3-1998

Double Photoionization of Helium

James A.R. Samson
Behlen Laboratory of Physics

Wayne C. Stolte
University of Nevada, Las Vegas, wcstolte@lbl.gov

Z. X. He
IBM, Inc.

Y. Lu
Behlen Laboratory of Physics

J. N. Cutler
Wright Patterson AFB

See next page for additional authors

Authors
James A.R. Samson, Wayne C. Stolte, Z. X. He, Y. Lu, J. N. Cutler, and R. J. Bartlett

Double photoionization of helium

J. A. R. Samson, W. C. Stolte, Z.-X. He,* J. N. Cutler,† and Y. Lu
Department of Physics and Astronomy, University of Nebraska, Lincoln, Nebraska 68588-0111

R. J. Bartlett
Los Alamos National Laboratory, Los Alamos, New Mexico 87545

(Received 19 September 1997)

The cross sections for double photoionization of helium and the ratios of double to single ionization have been measured from the double-ionization threshold to 820 eV. The results are in very good agreement with several recent calculations. [S1050-2947(98)05403-1]

PACS number(s): 32.80.Fb

I. INTRODUCTION

Calculations of multiple photoionization cross sections of an atom necessarily must involve electron correlation processes. Helium with only two electrons presents the simplest atom to study, yet determinations of its double-photoionization cross sections and of the ratio of double to single ionization have proven to be difficult. Similarly, accurate experimental values of these cross sections have proven to be difficult to obtain.

The experimental determination of double-photoionization cross section of He requires accurate measurements of the total photoionization cross section and of the ratio of double to single ionization. This year marks the 30th anniversary of the first measurement [1] and first calculation [2] of this ratio. During this period, calculations of the magnitude of the ratio [3–22] have varied by as much as 50% from one another. Among the experimental results [23–33] the maximum variation has been about 40% at the peak value of the ratio. However, in the past few years the theoretical and experimental results have been converging toward each other. The relative values of many of the experimentally determined ratios, measured as a function of the incident photon energy, are in reasonable agreement. Whereas, their absolute values differ greatly. This is evidence of the presence of systematic errors. These errors will be discussed in detail in Sec. II.

Our original measurements of the double-ionization cross sections and of the $I(\mathrm{He}^{2+})/I(\mathrm{He}^{+})$ ratio were reported in 1992 [30]. These results were about 30% lower than the existing data at that time. Thus, in order to compare the relative values of our ratio to the other experimental data we multiplied our results by 1.3 (the data presented for the double-ionization cross section were not subjected to this multiplication factor). Since the publication of our earlier data, we repeated the measurements of the ratio many times and under a variety of conditions in an attempt to understand and eliminate all possible systematic errors and to produce accurate values for the $I(\mathrm{He}^{2+})/I(\mathrm{He}^{+})$ ratio. Our early measurements were made at the National Synchrotron Light Source (NSLS) at Brookhaven, NY, whereas our present data were taken at the Synchrotron Radiation Center (SRC), Stoughton, WI, and at the Advanced Light Source (ALS), Berkeley, CA. We found that our new data are reproducible in the 1–3 % range, and in most cases the statistical errors lie within the same range. Also, during this time period we published precision values of the total photoionization cross sections of He [34], which combined with our new data allows us to present precision measurements of the double-photoionization cross sections of He. This provides benchmark data for comparison with the several new theoretical results published in the last two or three years.

II. EXPERIMENT

The $I(\mathrm{He}^{2+})/I(\mathrm{He}^{+})$ ratio was measured with a time-of-flight (TOF) mass spectrometer which was used in the space focusing mode [35]. The total distance traveled by the ions was about 8 cm, and the total time to travel that distance by He^{+} was about 200 ns. The voltages on the various elements were chosen so that the He^{+} ions would strike the front surface of a microchannel plate (MCP) with an energy between 4 and 5 keV. The output pulses from the MCP were fed into conventional electronic counting equipment. However, in some cases simple discriminators were used to suppress electronic noise, and in other measurements constant fraction discriminators were used. In all cases the discrimination level was kept as low as possible. Gas pressures were typically in the 10^{-6}-Torr range. The dwell time at each data point was sufficient to provide counting statistics in the 1–3 % range. At SRC, measurements were made with the single electron bunch mode (300-ns period). The ion signal was used to start the time-to-amplitude converter (TAC), and a signal from the synchrotron ring provided a reliable stop pulse for the TAC. At ALS, measurements were made with the two bunch mode (328-ns period) with similar start-stop pulse techniques.

