PHYSICAL REVIEW A VOLUME 47, NUMBER 4 APRIL 1993

Doubly differential cross section for the ionization of the hydrogen molecule by the impact of 100-eV electrons

A. H. Al-Nasir, M. A. Chaudhry,* A. J. Duncan, R. Hippler,† and H. Kleinpoppen
Unit of Atomic and Molecular Physics, University of Stirling, Stirling FK9 4LA, Scotland

(Received 24 September 1992)

Doubly differential cross sections (DDCS) for the molecular single ($H_2{}^+$) and dissociative single ($H+H^+$) ionization of the hydrogen molecule have been measured employing the ejected-electron and produced-ion coincidence technique for an incident electron energy of 100 eV and ejected-electron energies between 20 and 80 eV. Angular variations of partial and total DDCS's have also been measured in experiments in which electrons are detected at angles between 2° and 110° relative to the direction of the incident electrons. A comparison of the present measurements has been made with the data from the published literature.

PACS number(s): 34.80.Gs, 35.80.+s, 35.20.Wg

INTRODUCTION

The hydrogen molecule being the simplest molecule has been extensively studied both experimentally and theoretically. Investigations have been made for single- [1–4], double- [1,2,4–6], and dissociative single- [1,4,7–15] ionization cross sections of molecular hydrogen by electron impact. Also, measurements of singly and doubly differential cross sections for electron ejection which do not discriminate between single or double ionization have been reported in the literature [16–18]. This paper presents measurements of partial doubly differential cross sections (PDDCS) for molecular single ionization [PDDCS ($H_2{}^+$)], dissociative single ionization [PDDCS ($H+H^+$) or more simply PDDCS (H^+)], and doubly differential cross sections (DDCS) [=PDDCS ($H_2{}^+$) plus PDDCS (H^+)]. Dissociative double ionization, often referred to simply as double ionization [PDDCS($H^+ + H^+$)], will be the subject of another paper [19]. These investigations have been carried out by using the ejected electron and produced-ion coincidence technique [20]. In this crossed-beam-type experiment an incident electron energy of 100 eV was used. Ejected- (variously called secondary or inelastically scattered) electron energies between 20 and 80 eV were detected. Measurements are presented for the angular variations of the PDDCS and DDCS for electron ejections at angles between 2° and 110° relative to the incident-beam direction.

EXPERIMENTAL TECHNIQUE

The experimental technique employed is similar to that described previously [20]. Briefly, an energetic beam of electrons is allowed to cross a dilute beam of molecular hydrogen which diffuses out of a multicapillary array. From the resulting ionization events electrons ejected in a particular direction are energy analyzed by a 30° parallel-plate electrostatic analyzer and detected by a channeltron. The produced ions are extracted from the interaction region by an electrostatic field and are analyzed by a time-of-flight-type (TOF) analyzer, being finally detected by a channeltron after their acceleration to an energy of 3 keV. Amplified pulses from the electron analyzer act as the start pulses while the pulses from the ion analyzer act as stops for the time-to-digital converter (TDC). In parallel with the conventional TDC a multihit type of TDC is used to allow collision events resulting in the production of only one ion to be distinguished from events leading to the production of more than one ion. Figure 1 shows a typical TOF spectrum obtained and the area under any peak, after subtraction of random coincidences, gives the true number of coincidences $N_c^{(n)}$

