan *et al* (1978) and Vigilante *et al* (1991) for energies from 1 to 3 MeV and 1.0 to 4.6 MeV, respectively. The data of Vigilante *et al* (1991) for L_γ fall about 20% below ours for energies at and above 3.0 MeV. Good agreement is seen between the data of Braziewicz *et al* (1991) for L_γ and ours except for energies below 1.6 MeV. However, for L_α and L_β there appear some discrepancies of 30% and 12%, respectively, between their data and ours.

For gadolinium, the current data for the L_α and L_β cross sections match well with the data of Avaldi *et al* (1984) and Vigilante *et al* (1991) for energies from 1 to 2 MeV and 1.0 to 1.8 MeV, respectively. The data of Jesus *et al* (1987) for L_α and L_γ agree quite well with our data at 1.0 MeV. The values obtained by Avaldi *et al* (1984) for L_γ are in fair agreement with ours except for at energy 2.0 MeV.

For erbium, our data for L_α and L_β agree with the previous measurements by Vigilante *et al* (1991) in the energy range 1.0–1.8 MeV. However, their results for L_γ lie approximately 20% below this work. The data of Braziewicz *et al* (1991) for L_γ are in good agreement with our data, while their data for L_α and L_β lie approximately 15% higher. Our experimental values for L_α, L_β and L_ℓ x-ray production are in good agreement with the ECPSSR theory, while the data for L_γ lie 20% above the theory. These findings of discrepancy are consistent with those reported earlier by Cohen (1990).

For lutetium, the results are similar for gadolinium and erbium. There is a consistent deviation between our data for L_γ x-ray production and the predictions of the ECPSSR theory. No experimental cross sections for lutetium by 1–5 MeV proton impact were reported previously.

The comparison of experimental and theoretical L x-ray intensity ratios L_β/L_α and L_γ/L_α are illustrated in figures 5 and 6, where the results for La and Lu were chosen for representative display. The ratios are shown as a function of v_1/v_{2L}, where v_1 is the proton velocity and v_{2L} is the orbital electron velocity. The electron velocity is calculated from the Bohr model with $v_{2L} = v_0 Z_{2L}/2$, where $Z_{2L} = Z_2 - 4.15$ is the screened target atomic number and v_0 is the Bohr velocity. The curve represents the intensity ratios calculated from the ECPSSR theory.

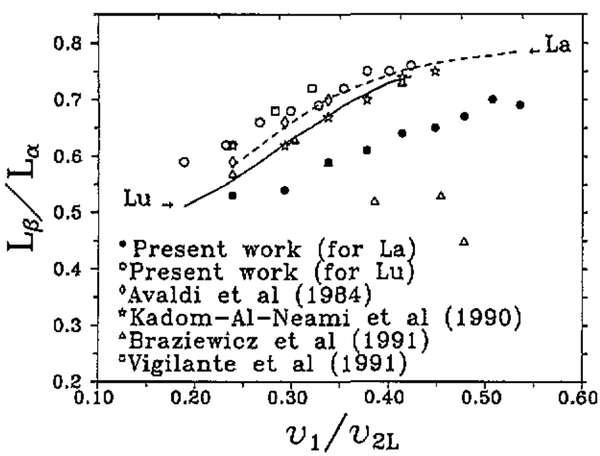

Figure 5. Comparison of experimental and theoretical L x-ray intensity ratios L_β/L_α of La and Lu as a function of v_1/v_{2L}. The broken and full curves represent the results for the ECPSSR theory in La and Lu, respectively.

With the exception of a slight underprediction for the La, Er and Lu data, our obtained L_β/L_α intensity ratios for Nd and Gd agree well with the ECPSSR theory. For La, the measurement by Braziewicz *et al* (1991) gives their L_β/L_α intensity ratios considerably lower than the ECPSSR predictions for energies 2.6, 3.6 and 4.0 MeV. For the L_γ/L_α intensity ratios, the present data lie systematically higher than the ECPSSR ratios with the discrepancy up to 30% except for the La data; for La, the present and earlier measurements gave values of the ratio L_γ/L_α close to the ECPSSR predictions, except for the data of Avaldi *et al* (1984) which are comparatively lower by 30%. For Lu, the present data are new; the ECPSSR theory seems to underpredict the L_γ/L_α data by about 20% over the entire energy range investigated.

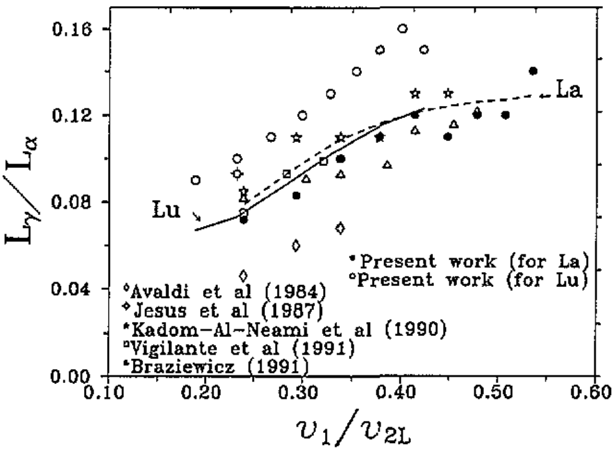

Figure 6. Comparison of experimental and theoretical L x-ray intensity ratios L_γ/L_α of La and Lu as a function of v_1/v_{2L}. The broken and full curves represent the results for the ECPSSR theory in La and Lu, respectively.