Different types of monochromators were used at the various synchrotron facilities, all with quite different types of diffraction gratings, for example, a variable line-spaced plane grating, a spherical laminar profile type, and spherical blazed gratings. The incident light on the gratings came from either an undulator or a bending magnet beamline. Therefore, our measurements were all made with monochromatic radiation of different quality with respect to the amount of scattered radiation and higher-order spectra present in the

*Present address: IBM, Inc., Burlington, VT 05452.
†Present address: Wright Patterson AFB, Dayton, OH 45433.

1050-2947/98/57(3)/1906(6)/$15.00

TABLE I. The following filters were used just below an absorption edge. The thickness of the filters ranged from 300 to 700 nm.

Filter	Energy range (eV)	Filter	Energy range (eV)
Al	50–70	Ti	360–440
Si	80–98	Cr	500–580
B	100–184	Ni	600–820

emerging beam. However, to eliminate or minimize this unwanted radiation, a series of filters was used as listed in Table I. This precaution essentially eliminated a major source of systematic error. Several other sources of systematic errors have been identified and their effects on the ratio of double to single charged ions are discussed below.

(i) Several investigators [26,36,37] observed that the double to single charged rare-gas ion ratios produced in TOF mass spectrometers increase with increasing background gas pressure. This effect can be understood if the cross section for charge transfer and/or momentum transfer is greater for the singly charged ions. Thus, the rate of loss for He^+ is greater than that for He^{2+}. That this is the case for all the rare gases can be seen from the compilation of ionic collision cross sections produced by Barnett [38]. A more recent experiment by Bruce and Bonham [37] measured these specific cross sections for Ar^+ and Ar^{2+} in order to explain their TOF observations that the $I(He^{2+})/I(He^+)$ ratio increased with gas pressure.

(ii) When a fixed ion input signal is incident on the front surface of a MCP multiplier, the output count rate increases as a function of the ion input energy, reaching a plateau at about 3.5–4 keV for either singly or doubly charged ions [37]. This occurs presumably because the number of secondary electrons emitted is now more than sufficient to saturate the MCP output signal. Thus a plateau is reached in the count rate vs impact energy curve. However, below this saturation level the detection efficiency of singly charged ions appears to decrease more rapidly than for doubly charged ions, causing an increase in the measured double-to-single-charge ratio. Measurements of the ratio should, therefore, be made when both are in the saturated mode. Even then care must be taken because the output-pulse-height distributions from the MCP detector produced by different ion species are likely to be different, as discussed below.

(iii) If the output pulse height distribution for He^+ detection is different from that of He^{2+}, the observed ratio will depend on the discriminator setting [37,39]. Because the He^{2+} ions strike the MCP with an energy twice that of He^+ the pulse height distribution maximum for He^{2+} may be shifted to higher values. If the discriminator threshold level is set too high, then more counts from the singly charged ions will be lost relative to that of the doubly charged ions. This will produce a ratio that is too high.

(iv) Care has to be taken to eliminate the presence of any stray electrons from the vicinity of the TOF because these electrons can be accelerated into the ionization target area causing secondary ionization, which produces He^+ and He^{2+} ions. Nagy, Skutlartz, and Schmidt [40] have shown that the $I(He^{2+})/I(He^+)$ ratio produced by electron impact is much smaller than that produced by photon impact. Thus this effect would cause the observed ratio to be too small.