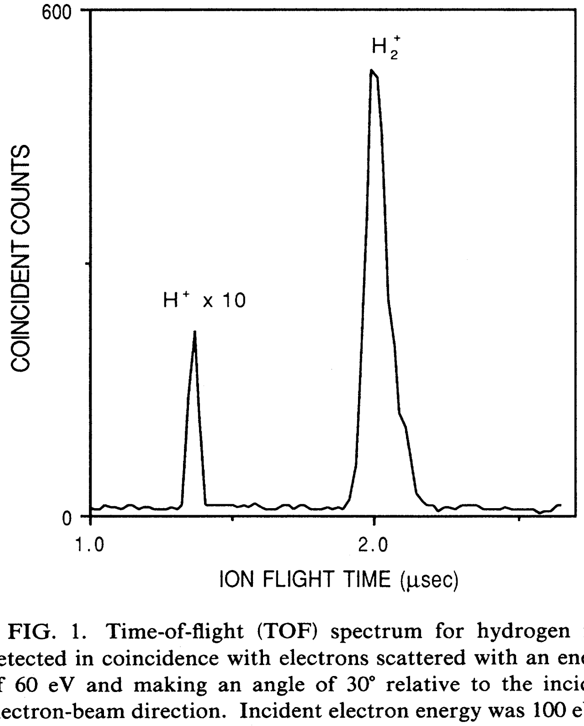

FIG. 1. Time-of-flight (TOF) spectrum for hydrogen ions detected in coincidence with electrons scattered with an energy of 60 eV and making an angle of 30° relative to the incident electron-beam direction. Incident electron energy was 100 eV.

which is related to the partial doubly differential cross section for ionization $d^2\sigma^{(n)}/(dE\,d\Omega)$ by

$$\frac{d^2\sigma^{(n)}}{dE\,d\Omega}=\frac{N_c^{(n)}}{N_i}\frac{\sigma_i}{\Delta E\Delta\Omega\epsilon_\delta}\ ,$$

where n refers to a particular ionization state, σ_i is the total cross section for ionization, N_i is the number of detected ions, and ϵ_δ is the efficiency of the electron-detection system, while ΔE is the energy bandwidth of the electron analyzer and $\Delta\Omega$ is the solid angle subtended by the entrance aperture of the electron analyzer at the interaction region. The electron analyzer was operated with a resolution of about 10% and had an acceptance angle of approximately 0.002 sr. The value for σ_i is taken from the data of Kossmann, Schwarzkopf, and Schmidt [1]. It may be mentioned that the present measurements for angles of electron ejection less than 10° were made possible only by moving the Faraday cup out of the way. However, due to the coincident nature of our experiment there was no measurable increase in the PDDCS values due to the scattered electrons. Also due to the ion-extraction field the ejected electrons tend to move out of the plane of the experiment. Measurements, therefore, for angles less than 10° in the horizontal plane were made only for 60 and 80 eV ejected-electron energies since the deflections in the vertical plane for these electrons were equal to an angle of approximately only 2°.

RESULTS AND DISCUSSION

Figures 2–5 show the present measurements of PDDCS (H_2^+), PDDCS (H^+), and DDCS while Fig. 6 shows the measurements of DDCS as a function of the angle of electron detection Θ_δ, measured relative to the incident-beam direction, for detected electron energies E_δ of 20, 30, 40, 60, and 80 eV, respectively, and for an incident electron energy E_i of 100 eV. Tables I–III show together in a tabulated form the present measurements of

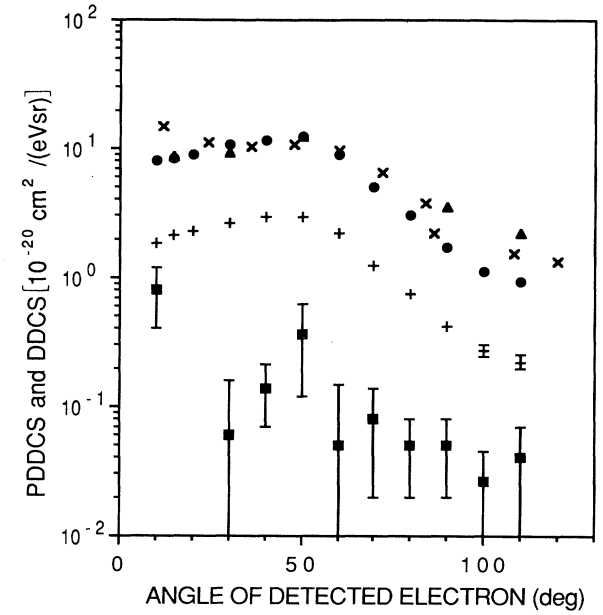

FIG. 3. Present measurements of (+) PDDCS (H_2^+)/4, (■) PDDCS (H^+), and (●) DDCS as a function of Θ_δ for $E_i=100$ eV and $E_\delta=30$ eV. The symbols (×) and (▲) show the published values of Shyn, Sharp, and Kim [18] and DuBois and Rudd [17], respectively.