The L_ℓ/L_α intensity ratios of La and Lu plotted in figure 7 are, in principle, ionization theory and projectile energy independent and thus test directly calculations of radiation transition rates. Our results agree with the data of Kandom-Al-Neami *et al* (1990) for L_ℓ in the energy range 1.5–3.0 MeV. Except for Er, where some data scatter and 15% discrepancies are observed, the calculation based on Scofield's theory, as tabulated by Campbell and Wang (1989), gives a straight line as shown in figure 7 and is well verified by our data.

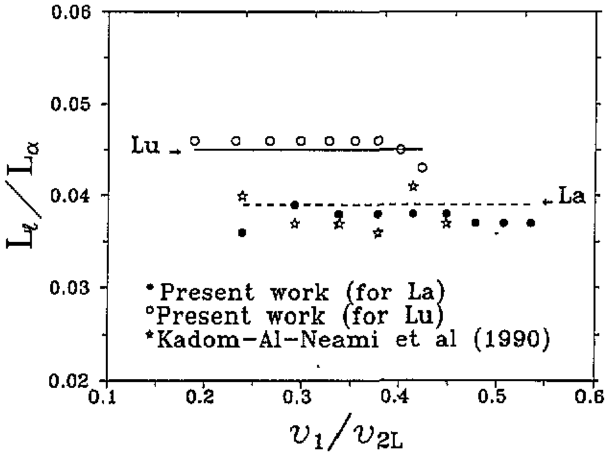

Figure 7. Measured L x-ray intensity ratios L_ℓ/L_α of La and Lu as a function of v_1/v_{2L}. The straight lines represent the calculation for the emission rates based on Scofield's theory as tabulated by Campbell and Wang (1989).

4. Conclusion

The individual L_α, L_β, L_γ and L_ℓ x-ray production cross sections in La, Nd, Gd, Er and Lu have been measured for 1–5 MeV protons and are compared to theoretical predictions of the ECPSSR theory. The ECPSSR theory reproduces the experimental data quite well except for a slight underprediction for the L_γ x-ray production measurements and it seems to be adequate for the rare-earth elements studied in this work. The measured L x-ray intensity ratios are in overall good agreement with the ECPSSR theory. Recently, Vigilante *et al* (1990) suggested a modified ECPSSR theory in which the binding energy saturates at the binding energy of a united atom in slow collisions. In collision systems under our consideration, however, no such saturation occurs and the ECPSSR-UA of Vigilante *et al* (1990) is identical with the standard ECPSSR of Brandt and Lapicki (1981). A calculation of the ratio σ (ECPSSR-UA)/σ(ECPSSR) shows that, except for Er, no ratio was found to be larger than 1.03, which is well within experimental uncertainties.

It is noted that the use of a different atomic parameters data base could lead to significant discrepancies, as previously reported by several authors (e.g. Cohen 1984, Campbell 1988). Therefore, more careful measurements of L x-ray production cross section and intensity ratios as well as a comprehensive review of atomic parameters are clearly needed for further theoretical studies on the inner-shell ionization mechanism.

Acknowledgments

We would like to thank Professor G Lapicki for providing us with the theoretical calculations and for discussions during his visit to our Institute. Thanks are also due to the accelerator group at the Institute of Physics for their technical assistance. This work is partially supported by the National Science Council of the Republic of China under contract NSC 82-0208-M001-057.

References

Avaldi L, Mitchell I V, Eschbach H L and Domba W 1984 *J. Phys. B: At. Mol. Phys.* **17** 2851
Brandt W and Lapicki G 1981 *Phys. Rev.* A **23** 1717
Braziewicz E, Braziewicz J, Czyzewski T, Glowacka L, Jaskola M, Kauer T, Kobzev A P, Pajek M and Trautmann D 1991 *J. Phys. B: At. Mol. Opt. Phys.* **24** 1669
Campbell J L 1988 *Nucl. Instrum. Methods* B **31** 518
Campbell J L and Wang J-X 1989 *At. Data Nucl. Data Tables* **43** 281
Close D A, Bearse R C, Malanify J J and Umbarger C J 1973 *Phys. Rev.* A **8** 1873
Cohen D D 1984 *J. Phys. B: At. Mol. Phys.* **17** 3913
—— 1990 *Nucl. Instrum. Methods* B **49** 1
Jesus A P, Ribeiro J P and Lopes J S 1987 *J. Physique Coll.* **9** 235
Kadom-Al-Neami A, Al-Neami A, Bordas M, Larcher J and Heitz C 1990 *Int. J. Particle-Induced X-ray Emission* **1** 63
Khan M R, Hopkins A G and Crumpton D 1978 *Z. Phys.* A **288** 133
Krause M O 1979 *J. Phys. Chem. Ref. Data* **8** 307
Lapicki G and McDaniel F D 1980 *Phys. Rev.* A **22** 1896
—— 1981 *Phys. Rev.* A **23** 975E
Lin E K, Wang C W, Yu Y C, Chuang F T and Huang Y M 1992 *Phys. Scr.* **46** 522
Maxwell J A, Campbell J L and Teesdal W J 1989 *Nucl. Instrum. Methods* B **43** 218
Orlic I, Sow C H and Tang S M 1994 *At. Data Nucl. Data Tables* **56** 159
Sokhi R S and Crumpton D 1984 *At. Data Nucl. Data Tables* **30** 49
Scofield J H 1974 *At. Data Nucl. Data Tables* **14** 121
Vigilante M, Cuzzocrea P, de Cezare N, Murolo F, Perillo E and Spadaccini G 1990 *Nucl. Instrum. Methods* B **51** 232
—— 1991 *J. Phys. B: At. Mol. Opt. Phys.* **24** 2521