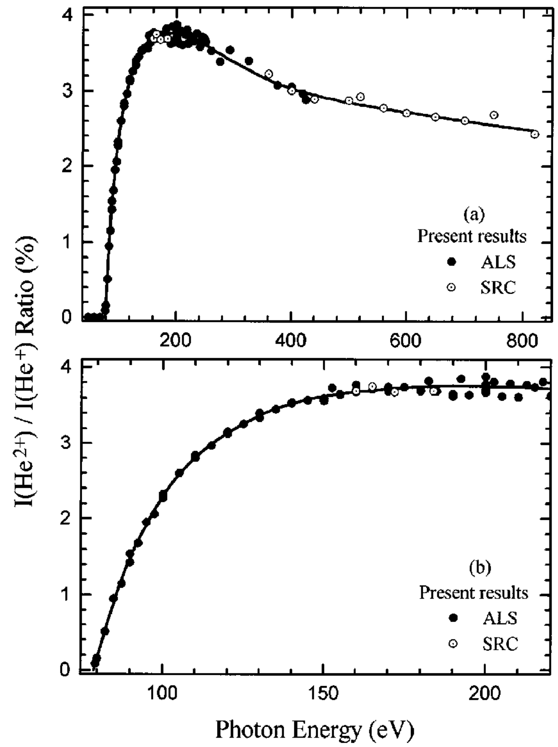

FIG. 1. The ratio of He^{2+} to He^+ ions measured as a function of the incident photon energy. The solid circle data points represent the results of four independent measurements at the ALS synchrotron source. The open circles represent data taken at the SRC facility. The solid line represents the best fit to our data.

Stray electrons are created by the pulsed photon beam from the storage ring ejecting photoelectrons from residual gas atoms, or by striking metal surfaces. For example, a metal filter placed in the photon beam to remove higher-order spectra can be a serious source of photoelectrons. Filters should be located inside the monochromator or at large distances from the TOF, where magnetic or electric fields can be used to prevent the photoelectrons from reaching the TOF.

(v) All mechanically ruled diffraction gratings will cause some scattering of the incident radiation. The intensity and spectral distribution of the scattered radiation is generally unknown, but if both quantities are relatively uniform within our spectral region we would expect this effect to decrease the ratio in our measurements. The reason for this is that light of energy between 25 and 80 eV is very effective in producing He^+, but cannot produce any He^{2+}. Gratings will also produce higher-order spectra. For the present measurements this effect will tend to increase the value of the ratio between 80 and 130 eV, but decrease the ratio for photon energies above 150 eV.

To minimize the effects of scattered light and higher-order spectra, the filters listed in Table I were used in all the measurements. Before every run the discriminator level was increased until an increase in the ratio was observed. The level was then set lower to accept a tolerable background signal. In all measurements the ions were accelerated through a potential difference of 4000–5000 V before striking the MCP detector. The gas pressure was generally varied to insure that there was no change in the ratio. But in all

TABLE II. The ratio R of double to single charged ions produced by photoionization of helium measured as a function of the incident photoenergy (smoothed data).

$h\nu$ (eV)	R (%)	$R\nu$ (eV)	R (%)	$h\nu$ (eV)	R (%)
79	0.0	125	3.26	380	3.08
80	0.16	130	3.37	400	3.03
81	0.30	135	3.45	420	2.99
82	0.45	140	3.52	440	2.95
83	0.60	145	3.58	460	2.91
84	0.74	150	3.62	480	2.88
85	0.88	160	3.68	500	2.85
86	1.02	170	3.72	520	2.82
87	1.15	180	3.74	540	2.78
88	1.27	190	3.76	560	2.77
89	1.38	200	3.75	580	2.74
90	1.48	210	3.74	600	2.72
92	1.67	220	3.72	620	2.70
94	1.84	230	3.68	640	2.67
96	2.00	240	3.65	660	2.65
98	2.15	250	3.61	680	2.63
100	2.28	260	3.56	700	2.60
102	2.42	270	3.51	720	2.58
104	2.53	280	3.47	740	2.56
106	2.63	290	3.42	760	2.53
108	2.73	300	3.38	780	2.51
110	2.81	320	3.30	800	2.49
115	3.00	340	3.22	820	2.47
120	3.14	360	3.15		