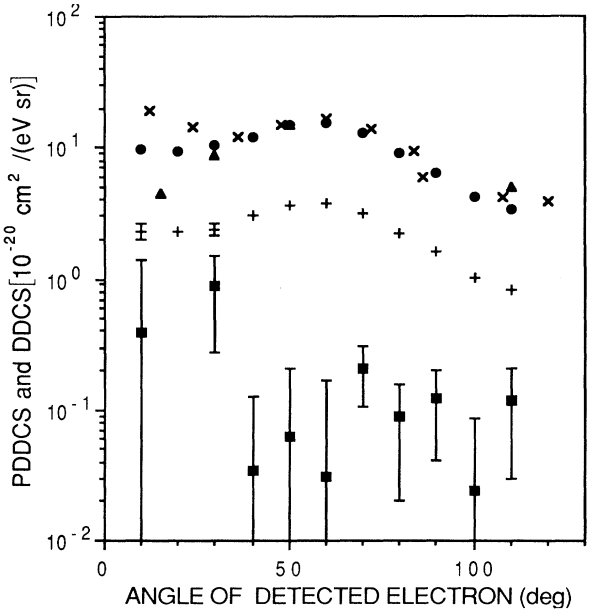

FIG. 2. The present measurements of (+) PDDCS (H_2^+)/4, (■) PDDCS (H^+), and (●) DDCS as a function of Θ_δ for $E_i=100$ eV and $E_\delta=20$ eV. The symbols (×) and (▲) show the published values of DDCS from Shyn, Sharp, and Kim [18] and DuBois and Rudd [17], respectively.

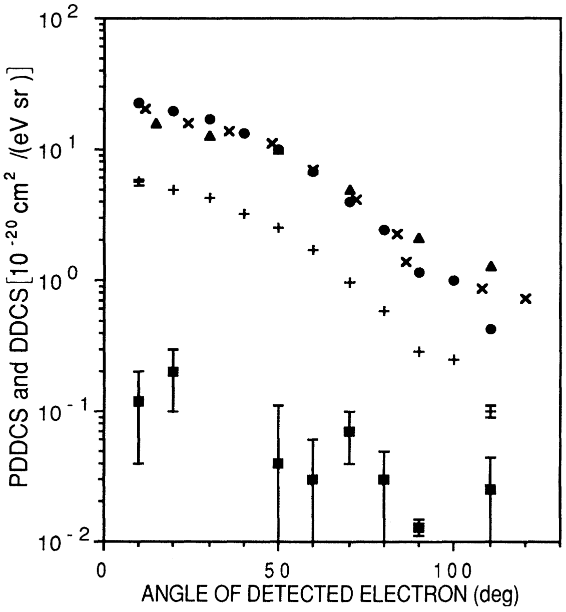

FIG. 4. Present measurements of (+) PDDCS (H_2^+)/4, (■) PDDCS (H^+), and (●) DDCS as a function of Θ_δ for $E_i=100$ eV and $E_\delta=40$ eV. The symbols (×) and (▲) show the published results of DDCS from Shyn, Sharp, and Kim [18] and DuBois and Rudd [17], respectively.

TABLE I. Present measurements of DDCS [=PDDCS (H_2^+) plus PDDCS (H^+)] for the ionization of molecular hydrogen by the impact of 100-eV electrons [in units of 10^{-20} cm^2/(eV sr)].

E_δ (eV) Θ_δ (deg)	20	30	40	60	80
2				654.70±26.00	8802.00±184.00
5				577.30±23.00	5887.00±135.00
10	9.66±1.70	8.27±0.80	22.42±1.30	416.00±10.60	2664.00±53.00
15		8.42±0.50		274.30±9.78	1014.00±13.60
20	9.32±0.70	8.99±0.38	19.60±0.60	151.60±1.68	320.60±5.10
30	10.35±1.09	10.76±0.40	17.30±0.70	53.90±1.00	57.60±2.30
40	12.03±0.44	11.74±0.50	13.07±0.51	15.60±0.50	21.10±1.30
50	14.79±0.60	12.30±0.73	10.00±0.44	8.10±0.48	9.10±1.00
60	15.27±0.53	8.88±0.40	6.88±0.34	3.45±0.28	5.80±0.80
70	12.87±0.52	5.08±0.20	3.92±0.26		2.20±0.47
80	8.97±0.40	3.05±0.20	2.38±0.14		1.40±0.20
90	6.46±0.38	1.75±0.16	1.15±0.14	0.72±0.11	0.50±0.18
100	4.12±0.20	1.13±0.10	1.00±0.08	0.50±0.10	
110	3.40±0.30	0.94±0.11	0.43±0.04		0.30±0.13
115				0.24±0.08	

TABLE II. Present measurements of PDDCS (H_2^+) for the ionization of molecular hydrogen by the impact of 100-eV electrons [in units of 10^{-20} cm^2/(eV sr)].

E_δ (eV) Θ_δ (deg)	20	30	40	60	80
2				654.70±26.0	8802.00±184.0
5				573.70±21.8	5887.00±135.0
10	9.26±1.40	7.47±0.70	22.30±1.30	416.00±10.6	2664.00±53.0
15		8.42±0.50		272.40±9.70	1014.00±13.6
20	9.32±0.70	8.99±0.38	19.40±0.60	150.40±1.67	320.60±5.10
30	9.48±0.90	10.70±0.40	17.30±0.70	53.40±1.00	57.60±2.30
40	12.00±0.40	11.60±0.50	13.07±0.51	15.60±0.50	21.10±1.30
50	14.73±0.57	11.93±0.70	9.96±0.43	7.90±0.44	9.10±1.00
60	15.24±0.50	8.83±0.39	6.85±0.34	3.30±0.27	5.80±0.80
70	12.67±0.50	5.00±0.19	3.85±0.26		2.20±0.47
80	8.88±0.40	3.00±0.20	2.35±0.14		1.40±0.20
90	6.36±0.38	1.70±0.16	1.14±0.14	0.64±0.10	0.50±0.18
100	4.10±0.20	1.10±0.10	1.00±0.08	0.47±0.10	
110	3.28±0.30	0.90±0.11	0.40±0.04		0.30±0.13
115				0.22±0.08	

TABLE III. Present measurements of PDDCS (H^+) for the dissociative single ionization of molecular hydrogen by the impact of 100-eV electrons [in units of 10^{-20} cm^2/(eV sr)].

E_δ (eV) Θ_δ (deg)	20	30	40	60
5				3.60±8.00
10	0.40±1.00	0.80±0.40	0.12±0.08	
15				1.90±1.30
20			0.20±0.10	1.20±0.20
30	0.87±0.60	0.06±0.10		0.50±0.16
40	0.03±0.09	0.14±0.07		
50	0.06±0.10	0.37±0.25	0.04±0.07	0.20±0.17
60	0.03±0.13	0.05±0.10	0.03±0.03	0.15±0.10
70	0.20±0.10	0.08±0.06	0.07±0.03	
80	0.09±0.70	0.05±0.03	0.03±0.02	
90	0.10±0.08	0.05±0.03	0.01±0.02	0.08±0.05
100	0.02±0.06	0.03±0.02		0.03±0.04
110	0.12±0.08	0.04±0.03	0.03±0.02	
115				0.02±0.01