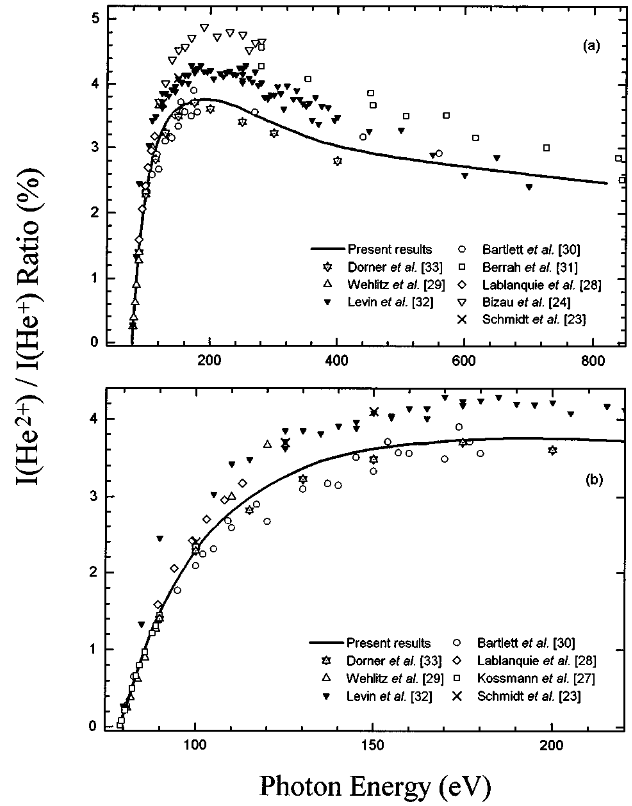

FIG. 2. The ratio of He^{2+} to He^{+} ions. Comparison of the present results (solid line) with the published experimental data. The absolute values of our earlier data (Bartlett *et al.*) are indicated by the open circles.

cases measurements were made with a background pressure less than 10^{-5} Torr.

III. RESULTS AND DISCUSSION

The present results for the $I(He^{2+})/I(He^{+})$ are shown in Fig. 1 by the solid circles and dot-centered circles. The solid line represents the best fit to our data. The three data points below 80 eV [Fig. 1(a)] were taken with an Al filter inserted into the light path. Without the filter an $I(He^{2+})/I(He^{+})$ ratio of 1.28% was obtained, indicating the presence of scattered light and/or higher-order spectra. Si and B filters were used up to 184 eV, but no filters were available between 185 and 330 eV. However, two sets of data were taken between 185 and 220 eV, one on an undulator beamline and the other on a bend magnet beamline. For a given setting of the undulator the radiation entering the monochromator is already highly concentrated in a narrow energy band centered on the chosen photon energy position, and with a half-width of approximately 4 eV. Hence there should be very little scattered radiation present. On the other hand, the bend magnet beamline allows the full spectral range of the synchrotron radiation to enter the monochromator, and could be expected to produce more scattered radiation than the undulator beamline. However, the spread in the data amounts to less than $\pm 3\%$ at 200 eV, and the median value blends smoothly into the data on each side of this region, which indicates that the effect of scattered light is minimal in this region. Use of the high-energy grating on the undulator beamline above 250 eV produced a large amount of higher-order spectra and was not used. Instead, the bend magnet beamlines at ALS and SRC were used to cover the range from 200 to 820 eV. Filters of Ti, Cr, and Ni were used to remove the higher-order spectra above 350 eV.

We believe that we have substantially eliminated all systematic errors, and that the absolute values of the $I(He^{2+})/I(He^{+})$ ratio represented by the solid curve are accurate to $\pm 2\%$ from threshold to 185 eV and about $\pm 4\%$ above 185 eV. The numerical values of the smoothed data are given in Table II. The smoothed data are compared to other experimental results in Fig. 2. No error bars are shown because of the unknown systematic errors. Most of the published data have statistical errors in the 1–4 % range. Our smoothed curve lies within the error bar limits of our earlier data, namely, $\pm 7\%$, and is in excellent agreement with most of the published experimental data from threshold to about 100 eV. However, above 100 eV our results are much lower than all previously published data, with the exception of the recent results of Dörner *et al.* [33]. In their studies they used the new technique of cold target recoil ion momentum spectroscopy. A major advantage of this technique is that ions of a given charge produced by higher-order spectra would have higher recoil energies than those produced by first-order radiation, thus allowing separation of the true from the unwanted signal. In addition, they used a MCP detector constructed with three channel plates. This insures a more equal detector response to ions of different charges. They also

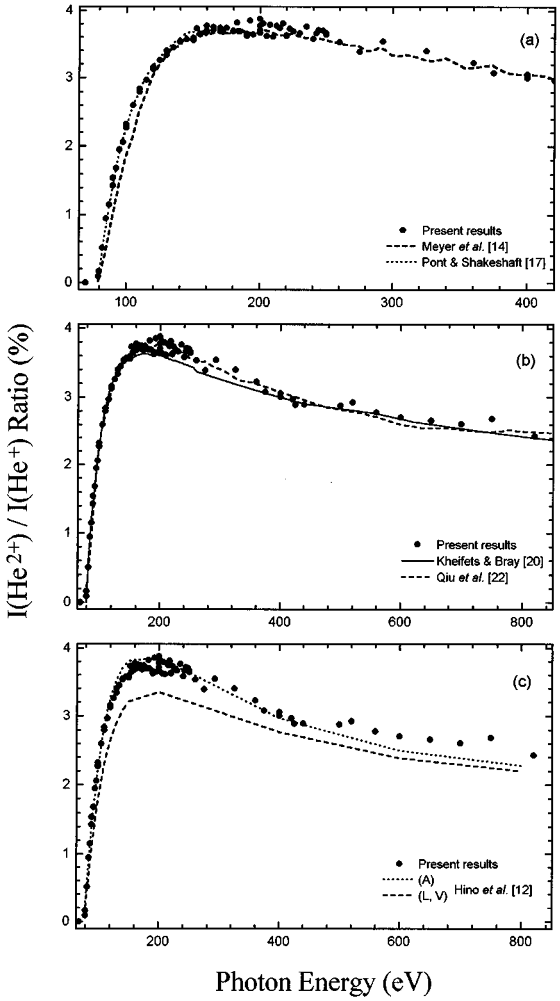

FIG. 3. The ratio of He^{2+} to He^{+} ions. Comparison of the present experimental data (solid circles) with the recent theoretical results.

monitored the pulse height distribution from their detector for both singly and doubly charged ions, and found the distributions to be identical. Our present data are in almost perfect agreement with their results up to 200 eV. Above that

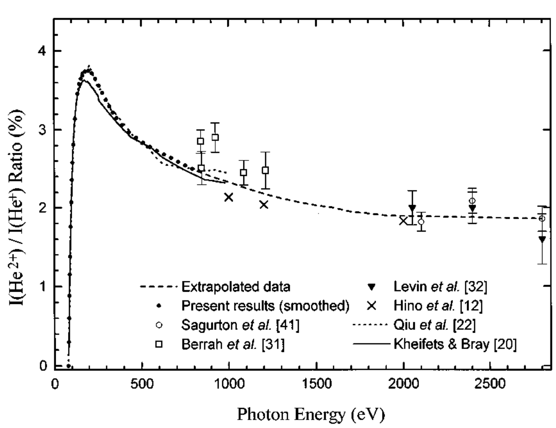

FIG. 4. Extrapolation of our present data (dashed line), to our previously reported data at 2800 eV illustrates the spectral regions that require further studies. The extrapolation is compatible with the trend toward higher energies in the calculated values of the ratio.

TABLE III. Absolute cross sections for double photoionization of helium. (Units in kb. 1 kb$=10^{-21}$ cm^2)

$h\nu$ (eV)	σ^{2+} (kb)	$h\nu$ (eV)	σ^{2+} (kb)	$h\nu$ (eV)	σ^{2+} (kb)
79	0.0	102	8.81	200	2.00
80	1.11	104	8.76	220	1.47
81	2.01	106	8.66	240	1.11
82	2.92	108	8.56	260	0.84
83	3.78	110	8.36	280	0.65
84	4.51	115	7.93	300	0.508
85	5.20	120	7.43	350	0.297
86	5.84	125	6.88	400	0.187
87	6.38	130	6.39	450	0.123
88	6.85	135	5.92	500	0.0864
89	7.23	140	5.44	550	0.0625
90	7.53	145	4.98	600	0.0465
92	8.02	150	4.58	650	0.0354
94	8.38	160	3.83	700	0.0269
96	8.59	170	3.21	750	0.0213
98	8.76	180	2.74	800	0.0169
100	8.76	190	2.33	820	0.0156

energy the two sets of data still fall within their respective error limits (Dörner *et al.* quoted an overall probable error of less than 6%). The recent recommended values of the double to single ionization ratio published by Bizau and Wuilleumier [24] are 20–37 % higher than the present data above 140 eV. We cannot reconcile this difference. However, we believe that the agreement of our data with that of