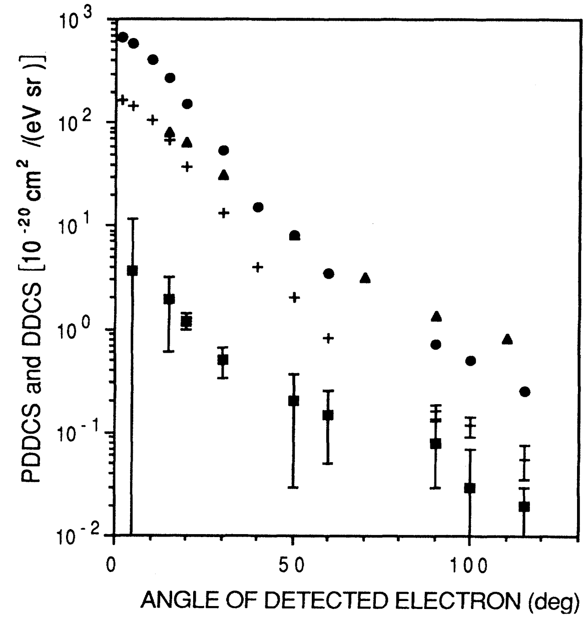

FIG. 5. Present measurements of (+) PDDCS (H_2^+)/4, (■) PDDCS (H^+), and (●) DDCS as a function of Θ_δ for $E_i = 100$ eV and $E_\delta = 60$ eV. The symbol (▲) shows the published results of DDCS from DuBois and Rudd [17].

DDCS, PDDCS (H_2^+), and PPDCS (H^+), respectively. Figures 2–6 also show for comparison the data of DuBois and Rudd [17] and Shyn, Sharp, and Kim [18] for DDCS. The present measurements for different detected electron energies have been normalized to the data of DuBois and Rudd [17] for $\Theta_\delta = 50°$.

First of all, examining Figs. 2–6, it is clear that, in general, PDDCS (H^+) is much less than PDDCS (H_2^+), although there is a considerable variation in the ratio of these two quantities with the angle of the detected electron. The form of the curves for DDCS is, therefore, not significantly affected by the presence of dissociative single ionization. Looking at the results for DDCS alone, there is a broad general agreement between the present results and those of DuBois and Rudd [17] and Shyn, Sharp, and Kim [18]. The agreement is particularly good in Fig. 4 corresponding to a detected electron energy of 40 eV. However, there are significant discrepancies at other detected electron energies. For example, Fig. 2 shows that, for $\Theta_\delta < 40°$, the present values of DDCS are relatively lower than those of Shyn, Sharp, and Kim [18], but higher than those of DuBois and Rudd [17]. Also in Fig. 3, for $\Theta_\delta < 30°$, it can be seen that, although the present measurements agree with those of DuBois and Rudd [17], they are lower than those found by Shyn, Sharp, and Kim [18]. The difference between the present results and those of DuBois and Rudd [17] shown in Fig. 5, which give the results of measurement at a detected electron energy of 60 eV, is quite large. In this case much stronger scattering in the forward direction and a more rapid decrease in the DDCS with increasing angle is indicated by the present results. A similar trend is indicated in Fig. 6 which shows the results of measurement for a detected electron energy of 60 eV.

An interesting feature of the measurements is the broad maximum which occurs in the present results as well as in the data of DuBois and Rudd [17] and Shyn, Sharp, and Kim [18]. Due to the law of conservation of