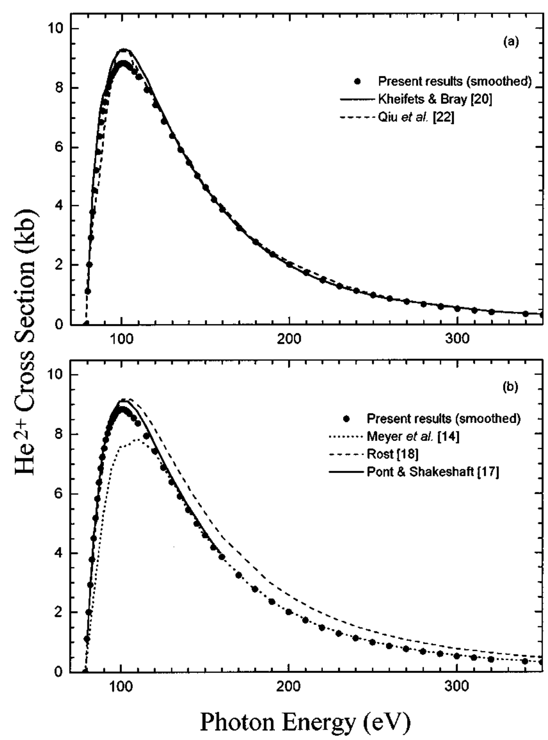

FIG. 5. The cross section for double photoionization of He as a function of the incident photon energy. Comparison of the present results (smoothed) (solid circles) with the recent theoretical results.

Dörner *et al.* (obtained by completely different techniques) is a measure of the accuracy of the two sets of results, and shows that they provide a new set of recommended values for the $I(He^{2+})/I(He^{+})$ ratio.

In Fig. 3 we compare our data for the $I(He^{2+})/I(He^{+})$ ratio with several of the most recent calculations [12,14,17,20,22]. With the exception of Ref. [12], the calculated results are all gauge independent, and are all in excellent agreement with the present results. There is also excellent agreement with the acceleration gauge calculations of Hino *et al.* [12] from threshold to about 400 eV. The recent calculations by Marchalant and Bartschat [21] from threshold to 200 eV (not shown) give separate results for the length, velocity, and acceleration gauges. The velocity form is in good agreement with the present results.

In Fig. 4 the dashed line shows an extrapolation of our present data to our previously reported high-energy data between 2100 and 2800 eV [41]. The only data available in this region are the experimental values of Refs. [31] and [32] and the theoretical values of Ref. [12]. Although the dashed line is compatible with the trend of the calculated ratios, obviously more experimental data are required in this spectral region to provide a more accurate comparison with theory.

To obtain the absolute value of the cross section for double photoionization, σ^{2+}, we use the relationship

$$\sigma^{2+} = B\sigma_{\mathrm{tot}} \quad (1)$$

where B is the branching ratio and equals $\sigma^{2+}/(\sigma^{+}+\sigma^{2+})$, and σ_{tot}, the total photoionization cross section, equals $(\sigma^{+}+\sigma^{2+})$. The measured ratio R of the doubly to singly charged ions produced is simply given by $R=\sigma^{2+}/\sigma^{+}$. Inserting the values of R, tabulated in Table II, and our previously published values for σ_{tot} [34] into Eq. (1) we obtain σ^{2+}. These results are tabulated in Table III and shown in Fig. 5. Comparison with theory shows almost perfect agreement with the latest quantum-mechanical calculations [14,17,20,22]. The deviations in the low-energy data of Meyer, Greene, and Esry [14], [Fig. 5(b)], are also reflected in their calculated values of the ratios [Fig. 3(a)]. This is a region where they expect their approximation of unequal energy sharing between the two electrons to break down. Otherwise, above 120 eV their data are in excellent agreement with the present results. The quantum-classical hybrid calculations by Rost [18] tended to overestimate the magnitude of the double photoionization cross section at energies above 100 eV. The calculated values of both the ratios and the double photoionization cross sections of Qiu *et al.* [22] and Kheifets and Bray [20] maintain very good agreement with the present data over the extended range from 80 to 820 eV.

ACKNOWLEDGMENTS

This work was supported by the National Science Foundation under Grant No. Phy-9317934 and is based in part upon research conducted at the Synchrotron Radiation Center, University of Wisconsin, Madison, which was supported by the NSF under Grant No. DMR-95-31009.

[1] T. A. Carlson, Phys. Rev. **156**, 142 (1967).
[2] F. W. Byron, Jr. and C. J. Joachain, Phys. Rev. **164**, 1 (1967).
[3] R. L. Brown, Phys. Rev. A **1**, 586 (1970).
[4] R. L. Brown and R. J. Gould, Phys. Rev. D **1**, 2252 (1970).
[5] M. S. Yurev, Opt. Spektrosk. **38**, 9 (1975) [Opt. Spectrosc. **38**, 4 (1975)].
[6] M. Ya. Amusia, E. G. Drukarev, E. G. Gorshkov, and M. P. Kazachkov, J. Phys. B **8**, 1248 (1975).
[7] S. M. Varnavshikh and L. N. Labzovskii, Opt. Spektrosk. **47**, 45 (1979) [Opt. Spectrosc. **47**, 24 (1979)].
[8] S. L. Carter and H. P. Kelly, Phys. Rev. A **24**, 170 (1981).
[9] S. N. Tiwary, J. Phys. B **15**, L323 (1982).
[10] T. Ishihara, K. Hino, and J. H. McGuire, Phys. Rev. A **44**, R6980 (1991).
[11] D. Proulx and R. Shakeshaft, Phys. Rev. A **48**, R875 (1993).
[12] K. Hino *et al.*, Phys. Rev. A **48**, 1271 (1993).
[13] K. W. Meyer and C. H. Greene, Phys. Rev. A **50**, R3573 (1994).
[14] K. W. Meyer, C. H. Greene, and B. D. Esry, Phys. Rev. Lett. **78**, 4902 (1997).
[15] C. Pan and H. P. Kelly, J. Phys. B **28**, 5001 (1995).
[16] J.-Z. Tang and I. Shimamura, Phys. Rev. A **52**, R3413 (1995).
[17] M. Pont and R. Shakeshaft, J. Phys. B **28**, L571 (1995); Phys. Rev. A **51**, 494 (1995).
[18] J.-M. Rost, Phys. Rev. A **53**, R640 (1996).
[19] A. S. Kheifets and I. Bray, Phys. Rev. A **54**, R995 (1996).
[20] A. S. Kheifets and I. Bray, Phys. Rev. A (to be published).
[21] P. J. Marchalant and K. Bartschat (private communication).
[22] Y. Qiu, J-Z. Tang, J. Burgdörfer, and J. Wang, Phys. Rev. A **57**, R1489 (1998).
[23] V. Schmidt *et al.*, Phys. Rev. A **13**, 1748 (1976).
[24] J. M. Bizau and F. J. Wuilleumier, J. Electron Spectrosc. Relat. Phenom. **71**, 205 (1995).
[25] G. R. Wight and M. J. Van der Wiel, J. Phys. B **9**, 1319 (1976).
[26] D. M. P. Holland, K. Codling, J. B. West, and G. V. Marr, J. Phys. B **12**, 1465 (1979).
[27] H. Kossmann, V. Schmidt, and T. Andersen, Phys. Rev. Lett. **60**, 1266 (1988).
[28] P. Lablanquie, K. Ito, P. Morin, I. Nenner, and J. H. D. Eland, J. Phys. D **16**, 77 (1990).
[29] R. Wehlitz *et al.*, Phys. Rev. Lett. **67**, 3764 (1991).
[30] R. J. Bartlett, P. J. Walsh, Z.-X. He, Y. Chung, E.-M. Lee, and J. A. R. Samson, Phys. Rev. A **46**, 5574 (1992).
[31] N. Berrah *et al.*, Phys. Rev. A **48**, R1733 (1993).
[32] J. C. Levin, G. B. Armen, and I. A. Sellin, Phys. Rev. Lett. **76**, 1220 (1996).
[33] R. Dörner *et al.*, Phys. Rev. Lett. **76**, 2654 (1996).
[34] J. A. R. Samson, Z.-X. He, Y. Lin, and G. N. Haddad, J. Phys. B **27**, 887 (1994).
[35] W. C. Wiley and I. H. McLaren, Rev. Sci. Instrum. **26**, 1150 (1955).
[36] V. Schmidt, N. Sandner, and H. Kuntzemüller, Phys. Rev. A **13**, 1743 (1976).
[37] M. R. Bruce and R. A. Bonham, Z. Phys. D **24**, 149 (1992).
[38] C. F. Barnett, in *Atomic Data for Fusion*, edited by H. T.

Hunter and M. I. Kirkpatrick (National Technical Information Service, U.S. Dept. Of Commerce, Springfield, VA, 1990), Vol. 1.

[39] Ce Ma, C. R. Sporleder, and R. A. Bonham, Rev. Sci. Instrum. **62**, 909 (1991).

[40] P. Nagy, A. Skutlartz, and V. Schmidt, J. Phys. B **13**, 1249 (1980).

[41] M. Sagurton, R. J. Bartlett, J. A. R. Samson, Z.-X. He, and D. Morgan, Phys. Rev. A **52**, 2829 (1995).