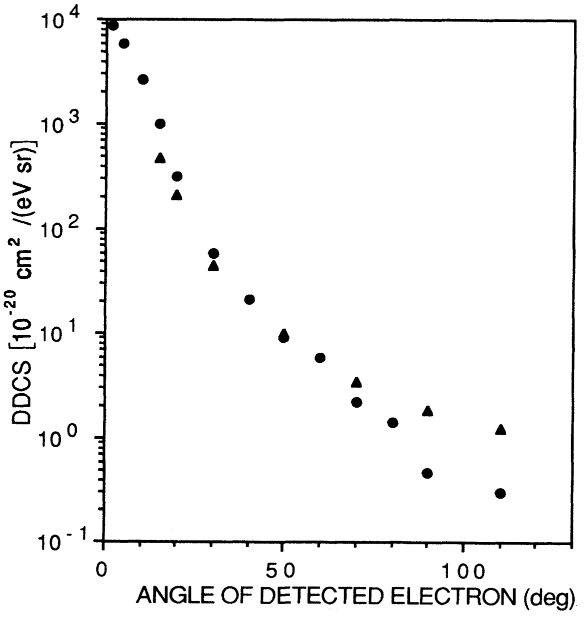

FIG. 6. Present measurements of (●) DDCS as a function of Θ_δ for $E_i = 100$ eV and $E_\delta = 80$ eV. The symbol (▲) shows the published results of DDCS from DuBois and Rudd [17].

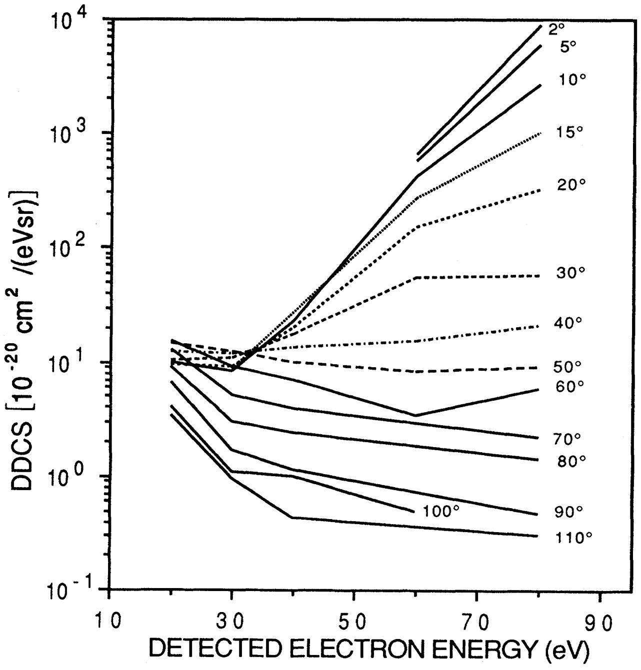

FIG. 7. Present measurements of DDCS as a function of E_δ for different values of Θ_δ from 2° to 110° relative to the incident electron-beam direction.

momentum in the binary collision between an incident electron and an atomic electron, a broad maximum is, in fact, expected [21–23] in the cross section at an angle given approximately by

$$E_\delta = E_i \cos^2\Theta_\delta - I \ ,$$

where I in eV is the binding energy of the molecular electrons. Using the value of 15.43 eV for I from Shyn, Sharp, and Kim [18], for $E_i = 100$ eV and $E_\delta = 20$ eV the maximum is expected at $\Theta_\delta = 53.5°$, whereas for $E_i = 100$ eV and $E_\delta = 30$ eV, the maximum is expected at $\Theta_\delta = 47.6°$. In Figs. 2 and 3, corresponding to measurements with $E_i = 100$ eV and $E_\delta = 20$ and 30 eV, respectively, the maxima occur at angles $\Theta_\delta = 60 \pm 5°$ and $50 \pm 5°$ in reasonable agreement with the above prediction. The above equation and the results also show that the maximum in Figs. 2 and 3 shifts to lower angles as the energy of the detected electron increases so that eventually the maximum becomes obscured by the strong increase in the electron scattering in the forward direction for larger values of the detected electron energy.

Figure 7 summarizes the present results for DDCS as a function of E_δ for different angles of electron detection. The measured values of DDCS are relatively higher for lower Θ_δ and higher E_δ values. This result is quite understandable since electrons having higher energies are known to be preferentially scattered in the forward direction.

CONCLUSION

Doubly differential cross sections for the molecular single- and dissociative single-ionization of the hydrogen molecule have been measured simultaneously using the ejected-electron and produced-ion coincidence technique for an incident electron energy of 100 eV and detected electron energies between 20 and 80 eV. The angular variation of the doubly differential cross section has also been measured in experiments in which electrons are detected at angles between 2° and 110° with the direction of the incident electrons.

It has been shown as expected that, in general, the cross section for dissociative single ionization is much less than that for molecular single ionization although quite large variation in the ratio of the two quantities occurs as the angle of the detected electron varies. Also, although the results for the total doubly differential cross section show general agreement with those of DuBois and Rudd [17] and Shyn, Sharp, and Kim [18], significant discrepancies have appeared, particularly for electron scattering in the forward direction.

Finally it should be observed that although the hydrogen molecule is the simplest molecule no theoretical calculation as yet appears to exist with which to compare the above measurements.

ACKNOWLEDGMENTS

The authors wish to acknowledge the financial support given to this research program by the Science and Engineering Research Council, London. Financial assistance by the North Atlantic Treaty Organization for collaboration between the universities of Bielefeld and Stirling is also acknowledged.

---

*Permanent address: Centre for Advanced Studies in Physics, Government College, P.O. Box 1750, Lahore, Pakistan.

†Permanent address: Fakultät für Physik, Universität Bielefeld, D-4800 Bielefeld, Germany.

[1] H. Kossmann, O. Schwarzkopf, and V. Schmidt, J. Phys. B **23**, 301 (1990).

[2] A. K. Edwards, R. M. Wood, and A. S. Beard, Phys. Rev. A **37**, 3697 (1988).

[3] D. Rapp and P. Englander-Golden, J. Chem. Phys. **43**, 1464 (1965).

[4] H. Tawara, Y. Itikawa, H. Nishimura, and M. Yoshino, J. Phys. Chem. Ref. Data **19**, 617 (1990).

[5] A. K. Edwards, R. M. Wood, J. L. Davis, and R. L. Ezell, Nucl. Instrum. Methods Phys. Res. B **40/41**, 174 (1989).

[6] K. E. McCulloh and H. M. Rosenstock, J. Chem. Phys. **48**, 2084 (1968).

[7] G. H. Dunn and L. J. Kieffer, Phys. Rev. **132**, 2109 (1963).

[8] H. Cho, K. C. Hsieh, and L. C. McIntyre, Jr., Phys. Rev. A **33**, 2290 (1986).

[9] A. Crowe and J. W. McConkey, J. Phys. B **6**, 2088 (1973).

[10] L. J. Kieffer and G. H. Dunn, Phys. Rev. **158**, 61 (1967).

[11] R. J. Van Brunt and L. J. Kieffer, Phys. Rev. A **2**, 1293 (1970).

[12] B. Brehm and G. De Frenes, Int. J. Mass Spectrom. Ion Phys. **26**, 25 (1978).

[13] K. Köllmann, Int. J. Mass Spectrom. Ion Phys. **17**, 261 (1975).

[14] K. Köllmann, J. Phys. B **11**, 339 (1978).

[15] A. Crowe and J. W. McConkey, Phys. Rev. Lett. **31**, 192 (1973).

[16] C. B. Opal, E. C. Beaty, and W. K. Peterson, At. Data **4**, 209 (1972).

[17] R. D. DuBois and M. E. Rudd, Phys. Rev. A **17**, 843 (1978).

[18] T. W. Shyn, W. E. Sharp, and Y.-K. Kim, Phys. Rev. A **24**, 79 (1981).

[19] M. A. Chaudhry, A. H. Al-Nasir, A. J. Duncan, R. Hippler, and H. Kleinpoppen (unpublished).

[20] M. A. Chaudhry, A. J. Duncan, R. Hippler, and H. Kleinpoppen, Phys. Rev. Lett. **59**, 2036 (1987); Phys. Rev. A **39**, 530 (1989); **41**, 4056 (1990).

[21] G. N. Ogurtsov, Zh. Eksp. Teor. Fiz. **64**, 1149 (1973) [Sov. Phys. JETP **37**, 584 (1973)].

[22] Yong-Ki Kim, Phys. Rev. A **28**, 656 (1983).

[23] M. E. Rudd, Phys. Rev. A **44**, 1644 (1991).